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***Green and Circular Economy*
ECOMONDO 2018**



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CONTENTS

ECOMONDO 2018

Editorial

- Green and Circular Economy ECOMONDO 2018
22th International Trade Fair of Material & Energy Recovery and Sustainable
Development
Fabio Fava 2105

Special issue papers

- A case study of indoor air quality in a classroom by comparing passive
and continuous monitoring
*Patrizia Aversa, Gaetano Settimo, Marco Gorgoglione, Emanuela Bucci,
Giorgio Padula, Alessandra De Marco*..... 2107
- Earthworms for feed production from vegetable waste: environmental
impact assessment
Cecilia Conti, Jacopo Bacenetti, Doriana Tedesco..... 2117
- Design and environmental assessment of bioplastics from
Hermetia illucens prepupae proteins
Silvia Barbi, Rosangela Spinelli, Anna Maria Ferrari, Monia Montorsi..... 2123
- Impact of effluents from wastewater treatments reused for irrigation:
strawberry as case study
*Luca Rivoira, Michele Castiglioni, Ahmed Kettab, Naaila Ouazzani,
Emad Al-Karablieh, Nesrine Boujelben, Donatella Fibbi, Ester Coppini,
Edgardo Giordani, Massimo Del Bubba, Maria Concetta Bruzzoniti*..... 2133
- Investigating circular economy urban practices in Centocelle, Rome District
*Francesca Cappellaro, Laura Cutaia, Carolina Innella,
Claudia Meloni, Rocco Pentassuglia, Vincenzo Porretto*..... 2145

Optimizing bioremediation of hydrocarbon polluted soil by Life Cycle Assessment (LCA) approach <i>Giuliana D'Imporzano, Ilaria Re, Federica Spina, Cristina Varese, Edoardo Puglisi, Giulia Spini, Carla Gramellini, Giacomo Zaccanti, Fabrizio Beltrametti Adriana Bava, Fabrizio Adani</i>	2155
Biomass exploitation for energy supply and quality compost production. An exemplary case of circular economy in the North East of Italy <i>Veronica Novelli, Paola Geatti, Luciano Ceccon, Sara Gratton</i>	2163
Secondary treated wastewater as a support strategy for tree crops irrigation: nutritional and physiological responses on apple trees <i>Giulio Demetrio Perulli, Giovambattista Sorrenti, Kushtrim Bresilla, Luigi Manfrini, Alexandra Boini, Maurizio Quartieri, Moreno Toselli, Luca Corelli Grappadelli, Brunella Morandi</i>	2171
The role of waste collection centers in a circular economy scenario: An empirical study on the citizens' perception <i>Marco Savastano, Mattia Belcastro, Francesco Dentale</i>	2181
Sustainability assessment of two digestate treatments: a comparative Life Cycle Assessment <i>Sofia Spagnolo, Anna Tinello, Cristina Cavinato, Alex Zabeo, Elena Semenzin</i>	2193
Technologies for the control of emerging contaminants in drinking water treatment plants <i>Sabrina Sorlini, Maria Cristina Collivignarelli, Marco Carnevale Miino</i>	2203
Sustainable management of sedimentary resources: A case study of the EGADI project <i>Sergio Cappucci, Mentore Vaccari, Marco Falconi, Terry Tudor</i>	2217
Real time monitoring of water quality in an agricultural area with salinity problems <i>Sara Simona Cipolla, Marco Maglionico, Marinella Masina, Alberto Lamberti, Irene Daprà</i>	2229
Start-up of the door-to-door municipal solid waste separate collection service in a large metropolitan area <i>Sabino De Gisi, Francesco Todaro, Vincenzo Campanaro, Michele Notarnicola</i>	2241
Nitrogen and <i>Escherichia coli</i> removal in facultative finishing lagoons receiving treated urban wastewater <i>Carmine Fiorentino, Maurizio Mancini</i>	2253
Membrane systems and water structures <i>Jan Koppe, Marietta Hesse</i>	2265

Smart water and soil-salinity management in agro-wetlands <i>Marinella Masina, Roberta Calone, Lorenzo Barbanti, Claudio Mazzotti, Alberto Lamberti, Maria Speranza</i>	2273
Combined assessment of chemical and ecotoxicological data for the management of contaminated marine sediments <i>Francesco Todaro, Sabino De Gisi, Claudia Labianca, Michele Notarnicola</i>	2287
Circular economy and upcycling of waste and pre-consumer scraps in construction sector. The role of information to facilitate the exchange of resources through a virtual marketplace <i>Marco Migliore</i>	2297



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EDITORIAL

Green and Circular Economy ECOMONDO 2018

22th International Trade Fair of Material & Energy Recovery and Sustainable Development

The papers collected in this Special Issue of *Environmental Engineering and Management Journal* were presented as lectures or posters at the scientific and technical conferences hosted by *Ecomondo 2018* held in Rimini, Italy, 6–9 November, 2018 (<http://en.ecomondo.com>).

Ecomondo is the one of largest European exhibitions in the field of *Green and Circular Economy*, with over than 100,000 delegates from 60 different nations and 1200 companies exhibiting their products and processes in 113,000 square meters. It is hosting over than 100 conferences and workshops, ensuring a weighted and rewarding balance between sales dimension and technical-scientific dimension, with extensive room dedicated to research and innovation, education and training and international networking.

As with the previous editions, the aim of *Ecomondo 2018* was to explore recent industrial advances and opportunities in industrial technical waste production reduction, recycling and exploitation; sustainable food and wood chains, biowaste collection and exploitation via integrated biorefinery schemes, with the production of biobased chemicals, materials and biofuels, including methane; industrial eco-design; industrial symbiosis, renewable and critical resources; water resources monitoring, protection and sustainable use in the civil and agrifood sectors; wastewater treatment and valorization with nutrients recovery and water reuse; marine resources protection and sustainable exploitation; sustainable remediation of contaminated sites, ports and marine ecosystems; indoor and outdoor air monitoring and clean up; and circular and smart Cities.

Some of the international workshops were focused on the advanced international trends in the in the circular economy domains and on the role of digitalization and industry 4.0 enabling technologies in the process efficiency, eco-design and waste collection in the major industrial value chains. Some other workshops were focused on the technical and regulatory constrains currently affecting the creation of circular economy value chains in the sectors of electronic and electric products, automotive, construction and demolition, packaging materials and textile and fashion. A special room has been dedicated to the recycling of plastic waste, biodegradable plastics and the monitoring, prevention and mitigation of marine litter. Finally, *Ecomondo 2018* dedicated a particular attention to the main challenges and needs of the Mediterranean macro-region, addressing, via dedicated workshops, the priorities of the area associated with the water scarcity, the Mediterranean Sea contamination (also due to marine litter) and the blue and sustainable growth of the area.

Ecomondo 2018 hosted about 140 conferences, more than 900 oral communications and almost 100 full papers. This special issue contains some of such papers and provides some of the key information presented and discussed in the frame of some of the most relevant technical and scientific conferences of 2018 edition of *Ecomondo*.

We believe that this collection of papers will be useful to people who could not visited and participate in *Ecomondo 2018*. It is primarily towards them but it also aspires to provide permanent records in the promotion,

adoption and implementation of the major priorities and opportunities of the circular economy in Europe and in the Mediterranean basin, with the conversion of some of the key local environmental challenges into new opportunities for a green and sustainable growth of the areas mentioned.

For additional info, please refer to the following links:

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Fabio Fava, PhD, Professor, *Alma Mater Studiorum - Università di Bologna*, Bologna, Italy



Fabio Fava, born in 1963, is Full Professor of “Industrial & Environmental Biotechnology” at the School of Engineering of University of Bologna since 2005.

F. Fava published about 240 scientific papers, 170 of which on medium/high IF peer-review international journals of industrial and environmental biotechnology. He has 5733 overall citations, a H-index of 46 and an i10 index of 123 (Google Scholar) along with 180 papers quoted by Scopus. He is actively working in the fields of environmental, industrial and marine biotechnology and of the Circular Bioeconomy in the frame of a number of national projects and collaborative projects funded by the European Commission. Among the latter, he coordinated the FP7 collaborative projects NAMASTE, on the integrated exploitation of citrus and cereal processing byproducts with the production of food ingredients and new food products, and BIOCLEAN, aiming at the development of biotechnological processes and strategies for the biodegradation and the tailored depolymerization of wastes from the major oil-deriving plastics, both in terrestrial and marine habitats. He also coordinated the Unit of the University of Bologna who participated in the FP7 collaborative projects ECOBIOCAP and ROUTES (on the production of microbial and biodegradable polymers from different organic waste and food processing effluents), MINOTAURUS and WATER4CROPS (on the intensified bioremediation of contaminated waste- and ground- water and the integrated valorization and decontamination of wastewater

coming from the food processing industry and biorefineries), and ULIXES and KILL SPILL (on the development of strategies for intensifying the *ex situ* and *in situ* bioremediation of marine sediments contaminated by (chlorinated)hydrocarbons and microplastics and the isolation and industrial exploitation of microbes from such contaminated matrices). F. Fava served and is serving several national, European and international panels, by covering the following positions:

- Italian Representative in the Horizon2020 Programme Committee of Societal Challenge 2: European Bioeconomy Challenges: Food Security, Sustainable Agriculture and Forestry, Marine, Maritime and inland water research" (European Commission, DG RTD) (2013-);
- Italian Representative in the "States Representatives Group" (SRG) of the Public Private Partnership "Biobased Industry" (PPP BBI JU) (Brussels) (2014-); he is chairing the SRG since October 2018;
- Italian Representative in the BLUEMED WG of the EURO-MED Group of Senior Officials (EU Commission DG RTD and Union for Mediterranean) (2017-);
- Italian Representative in the initiative on sustainable development of the blue economy in the western Mediterranean the "Western Mediterranean Initiative" WEST MED, promoted by the EU Commission (DG MARE) in close cooperation with 10 countries of the area (2016-);
- Member of the "Working Party on Biotechnology, Nanotechnology and Converging Technologies" of the Organization for Economic Co-operation and Development (OECD, Paris) (2008-);
- Chair (2011-2013) and currently Deputy Chair of the "Environmental Biotechnology section" of European Federation of Biotechnology (EFB) (2013-).

Finally, he is the scientific coordinator of the International Exhibition on Green and Circular economy ECOMONDO held yearly in Rimini (Italy)



“Gheorghe Asachi” Technical University of Iasi, Romania



A CASE STUDY OF INDOOR AIR QUALITY IN A CLASSROOM BY COMPARING PASSIVE AND CONTINUOUS MONITORING

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Abstract

Most people is aware that outdoor air pollution can impact human health, including indoor air pollution. Human exposure to air pollutants indicate that indoor levels of pollutants may be two to five times, and occasionally more than 100 times, higher than outdoor levels. These levels of indoor air pollutants are of particular concern because most people spend about 90 percent of their time indoors. In this manuscript we monitored indoor pollutants in a school building with two different methodologies (active and passive monitoring). We demonstrated that, even if in general the registered pollutants showed concentration below the threshold defined by WHO guidelines, the active monitoring is able to catch peaks of concentrations linked to particular school activities, such as educational arts, including single emitting episodes. The use of the monitoring equipment in continuous facilitated the identification of the pollution sources in a timely manner, identifying the impact of the best management practices on the microclimate, and particularly on internal temperature and CO₂ concentrations.

Key words: active monitoring, indoor pollution, school redevelopment

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1. Introduction

Because of increasing traffic and industrial emissions, outdoor air quality has become of growing concern during the past 50 years. As a consequence, monitoring of outdoor air pollution has become more and more systematic in western countries, either to check that national or international standards on contaminant emissions are applied, to ensure that outdoor air quality complies with the standards in force, to inform authorities if concentrations limit value are exceeded, and/or to assess people health hazard (Blondeau et al., 2005).

Evidence has been made that city-dwellers spend most of their time in buildings (Jenkins et al., 1992; Klepeis et al., 2001) and are far more exposed to pollution indoors than outdoors. As a result,

contaminant concentration measurements have been performed in various indoor environments during the past 40 years, providing more accurate information about human exposure.

In the last years public awareness about negative impacts of indoor air quality on human health was strongly increased, starting after occurrence of health problems associated with indoor air quality in building in the 1970s for occupants of commercial and institutional buildings (ASHARAE, 1999; Daisey et al., 2003; Finnigan et al., 1984; Latif et al., 2018; Mendell et al., 2007).

Poor indoor air quality at classrooms was demonstrated to exert a negative impact on children's learning performance, with absenteeism and adverse health effects such as increased risk of asthma and other health-related symptoms (Canha et al., 2013;

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EPA, 2014; Vassura et al., 2015). In schools, aerosol particles are between the pollutants that can cause the decrease of indoor air quality. Particles may be originated naturally such as by dust, salt, pollen, viruses, fungi and bacteria (Sohn et al., 2009) or from anthropogenic sources, such as industrial activity, and combustion processes (Almeida et al., 2011; Almeida-Silva et al., 2011; Canha et al., 2010, 2013; David and Kopac, 2017; Lage et al., 2014).

The WHO (2006) started a process of updating the guidelines for the 42 pollutants harmful for indoor environments and for the first time considered as integrated both the indoor and outdoor environments. According to WHO (2010), indoor pollution in schools can be traced for the presence of organic compounds such as VOCs (i.e. benzene, naphthalene, formaldehyde, toluene, xylene, styrene, trichlorethylene, tetrachlorethylene) and inorganic substances, such as PM₁₀ and PM_{2,5} particles, carbon dioxide (CO₂), oxides of nitrogen (NO_x), carbon oxide (CO), sulfur dioxide (SO₂) and ozone (O₃). Legislative acts are developed in several European countries, in particular in France, that responded to the obligation to monitor indoor air quality in nursery schools for 32 pollutants as stated Decree 5 June 2016, relating to monitoring procedures for indoor air quality in certain establishments receiving public.

Indoor atmospheric pollution is due not only to the behavior of occupants but to a series of variables such as the diversity of pollutant concentration levels in relation to the investigated environment and depends on different factors ranging from climatic parameters and typologies of natural or forced ventilation carried out, and from the characteristics of the environment, furnishings, building materials and surface finishes (Blondeau et al., 2005). Considering all the listed aspects, indoor monitoring is complex and unique, depending on the environment.

In the countries of the European Community already sensitive to the problem in recent years various strategies have been adopted to mitigate and contain the phenomenon (Godwin and Batterman, 2007; Shendell et al., 2004; Sohn et al., 2009; Stranger et al., 2007, 2008). Recent studies show that children aged 3 to 14 are vulnerable categories as they spend 90% of their day in indoor environments, both in winter and summer and that they reveal a greater susceptibility to some typical indoor pollutants (Firestone et al., 2008; Selgrade et al., 2008).

The Italian National statistical database (www.istat.it) shows that the school population (students of all orders and degrees, teaching staff excluding administrative staff) spending many hours in school environments is corresponding to around 9 million people safety and well-being in schools. The harmonization framework for health started in Europe, started in the year 2015, based on evaluation of indoor emissions from construction products for the obligation to certify indoor air quality. Recently, to comply, Italy has adopted in the year 2014 the Action Plan for environmental sustainability of consumption in the Public Administration sector, and will end in

2018 with a series of legislative acts and actions aimed at protecting the health of the population in line with what was defined in the First Accord of 27/09/2001 between the Minister of Health regions and autonomous provinces, where the Guidelines for Indoor Environments were established (National Prevention Plan).

It should be emphasized the growing interest in recent years in the monitoring of PM₁₀ and breathable PM_{2,5}, as several studies have shown that exposure to high concentrations of these pollutants are no longer due solely to the outdoor contribution. Indeed, the PM pollution is not attributable just to traffic, presence of industrial plants and/or residential thermal installations, but also to poor ventilation system inside the buildings through doors and windows, that are usually closed and to crowding itself in the indoor environments.

The aim of this paper is to propose a first set of methodological strategies for simultaneous monitoring of indoor air quality for chemical pollutants such as CO, NO₂, CO₂, Formaldehyde, Toluene, Benzene, Btex, acetaldehyde, m,o,p, xylene, acetaldehyde, acetone, styrene, chloroform, ethylbenzene, trichlorethylene, dichloromethane, tetrachlorethylene using in-situ, continuous and passive type methodology. In the same time to assess if the proposed methodology is able to identify management measures able to reduce indoor air pollutants concentrations.

2. Material and methods

2.1. The classroom

The classroom chosen to carry out monitoring is part of a school building that has been subjected to an energy-saving reconditioning constituted by an outer coat with natural insulating material. The choice was related to some feature such as the street exposure, the solar exposure, the building material characteristics and the type of furnishings.

The selected school was located in a village in South of Italy (Mesagne), characterized by Mediterranean climate, even if is located not on the coast but 20 km from the sea. The school building was redeveloped with a vertical and horizontal intervention called "external coat insulation", change of windows fixtures and an integrate hot water heater. The building is located far from industrial sites, out from the traffic area. During the experiment, the school community participated in a classroom training on issues related to energy efficiency and the role of indoor pollution, at the stage of detection and experimentation.

2.2. Monitoring activity

The duration of monitoring was 6 months, and was organized in the following way: measurements in continuous done by Aeroqual₁, Aeroqual₂ and Capetti, and passive sampler measurements by Radiello. For

indoor measurements Aeroqual₁, Capetti and passive samplers were located into the center of the classroom at 90 cm height, while Aeroqual₂ was located close to the desk, again at 90 cm height. The Capetti measured indoor temperature, relative humidity and CO₂ concentration and passive samplers indoor (Radiello) measured COV and formaldehyde. The Aeroqual₁ and Aeroqual₂ measured COV, temperature relative humidity and CO₂ concentration indoor.

For outdoor measurements the Capetti (to monitor temperature, relative humidity, wind speed and direction) were located on the school roof at 1.5 m height, and the passive samplers (to measure COV and formaldehyde) close to the classroom windows. For more details see Fig. 1 and Table 1. The measurements duration was variable between 4 to 6 days depending on the school vacation time.

All the measurements obtained by the campaign were compared with data provided by ARPA Puglia (www.arpa.puglia.it) for pollutants and (www.eurometeo.com) for meteorological data. The both analytical systems and passive samplers were

conditioned for 48h to adapt to the measurements environment, according to methodology described in ISSN, 2015. Testing averaged over 4 days was conducted between February 2017 and April 2017, considering the periods of the winter season and the spring season. The sampling cycles provided 2 reference tests (Blank), characterized by the absence of school activities corresponding to carnival holidays and Easter holidays. Other 2 tests were conducted during school day-to-day. For VOCs and Formaldehyde measurement, Radiello passive samplers and continuous analyzers were used for a week, during working days (Monday to Friday). The tests have taken into account several variables: external climate factors, green for shading, location and exposure of the classroom, classroom activities, number of attendances, adopted ventilation, school holidays, use or no of thermal conditioning in relation to different seasons, daily habits related to classroom cleaning, and finally vehicular traffic. The information on cleaning methods and the products used were obtained from interviews with the cleaning staff.

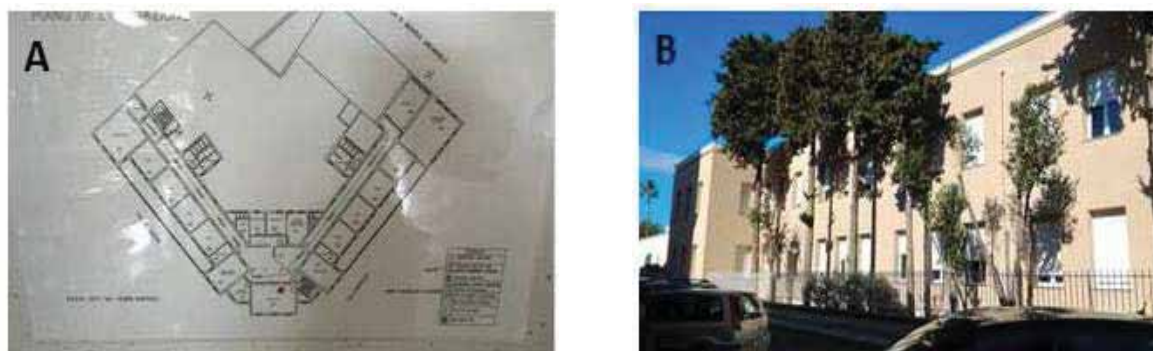


Fig. 1. Plan of the school (A), exterior view of the building (B)

Table 1. Monitoring system

<i>Aeroqual sensors 1</i>	<i>Technical data range</i>	<i>Technical data Sensitivity limit</i>	<i>Technical data Resolution</i>	<i>Technical data Accuracy</i>
Electrochemical sensor for Carbon Monoxide CO	0-25 ppm	0.05 ppm	0.01ppm	≤±/0.5 ppm
Electrochemical sensor for Formaldehyde CH ₂ O	0-10 ppm	0.01 ppm	0.01ppm	≤±/0.05 ppm
Semiconductor sensor for Ozono O ₃	0-0.5 ppm	0.001ppm	0.001 ppm	≤±/0.08 ppm
Electrochemical sensor for nitrogen dioxide NO ₂	0-1 ppm	0.005 ppm	0.001 ppm	≤±/0.02 ppm
NDIR non-dispersive infrared sensor for CO ₂	0-2000 ppm	10 ppm	1 ppm	≤±/10% ppm +5%
PID sensor for VOC	0-20 ppm	0.002 ppm	0.01 ppm	≤±/10%
Built-in temp / RH sensor	10-125°C			
Aeroqual sensors 2				
GSE electrochemical sensor for CO	0-100 ppm	0.2 ppm	0.1 ppm	≤±/10%
Built-in temp / RH sensor	10-125°C and 0-100%			
NDIR non-dispersive infrared sensor for CO ₂	0-2000 ppm	10 ppm	1 ppm	≤±/10% ppm+5%
PID photoionization sensor for VOC	0-20 ppm	0.02 ppm	0.01 ppm	≤±/10%
Capetti sensors				
System for the measurement of Temperature	-10-60°C		0.01°C	+/-0.1°C
Relative humidity	10-90%		1 ppm	+/-5% a 25°C
Concentration of ambient CO ₂	0-2000 ppm			+/-50ppm+2%
System for the measurement of Temperature	-40-80°C		0.2°C	
System for the measurement of wind	0.5-70 m/s		0.1 m/s	5%
wind direction	0°-360°		≤1°	3°
global solar radiation	0-2000 W/m ²		3 μm	
Radiello for HPLC e GCMS Method	www.radiello.it UNI EN ISO 16000-annex			

3. Results and discussion

3.1. Volatile Organic Compounds (VOCs) and Formaldehyde

The study with the continuous monitoring system for VOCs was necessary to detect the activation of the sources during the period of interest and the contribution from external sources. The commercially used Aeroqual₁ system was calibrated respect to 100 ppm of isobutylene. therefore, the concentration of total VOC is referred to this gas. Unfortunately, the type of sensor used doesn't give sufficient and unequivocal elements to define the amount of formaldehyde, most likely due to interfering as reported in the document of the technical characteristics of the electrochemical sensor interferences for reducing gases such as alcohols.

Indeed, the results, carried out from 7 am to 1 pm school activity with continuous analysis systems, show a recurring peak for formaldehyde and VOCs coming (Singer et al., 2006) from the process of classroom cleaning (Fig. 2).

The detergents, mostly degreasers, are composed by, alcohol, limonene, sodium hypochlorite, methylchlorothiazolinone, methylisothiazolinone, thus the amplitude peak became more evident when the cleaning procedure was carried out in form of aerosol. An important peak, was recorded during school activities in one specific day with an increase in VOCs concentration ranging around 240 µg/m³, determined by the presence of a leather bag and in proximity of the chair, in presence of a fixative jar left open, used for art activities (Fig. 2).

Fig. 3 shows the different information obtained by passive and active measurements for both VOC (a) and CH₂O (b). In the comparison periods, the VOCs selected have values lower than the main WHO references (WHO, 2006) defined by different European countries.

High importance is attributed to the location of the sensors inside the classroom, indeed the two different set of measures (continuous, located close to the desk and passive, located in the middle of the classroom) explains the different levels of pollutants detected. In addition, the active system was in the vicinity of the clean desk with the detergents.

In general, the concentrations observed do not present particular criticisms as compared to the limit values set by the European Community Generally, but is to be noticed that higher level of both pollutants were observed with active sampling methodology as compared to passive one in all the monitoring periods. Furthermore, passive methodology was not able to catch the peak of VOC registered by the active measurements. Fig. 4 shows a comparison between indoor and outdoor measurements of respectively formaldehyde, toluene and acetaldehyde in the main square and acetone and m,o,p-xylene in the small box in four weeks.

Very low differences, statistically insignificant, are present between indoor and outdoor measurements in all the sampling weeks. Generally, acetone and xylene show very lower values in comparison to the main pollutants. The concentration of some substances, such as styrene, chloroform and others are below the limit of detection of the instrument, as detected in Fig. 5.

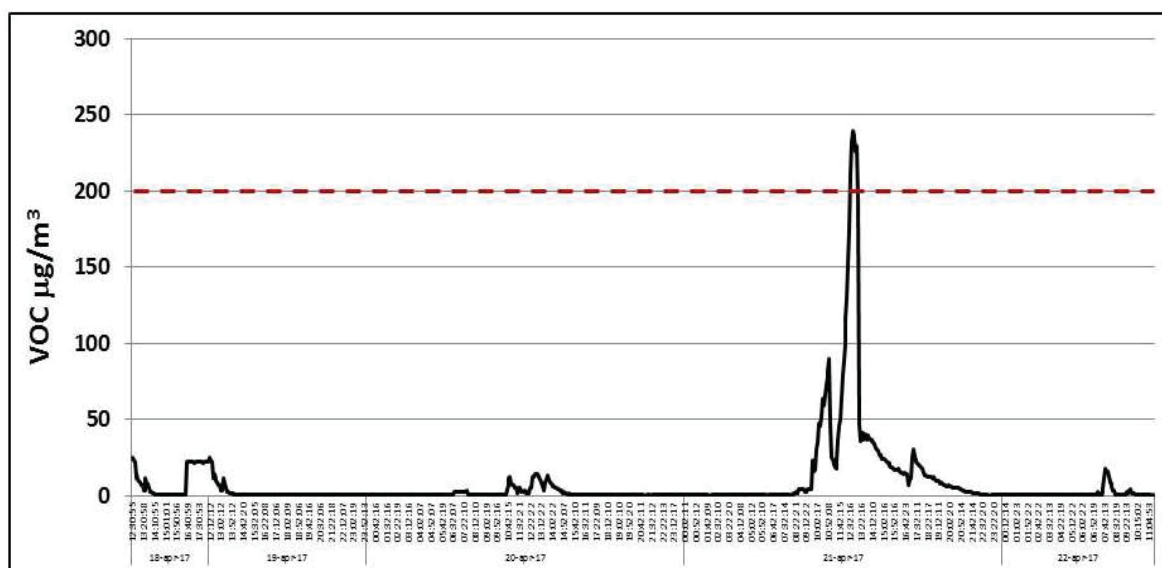


Fig. 2. Indoor total VOC measurements (April 18-22)

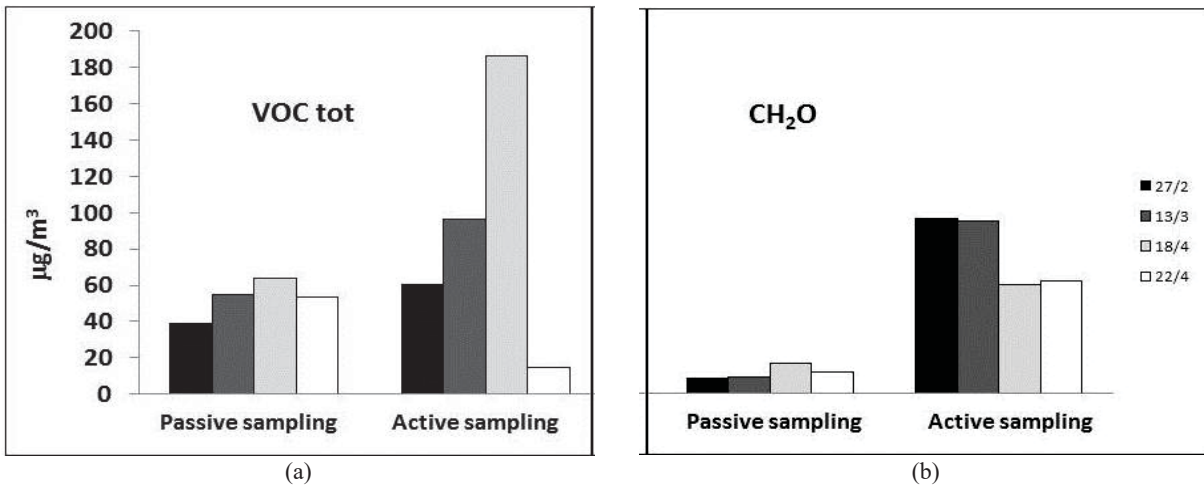


Fig. 3. Comparison of total VOC (a) and CH₂O (b) measurements obtained by passive sampling and active sampling methodology in four different days

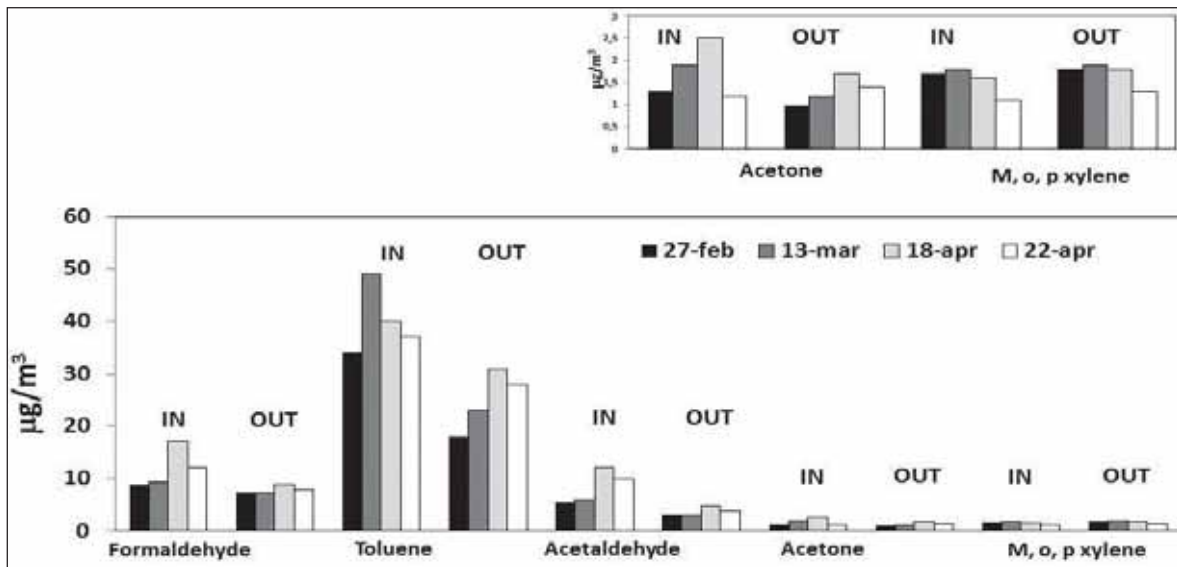


Fig. 4 Comparison of indoor (IN) and outdoor (OUT) pollutants obtained by passive sampling in four different weeks

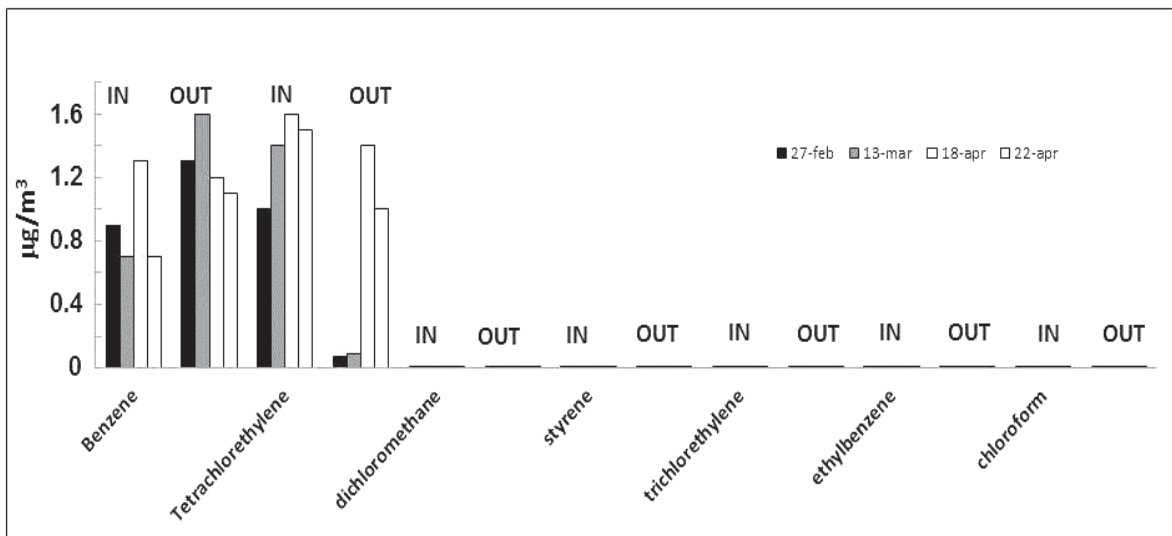


Fig. 5. Comparison of indoor (IN) and outdoor (OUT) pollutants obtained by passive sampling in four different weeks

3.2. Temperature and CO₂

The temperature increases according to the number of attendances in the classroom and decreases in pauses with the absence of students, end-of-activity or in relation to the type of ventilation (Daisey et al., 2003; Satish et al., 2012; Shendell et al., 2005) used. In Fig. 6 we showed the temperature trends into the classroom in a normal day, from measurements in the middle of the classroom. The maximum indoor temperature doesn't exceed the value of 23°C, even if the temperatures monitored in the proximity of the chair (data not shown) were higher than 26°C, thus well above the limits suggested by the 2013 dlgs74. In the spring period, during hot and sunny days, the same temperature registered is 23°C at the end of the morning or in the warmer hours, and this highlights the weight of the contribution of the thermal plants. The temperature profiles show that in the presence of

activities within the class the temperature increases faster and more consistently when all the fixture is closed (Fig. 6). When the windows are opened in Vasistas mode or totally opened, the temperature curve decreases proportionally. The same happens if the inner door is opened in the corridor (Fig. 6). During the interruption of school activities (Carnival and Easter holidays), the temperature profile measured within the class follows the increase observed in the outdoor temperature.

The CO₂ measurements in the classroom recorded values even higher than 2000 ppm, during the start of the activity, at the time of return from the gym, especially in winter more than in spring, where it is customary to keep the windows open. Moreover, an analysis of the period from 18 to 22 April showed how a significant increase in CO₂ was due to the strong winds that avoid the opening of the windows in the classroom, as shown in Fig. 6.

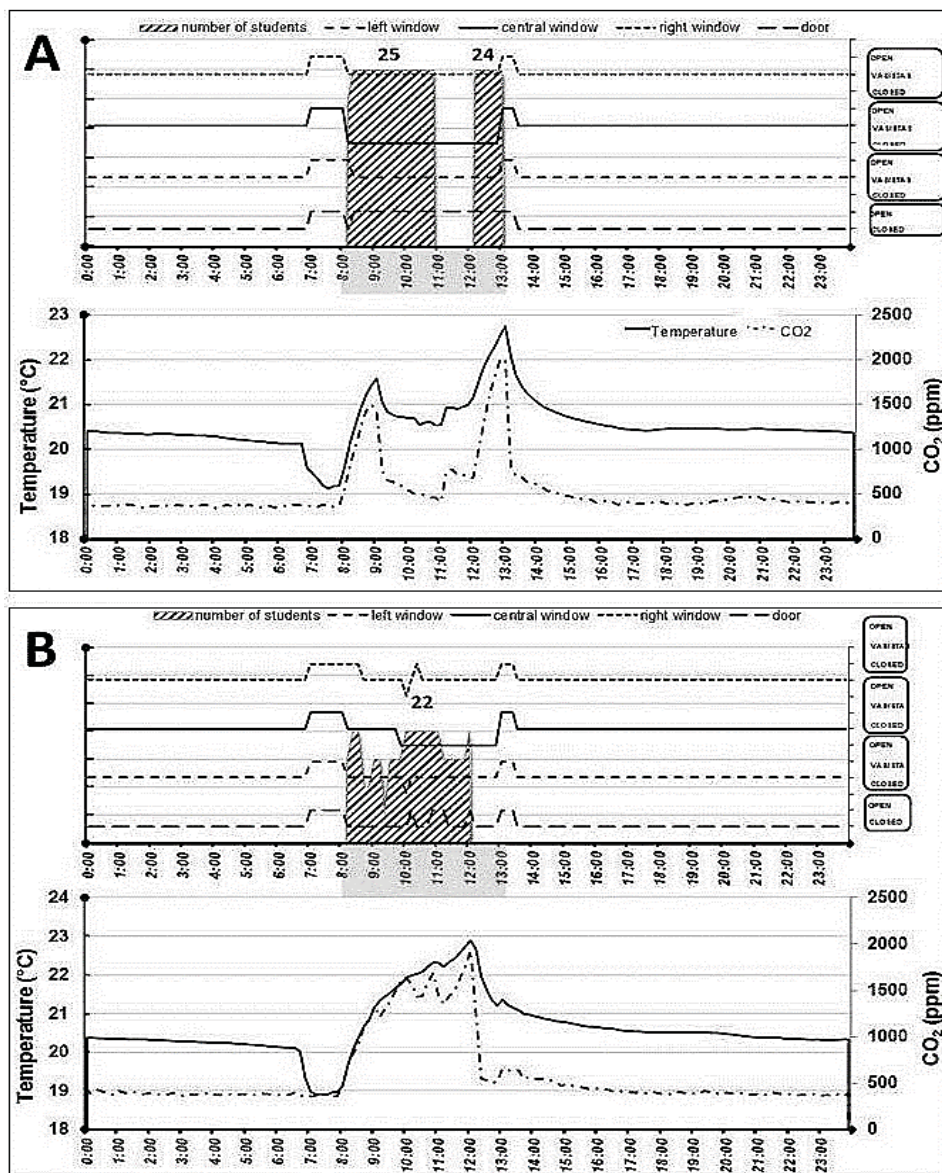


Fig. 6. Temperature and CO₂ measurements during two different days, 15th March (A) and 16th March (B) as related to number of students into the classroom and opening and closure of the three different windows and the door

3.3. Relative humidity

The relative humidity trend follows the same pattern as temperature, increases in classroom activity when windows and door are locked (data not shown). Moisture decreases with the opening of the windows while the opening of the door generally produces a dampening of the rapidity of the moisture increase. From data analysis, however, recorded values follow the external trend and do not generally exceed 55%, in line with dlgs n ° 74 of 16/4/2013, on days not rainy or humid as those characterized by winds typical of the Mediterranean climate.

3.4. CO, NO₂, O₃

There were no significant increases in the level of CO, but just a sufficient recurrence at the start and end of school time as shown in Fig. 7, probably due to vehicular traffic and to the discharges from thermal plants. The values generally not exceeding 2 ppm still remain below the expected limits or guide values. Immediately before the start of school activities there is an increase in the concentration of 0.02 ppm of NO₂ compared to a baseline level.

During the activity there is usually a reduction in concentration and the opening of doors or windows always appears to have the same effect. The oscillations found are still included within the limits and the guide values taken into the reference. It is assumed, as shown in Fig 7 that a short variation can be attributed to the discharges of vehicular traffic in the presence of sunny days. The pollutant concentrations into the environment don't exceed the current WHO guideline values to protect public health (WHO, 2006). The monitoring carried out did not reveal any particular criticisms in the measurement of ozone, whom concentrations appear, however, affected by the aperture of the windows. However, the

ozone levels remain within the limits of the experimental tolerance, as shown in Fig 7. From temporal trends shown in Fig 7 it appears clear that the school area is not affected by significant traffic levels.

The results shown above demonstrated that the communication and the interactive training can be considered as an effective scientific tool to be included in the experimental campaigns and in the protocols to prevent and mitigate the emissions of indoor pollutants in the daily living environment.

Indeed, during the experiment together with the experimental measures, the students were involved in the achievement of results, by compilation of appropriate sheets, reporting information on the classroom attendance, frequency and type of opening of the windows and doors.

Comparisons of total VOC levels across studies can be problematic due to differences in definition, sampling times, measurement, and analysis (Zhang et al., 2006), and examination of specific VOC species is often more informative.

The current study showed an increase in total VOC levels during cleaning activities, artistic activities with solvents, paints and in particular with closed windows. The decrease in total VOC levels is associated with the increase of ventilation due to the windows. These results, and the outdoor levels suggest that the indoor sources are more effective than the outdoor sources. Ultimately, it can be stated that the site investigated after energy redevelopment intervention during the observation period shows levels of concentration of VOCs and Formaldehyde below the European limits. Some criticalities are reported, however, in this study relatively to some microclimatic parameters, such as temperature and CO₂ concentration. Indeed, high values of CO₂ concentrations were recorded during school activities, with levels reaching concentrations above 2000ppm. These values are in agreement with the CO₂ levels registered by Pegas et al., 2010.

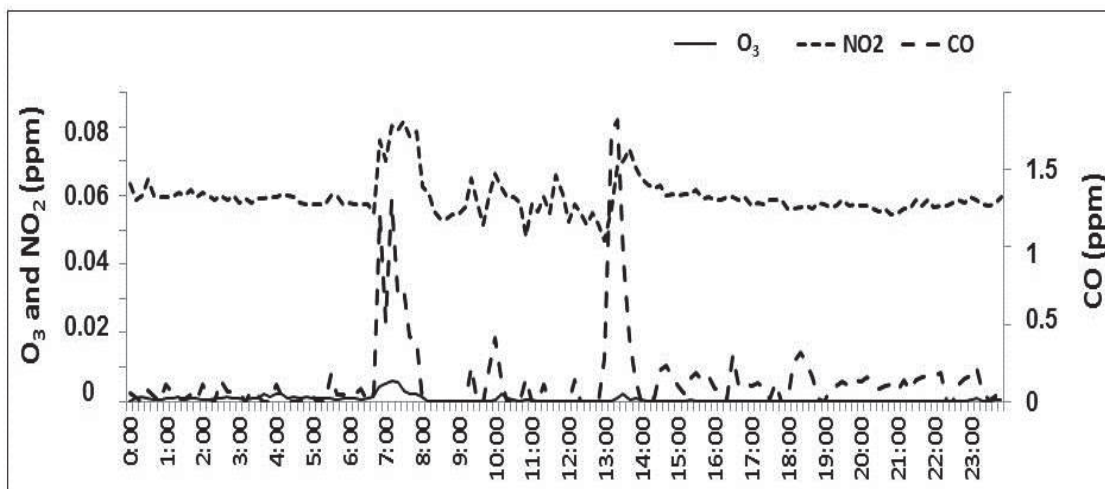


Fig. 7. Temporal trend of O₃ (solid line), NO₂ (short dashed line) and CO (high dashed line) during a single typical day

Good practices were suggested to decrease the high levels, through natural ventilation by regular window openings. This allowed to reduce the CO₂ value during the activities. To this purpose, management procedures have been identified in order to mitigate the effects consciously. Firstly, the total aperture of windows and doors for short periods rather than set the windows in the Vasistas mode for longer periods of time. The importance of ventilation of the rooms were demonstrated in the past. Indeed, the type of ventilation (mechanical ventilated or natural ventilated) plays a major role on ventilation rates in primary schools (Canha et al., 2013; Dimitroulopoulou, 2012) demonstrated the importance of ventilation during winter, comparing a Finnish school, not very much ventilated during the cold winter and a Portuguese school. Other critical points are the cleaning activities and specific arts education activities during in the school time.

4. Conclusions

To further improve the air quality one of the suggested actions is to not perform the room cleaning procedures using detergents in aerosol mode as highlighted by Fig. 2 for CH₂O, (period 27/02 and 13/03), but using the cleaning products directly on a wet cloth as reported in the spring period (18/4 and 22/4) where there is a decrease in the pollutant concentrations.

The monitored results and comparisons with the methodologies already recognized and reported in for the determination of VOCs showed the effectiveness of the integrated approach proposed for quantitative and limited surveillance of air quality in order to be able to minimize the issues related to indoor pollution in short time through the conscious management of natural ventilation. In this particular case, the use of the monitoring equipment in continuous facilitated the identification of the pollution sources in a timely manner, identifying the impact of the best management practices on the microclimate, and particularly on internal temperature and CO₂ concentrations.

The limitation of this work is the relative low number of measurements, due to limited funding availability. The study is to be considered as a starting point to develop the appropriated methodology for indoor air pollution monitoring into the classroom.

The authors in the future through other case studies, already in the pipeline, propose to implement the sensor network and to develop a prototype of the Artificial Neural Network to make the measures less and less invasive and to demonstrate the effectiveness in the surveillance action to promote the use in all indoor environments.

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EARTHWORMS FOR FEED PRODUCTION FROM VEGETABLE WASTE: ENVIRONMENTAL IMPACT ASSESSMENT

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Abstract

In the European Union, 88 million tons of food is wasted annually, 30% of which comes from the production and processing sectors. Among the different food waste, vegetable ones represent a remarkable share and their management is complicated by the usually high-water content and the difficult storage. In this context, the earthworms are an interesting solution because transform vegetable waste into valuable products: the vermicompost, that can be sold as organic fertilizer, and the earthworms that, thanks to their high protein content can be used for feed and food production. This study aims to evaluate the environmental impact related to the production of vermicompost and dry earthworm meal. LCA approach was applied, 1 kg of dry meal for feed production was selected as functional unit. Inventory data were collected during experimental tests carried out in 2017 in a composting plant located in Northern Italy where earthworms were fed with vegetable waste. Secondary data were used about emissions during earthworms rearing. A quantity of 1 kg of fresh earthworms (16% of dry matter with 67% of protein content) and 13 kg of vermicompost were produced from 45 kg of vegetable wastes. Between earthworm rearing and processing, the first one is the main responsible for the environmental impact for all the evaluated impact categories except for freshwater eutrophication and ecotoxicity. GHG emissions during composting are the main hotspots for Climate Change.

Key words: feed, protein source, vermicompost, waste valorization

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1. Introduction

Food loss and waste have a negative environmental impact due to the natural resources used for food production as well as for their management and disposal. In the European Union, 88 million tons of food is wasted annually, 30% of which comes from the production and processing sectors. In particular, the fruit and vegetable retail sector generates large amounts of waste. In industrialized countries, fruit and vegetable waste (FVW) are mainly generated before reaching consumers, during all phases of the supply and handling chain, such as market oversupply or nonfulfillment of aesthetic and quality standards (Plazzotta et al., 2017). Even without official quality standards, food retailers generally do not offer food with abnormal appearance, based on the

assumption that consumers do not purchase or consume foods that deviate from regular products, which can mean yielding lower profits (Loebnitz et al., 2015). For this reason, related to not reflecting aesthetic standards (shape, color or size), many products are discarded, even if they were produced for human consumption, they are still healthy, safe, and edible and could still reach the consumers (Stuart, 2009). FVW poses environmental problems due to the squandering of environmental, human and economic resources used to produce it and represents also a loss of valuable biomass (Plazzotta et al., 2017).

In order to reduce the impacts associated with food waste and to avoid the squandering of valuable resources, the search for sustainable solutions to the valorization of food waste is highly necessary and encouraged. A possible strategy is the utilization of

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FVW as feeding substrate for the rearing of terrestrial invertebrates to be used as potential protein source for feed and/or food supply chains. Among terrestrial invertebrates, earthworms could be an interesting solution. Earthworms grown on FVW can contribute to the waste disposal efficiency and bio-transform FVW into valuable products: the vermicompost, which can be sold as organic fertilizer, and the earthworms themselves that, thanks to their high protein content, can be a new food/feed source. Earthworms are rich in proteins, particularly in essential amino acids and they can contribute to human and animal nutrition (Yadav and Garg, 2011; Zhejun and Jiang, 2017). Currently, earthworms are just employed to convert food waste (FW) in a bioconversion process to mitigate the FW problem as a sustainable, cost-effective and ecological approach in dealing with FW management (Huang et al., 2016). Up to now, the attention on alternative protein sources has regarded mainly the insects both as human food (Halloran et al., 2016; Oonincx and De Boer, 2012; San Martin et al., 2016) and as animal feed (Smetana et al., 2016; Salomone et al., 2017). No studies addressed the environmental performances of dried meal production from earthworms.

In this context, this study aims to evaluate the environmental impact of the earthworms' dried meal production for feed purposes using fruit and vegetable waste (FVW) as feedstock. Primary data collected during field trials were combined with secondary data coming from literature; the environmental impact was quantified, and the environmental hotspots identified

2. Methodology

Life Cycle Assessment (LCA) is a holistic approach, structured and recognized worldwide that consists of a systematic set of procedures to convert inputs and outputs of the studied system into its related environmental impact (ISO 14040, 2006; ISO 14044, 2006).

In details, there are 4 steps in LCA:

- (i) goal of the study definition that foresees the selection of the functional unit, the definition of the system boundary and the solving of multifunctionality;
- (ii) Life Cycle Inventory (LCI) data collection, in which the flow of materials and energy from the studied systems and the environment are identified and quantified;
- (iii) Life Cycle Impact Assessment; during which, thanks to specific characterization factors, the inventory data are converted in few numeric indicators of environmental impact;
- (iv) interpretation of the results and identification of the process hotspots.

Over the last years, although originally developed for industrial processes, LCA has been

more and more applied also to agricultural systems (Moudry et al., 2018; Schmidt Rivera et al., 2017) and waste to energy processes (Bacenetti and Fiala, 2015; Lijó et al., 2015; Vida and Tedesco, 2015) and waste treatment solutions (Bacenetti et al., 2016; Bjelic et al., 2017; Lijó et al 2017; Salomone et al., 2017; Smetana et al., 2016).

2.1. Goal and scope definition

The goal of the present study is to evaluate the environmental impacts of the earthworms' (*Eisenia foetida*) production system reared on a low-quality substrate made of fruit and vegetable waste (FVW).

Concerning the functional unit, in this study, to avoid allocation between vermicompost and earthworms dried meal, a mixed functional unit was selected. According to ISO standards for LCA (ISO, 2006), the functional unit is defined as the quantified performance of a product system, and is used as a reference unit in an LCA. In this study, the FU is the production of 1 kg of dried earthworm meal and 80 kg of vermicompost.

Concerning the system boundary, a "from cradle to gate" approach was applied. Fig. 1 reports the system boundary for the evaluated process; two different subsystems were identified:

- Subsystem 1 (SS1), a mix of young-non-clitellum and adult-clitellate earthworms was reared on a feeding substrate consisting of FVW and straw. FVW, constituted mainly by tropical fruits, was ground and then used as feed for earthworms three times a month. Besides earthworms, during the decomposition of FVW also an odour-free and hummus-like substance is produced: the vermicompost. Vermicompost is the co-product of the production system, it can be used as organic fertilizers. After 3 months of earthworms rearing, the vermicompost and the earthworms were collected through mechanical separation;

- Subsystem 2 (SS2), the collected earthworms were processed to produce meal. First, they were repeatedly washed, then kill by cooling and, finally, dried. During the experimental trials, the dry meal was produced in a laboratory by drying earthworms in an oven at 50°C and then proceeding with grinding.

The following activities were included: raw materials extraction (e.g., fossil fuels, metals and minerals), manufacture of the different inputs (e.g., diesel, electricity, water and trucks for FVW transport), use of the inputs (diesel fuel emissions), maintenance and final disposal of capital goods (e.g., the trucks used for the FVW transport). The emissions of methane, dinitrogen monoxide and ammonia related to the vermicomposting of FVW were included too. The packaging, the distribution as well as use and end-of-life of the produced meal were excluded from the system boundary.

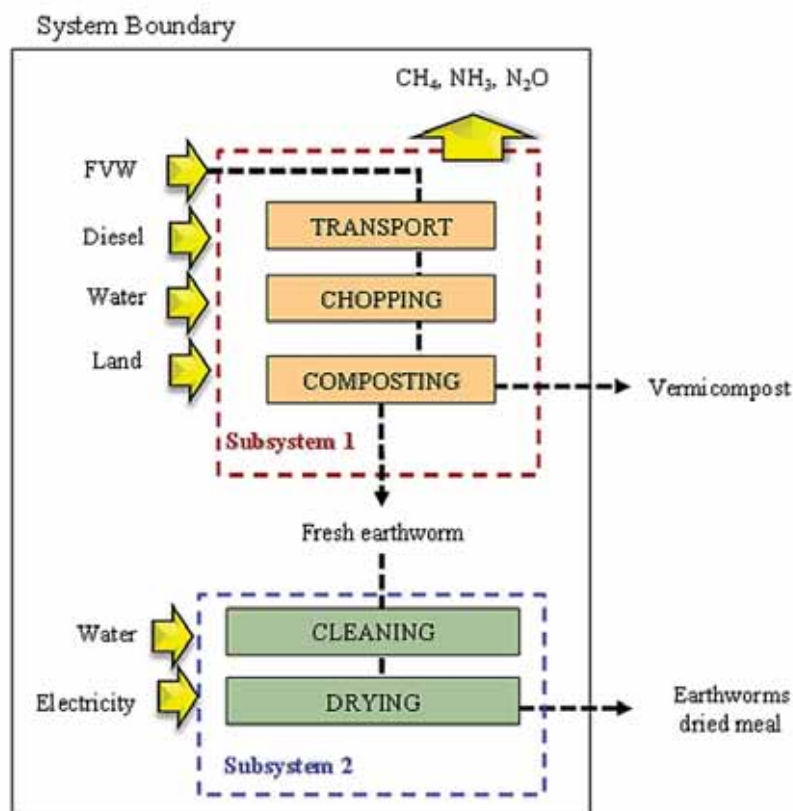


Fig. 1. System boundary

2.2. Description of the process

Earthworms were provided by a small-scale production system, located in the province of Lecco (North Italy). Earthworms were reared on an area of about 30 m² made up of FVW (growth substrate), placed above a non-woven textile sheet and covered with a net. During the rearing phase, moisture, temperature, and pH of the growth substrate were kept under control in order to guarantee optimal living conditions.

After 3 months, samples of *Eisenia foetida* at the adult stage of development were collected. The first cleaning procedure consisted of a mechanical separation from the growth substrate with the use of a trommel.

As the material rolls, anything smaller than the holes in the screen falls through, and the rest continues until it comes out the output end. Subsequently, they were washed with running tap water and soaked for some hours, to remove the residual particles of waste and to clear their gut. Finally, to produce the meal, after being frozen at -28°C, they were dried at 65°C and ground.

2.3. Life Cycle Inventory

Inventory data concerning inputs and outputs relevant to the production of earthworms' biomass were collected over a three-month experimental test

performed in year 2017.

Primary data were collected with questionnaires during interviews with the farmer and during surveys to the experimental site. More in detail the following data were directly collected: amount of FVW used as feed, fossil energy for preparing the feed substrate, water volumes and land occupation for earthworms breeding. The main secondary data refers to the emissions during vermicomposting. These emissions were retrieved from literature (Yang et al., 2017). Table 1 reports the main inventory data for the analyzed production process.

Background data was retrieved from the Ecoinvent Database v.3.5 (Moreno Ruiz et al., 2018; Weidema et al., 2013).

Table 1. Inventory data

Subsystem	Inputs/Outputs	Amount
1	Fruit and vegetable waste	285.8 kg
1	Transport of FVW	25 km
1	Diesel	1.2 kg
1	Water	22.9 kg
1	Land	2.6 m ²
1	Ammonia	99.03 g
1	Dinitrogen oxide	9.56 g
1	Methane	31.60 g
2	Electricity	2.0 kWh
2	Water	22.4 kg
2	Vermicompost	80.0 kg
2	Dried meal	1.00 kg

2.4. Life Cycle Impact Assessment

The systems considered here have been modeled using SimaPro LCA software 8.05 and the impacts estimated according to the ReCiPe method (Goedkoop et al., 2009). The following 10 impacts are considered: *Climate change* (CC), *Ozone depletion* (OD), *Terrestrial acidification* (TA), *Freshwater eutrophication* (FE), *Marine eutrophication* (ME), *Human toxicity* (HT), *Photochemical oxidant formation* (POF), *Particulate matter formation* (PM), *Metal depletion* (MD) and *Fossil depletion* (FD).

3. Results and discussions

Table 2 reports the environmental results for the different evaluated impact categories while Fig. 2 shows the environmental hotspots (i.e. the inputs or emissions mainly responsible for the total impact).

The main environmental hotspots are:

- Diesel production: the consumption of diesel fuel for grinding a share of the FVW (e.g., pineapple leaves) is the main contributor for OD (72%) and FD (69%) while for the other evaluated impact categories it is responsible of a share of the total impact ranging from 10% in CC to 31% in POF;
- Transport of the FVW to the composting plant is the main responsible for HT and POF (51%)

and 52%, respectively, mainly due to the emissions of pollutants related to the diesel combustion) and MD (54%, mainly due to the manufacturing of the truck). Similarly, to the diesel production, the transport plays a non-negligible role for all the other evaluated impact categories (from 2% for TA to 21% in TE);

- Electricity consumption during earthworm processing in SS2 is the main contributor of TE (57%) and it is responsible for about one-third of HT. For the other evaluate impact categories the role of electricity ranges from 1.8% in ME and 18% in MD. For CC, the consumption of electricity; is responsible for 13% of the total impact;

- Ammonia emission during vermicomposting is the main contributor to TA (94%), ME (94%) and PM (85%, due to the formation of secondary particulate);

- Dinitrogen oxide emission deeply affects CC with 45% of the total impact.

With regard to the other inputs or emissions:

- The consumption of water in SS1 (for rearing humidity maintenance) as well as during SS2 (for cleaning) is responsible for a small impact (<2% for all the evaluated impact categories);

- The emissions of methane only slightly contribute to CC (about 12%) and POF (2.6%, due to the emission of CH₄).

Table 2. Environmental impact for the selected FU

Impact category	Acronym	Unit	Score
Climate change	CC	kg CO ₂ eq	6.327
Ozone depletion	OD	mg CFC-11 eq	1.142
Terrestrial acidification	TA	kg SO ₂ eq	0.257
Freshwater eutrophication	TE	g P eq	0.429
Marine eutrophication	ME	g N eq	9.637
Human toxicity	HT	kg 1,4-DB eq	0.618
Photochemical oxidant formation	POF	g NMVOC	12.332
Particulate matter formation	PM	g PM10 eq	36.997
Metal depletion	MD	g Fe eq	72.318
Fossil depletion	FD	kg oil eq	2.212

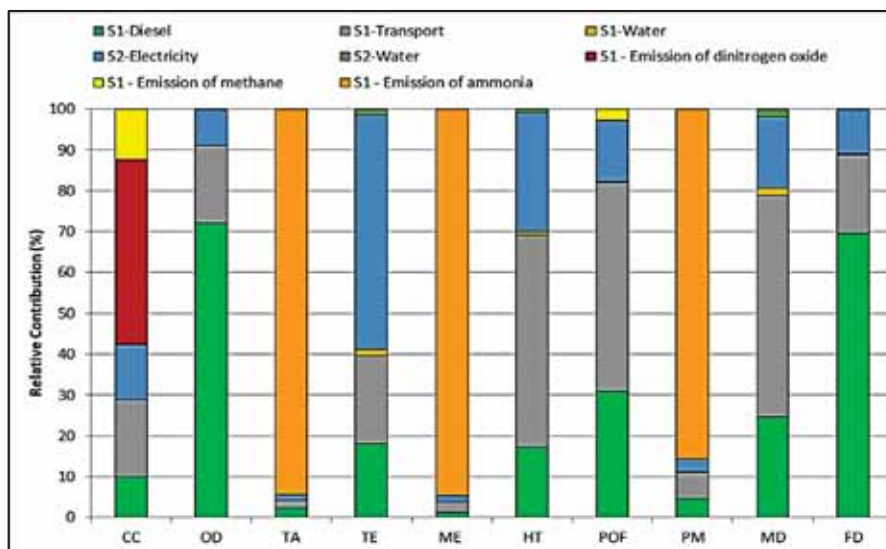


Fig. 2. Identification of the environmental hotspots (S1 = subsystem 1, S2 = subsystem 2)

Between the two subsystems, SS1, with a share of the total impact ranging from 70% in HT to 98% in TA and ME, is the main contributor for all the evaluated impact categories except than for TE. For this last impact category, 59% of the impact is related to SS2.

The environmental impact of the earthworms' dried meal production for feed purposes using fruit and vegetable waste (FVW) as growth substrate showed a higher CC value associated with its production; this was caused by the considerable energy input for FVW transport and drying process. This could be reduced if the vermicomposting process takes place at the FVW production site. Moreover, Europe's reliance on imported protein, particularly soybeans, to feed livestock is inconsistent with sustainability objectives because soybean is associated with deforestation and impacts from pesticide use and transportation (Tallentire et al., 2018). The environmental burden for soybean meal is 3.05 kg CO₂ eq kg⁻¹ (Tallentire et al., 2017). This means that improvements of the earthworms' dried meal production for feed purpose using FVW could be a promising research field how even the necessity of alternative protein sources in terms of minor warming potential and reduction of food waste (Conti et al., 2018).

4. Conclusions

By means of the Life Cycle Assessment (LCA) method, the environmental impact of the production of earthworm-dried meal was quantified. The feed substrate for earthworms is made of fruit and vegetable waste (FVW) that, therefore, is highly valorised respect to wasting. Given the increasing importance worldwide of issues related to food waste, the transformation into feed and/or food meal is very promising. Besides the not negligible environmental impact, this production system brought benefits such as the recovery of FVW as feeding substrate, the earthworm production as a food/feed source with high nutritional profile, and the availability of vermicompost as an organic fertilizer that allows reducing the use of mineral fertilizers in other production systems.

Similarly, to other protein sources, earthworm dried meal currently has high environmental impacts mostly due to the transport of FVW for fresh earthworm production and energy use during processing. To make earthworm meal sustainable and competitive on the market, enhancing earthworm productivity and reducing energy costs of the processing stages by shifting towards renewable energy sources is essential.

Additional research and integration with innovations among different sectors are the key drivers for the near future. However, the outcomes of this study can be useful for the development of a subsidy framework supporting the earthworm dried

meal production chain thanks to the identification of the hotspot stages and their possible mitigations.

Acknowledgements

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DESIGN AND ENVIRONMENTAL ASSESSMENT OF BIOPLASTICS FROM *Hermetia illucens prepupae* PROTEINS

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Abstract

Proteins from *Hermetia illucens* (or Black Soldier Fly, BSF) have been employed in this study as possible source for bioplastic formulation. This type of bioplastic can replace the actual materials employed in agriculture, avoiding the critical issues concerning the soil pollution due to conventional plastic end-life. Different plasticizing agents (glycerol and polyethylene glycol) have been tested and the ability to generate a homogenous film, through wet casting, has been evaluated. Characterizations on tensile properties and water absorbance have been performed to estimate the effect of different plasticizers employed. Bioplastic formed by proteins/glycerol ratio 50:50 has shown interesting properties, contributing to the formation of homogeneous and free-standing film with tensile stress at break near to 2.5MPa, almost constant during degradation profile test. At the same time the high degree of solubility in water has been verified for the same sample (~70%). The environmental impact of the laboratory scale production of bioplastics obtained from BSFs proteins has been evaluated through the Life Cycle Assessment (LCA) methodology. Inventory analysis has been conducted using primary data and Ecoinvent database. LCA analysis has been conducted using the SimaPro 8.3 software and the IMPACT 2002+ method of evaluation. The analysis show that the energy consumption is high (63%), but this can be mainly attributed to a laboratory-scale production process and related with the energy consumption of aspiration system (93%). Therefore, these results will help to the design of industrial production of innovative bioplastics in order to minimize these environmental issues.

Key words: bioplastics, glycerol, *Hermetia illucens*, LCA, waste bioconversion

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1. Introduction

The worldwide plastic production and consumption is continuously increasing, generating in Europe an economic return of trillions of dollars (PlasticEurope, 2018, Comăniță et al. 2016). Plastic consumption for agricultural purpose is estimated as 2–3 million tons for year and at least half of these are employed for the cultivation's protection as soil mulching films, low tunnels, greenhouses and temporary coverings (Dilara and Briassoulis, 2000; Espi, 2006; Huang et al., 2017; IBAW, 2018). Low-density polyethylene (LDPE) is the most common materials employed for the protections of the cultivation, considering its low cost compared with

good mechanical and optical properties (Briassoulis, 2005). One of the main drawbacks of LDPE employment is about lifetime that ranges from few months to 3–4 years depending on peculiar conditions near the soil and geometrical properties (e.g. thickness) (Desriac, 1991; Lemaire, 1993). In fact, LDPE degradation comes as a fragmentation of the materials in small pieces of plastic that are too small to be removed and which often can be founded as pollutant in the soil (Scott, 1999). To avoid this type of pollutant on soil and cultivation and to reduce the disposal costs, films based on biodegradable materials must be considered as favorable alternative, even if concerns arise from the vegetable exploitation for plastic production (Briassoulis, 2005; Jachowicz et al.,

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2017; Krzan et al., 2006; Malinconico et al., 2002; Wu and Wang, 2018). Moreover, biodegradable films, if properly studied, can act as fertilizers, releasing nutrients, such as nitrogen, during their decomposition, and therefore producing an innovative and totally green sustaining source for cultivation (Scott, 2000).

As innovative aspect, the present study investigates the exploitation of prepupae extracted from *Hermetia illucens* (Linnaeus 1758-Diptera: Stratiomyidae), also known as black soldier fly (BSF), to generate a free-standing bioplastic film. BSF is a considerable source of nutrients, in fact its proteins are employed as base for animals feed, and fats in biodiesel production (Cummins et al., 2017; Newton et al., 1977). BSF is also known to act as safe bio-converter for the human being, acting as waste reducer without the transmission of pathogen agents, generating a strongly beneficial effect in the circular economy perspective (Caligiani et al., 2018; Diener et al., 2011; Sheppard et al., 2002; Spranghers et al., 2017). In the present study, the formulation of bioplastic for agricultural purpose has been studied taking in account proteins extracted from BSF prepupae, previously reared on poultry manure, by mixing with selected plasticizers. Therefore, insects are employed to reduce waste's volume, and their fractionated prepupae as source of proteins, useful as base for a new category of materials interesting for agricultural purpose following the scheme shown in Fig. 1.

In order to evaluate the environmental profile of insect-based products, the environmental impacts associated with the whole life cycle of these processes has been quantified through a Life cycle assessment (LCA) evaluation. LCA is essential to identify and assess entails identifying and assessing the potential environmental impact associated with a material, product, service or process throughout its entire life cycle, from the raw material extraction and processing, through manufacturing, transport, use and final disposal. However, the scientific literature on bioplastic obtained from insects is still limited. Using the LCA approach, Oonincx et al. (2012) published a detailed environmental impact assessment in terms of global warming, agricultural land use and energy

consumption for the mass farming of two species of mealworms (*Tenebrio molitor* and *Zophobas morio*) in comparison with traditional protein sources for human consumption (milk, chicken, pork, beef). Their results highlighted greater GHG emissions and land use associated with milk, chicken, pork and beef systems, whereas similar amounts of energy are required in conventional and mealworm protein production. However, Oonincx et al. (2012) reported that the high energy consumption observed for rearing of insects is due in large part to the need to air-condition the breeding environment. Similar conclusions are reported by Van Zanten et al. (2015), that focused the attention on the use of housefly larvae grown on poultry manure and food waste as livestock feed. Although, Boer et al. (2014) employed LCA to find that mealworms seem to have little potential for inclusion in compounded feed without increasing the carbon footprint, they also concluded that the use of other insect species, with a low energy requirement during rearing and higher nutrition values, reared on waste products instead of feed ingredients, could increase the replacement potential of insects.

LCA methodology was employed by Salomone (2017) to evaluate possible environmental impacts of bioconversion from food-waste to *Hermetia illucens* dried larvae. Their results confirm that Energy Use category is the main burden, although significant benefits are related to Land Use category if data from food-waste conversion are compared with alternative sources for biodiesel or feed. Therefore, in the present study, the environmental impact evaluation has been carried out adopting the LCA methodology (ISO 14040, 2006; ISO14044, 2006) to evaluate the environmental performance of the production process of the best innovative bioplastic formulation obtained from the experimental work, in order to estimate if, as base material for plastic, the insect exploitation demonstrates a lower environmental impact, than as alternative for feeding. In this study the focus has been concentrated on the environmental impact associated with the laboratory-scale production. The aim has been to develop a detailed picture of the environmental profile of the bioconversion process, generating new data-set to be available for future similar studies.

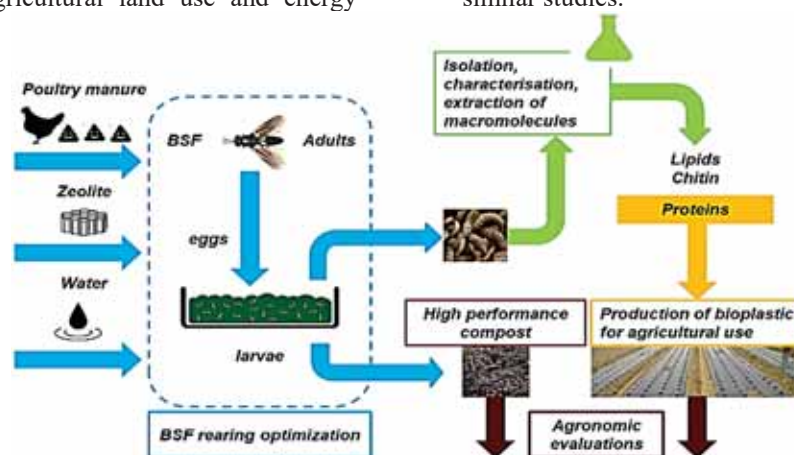


Fig. 1. BSF as renewable resource for bioplastic production

2. Material and methods

2.1. Films preparations

The BSF prepupae protein fraction was derived through a conventional chemical route, as reported in literature, obtaining a protein amount of 32wt% on the overall BSF protein content (Caligiani et al., 2018). The protein fraction was firstly ground with an analytical mill (IKA, A10 basic) and then sieved below 40 μm to obtain a powder with homogeneous grain size and able to react consistently with the selected plasticizers. Glycerol (GL, 99%, Sigma Aldrich), sodium hydroxide solution (NaOH, 1M, Sigma Aldrich) and polyethylene glycol (PEG400, 99%, Sigma Aldrich) were employed as additives for protein-based film formulation.

Films were obtained by mixing GL (or PEG 400) and protein, in distilled water (DI), adjusted to pH 10 with NaOH (1M) following the formulations expressed in Table 1. These suspensions were firstly heated for 30 min at 70°C and stirred at 200 rpm, then poured into aluminum dishes. The cooling was done under fume hood at room temperature for 24 h.

2.2. Films characterization

A dynamic mechanical analyzer (DMA, TA Q800) was employed in film tension set-up for tensile properties evaluation. Before measure each specimen was kept at standard conditions (50% Relative Humidity; 25°C) for 24 h. Rectangular specimen of 20 \times 5 mm² were employed and run in duplicates for each mixture shown in Table 1. The effective sample length was measured in the film stage assembly under a pre-load force of 0.05 N at room temperature. During testing the load force was raised to 18 N at the rate of 0.05 N min⁻¹ to measure the relative elongation.

Specimen's thickness and diameter were evaluated with a digital micrometer (Mitutoyo, YY-T1BD-2GYE) in fifteen different points, and the average value was taken as reference together with its calculated standard deviation. The sensibility of the instrument was 0.02 mm.

The moisture content (MC) was evaluated following Eq. 1 and measuring each sample's weigh before (w_0) and after (w_1) drying in oven at 105 °C for 24 h. (AOAC, 1995): (Eq. 1)

$$MC(\%) = \frac{(w_0 - w_1)}{w_0} * 100 \quad (1)$$

The water solubility (WS) was determined through Eq. (2), where w_2 is the weight of each sample after immersion in 200ml of distilled water for 24 h and drying in oven at 105 °C for 24 h (Gontard et al., 1994):

$$WS(\%) = \frac{(w_1 - w_2)}{w_1} * 100 \quad (2)$$

An analytical balance with sensibility of 0.00001g was employed to measure all the weights.

EN 17033:2018 and EN ISO 4892-2:2013 Method A cycle 1 were employed for the measurement of the degradation profile due to artificial weathering. According with these standards rectangular specimens (50x15mm) were employed and exposed in a closed chamber at irradiance of 0,51 W/(m² x nm), at 38°C with 65% of relative humidity 65% for 500 hours. At the same time deionized water was sprayed into the closed chamber for 18 minutes over 2 hours. After the exposure tensile mechanical properties were measured.

2.3. Life Cycle Assessment (LCA)

The functional unit was represented by the amount of bioplastic produced (0.403 gr). The system boundaries (Fig. 2) range from the obtained BSFs protein to the bioplastic production following the procedure mentioned in paragraph 2.1. Energies, materials, water, main equipment with their end of life, transport, waste and their treatment, emissions into the air at the continental level, the aspiration system and purification plant as well as the recovery and reuse of certain solvents were also considered.

Table 1. Material's formulations (wt%)

<i>Experiment</i>	<i>Protein</i>	<i>GL</i>	<i>PEG400</i>	<i>DI</i>	<i>Score</i>
GL1	12	12	/	76	5
GL2	14	10	/	76	3
GL3	16	8	/	76	3
PG1	12	/	12	76	3
PG2	14	/	10	76	2
PG3	16	/	8	76	1

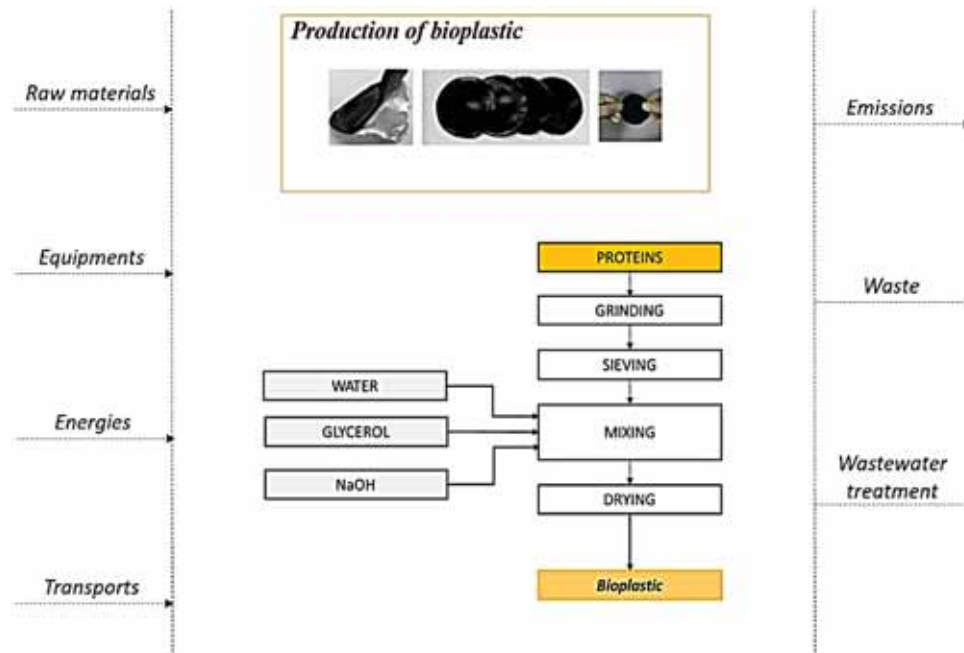


Fig. 2. LCA scheme for bioplastic production

2.3.1. Life Cycle Inventory

Regarding the quality of data used for the Life Cycle Inventory (LCI), lab-scale data were directly collected from the experimental procedure. Where the data were not available, the study was completed on the basis of secondary data obtained from the Ecoinvent databases v3 (Ecoinvent, 2013) which were used to model the background processes (land use, material production, fuel and electricity production, and transport). The emissions were calculated assuming that the laboratories were fully ventilated after the conclusion of each working day (8 hr). For these emissions, the indoor concentrations were calculated considering a total laboratory volume of 480 m³ and assuming that 1% of the emissions come from the aspiration filters with 99% efficiency. The other extracted fraction, related with lipids and chitin content, were not used for the production of bioplastic, and thus were considered as co-products. A mass allocation was used. A composting process was hypothesized as bioplastic end of life.

2.3.2. Impact assessment methodology (LCIA)

The life cycle impact assessment (LCIA) results were modeled using the IMPACT 2002+ method (Jolliet et al., 2003) with Simapro 8.5 (PRè Consultants, 2013) to determine the environmental impact. This impact assessment method covers more impact categories than other methods and includes more substances. Since it is midpoint and endpoint oriented, it provides a complete overview of the environmental performance. However, the following additions and modifications were implemented to describe the system considered in a more representative manner i.e. modification to Land use (different types of land transformations were considered), Mineral extraction categories (additional resources were added) and Radioactive waste

(radioactive waste and its occupied volume were evaluated) (Spinelli et al., 2014). The LCIA results were derived from both midpoint and endpoint levels. However, for the sake of brevity we report only the endpoint results. These are usually shown as the impact on human health, ecosystem quality, climate change and resource depletion. We decided to report only the endpoint results as the interpretation of these results does not require extensive knowledge of the environmental effects. Moreover, midpoint results can be more difficult to interpret because they consider many impacts which are often difficult to understand.

3. Results and discussion

3.1. Materials evaluation and characterization

The evaluation of the capability to constitute a free-standing plastic film has been performed firstly from a qualitative point of view through a consensual panel based on the judgment in blind of five people. The output of each experiment has been evaluated taking into account the homogeneity of the sample after drying and, therefore, its compactness and detachability from the aluminum support. Panel test grouped all experiments in 6 categories and a score from 1 to 6 has been attributed at each one category. In details, 1 is the value associated to the lowest quality (totally not homogeneous), whereas 6 is attributed to samples with the highest quality (completely compact, homogenous and with good detachability). From the collected scores, as expressed in Table 1, is clear that the decreasing of plasticizer content leads to the weakest quality in terms of homogeneity, compactness and detachability. Moreover, the type of plasticizer plays a key role, in fact, an improved compactness and detachability can be related to the employment of GL, whereas PEG400

leads to the worst overall result. Independently of plasticizer type and content, detachability has to be improved, in fact none of the samples studied in this work reached the highest score of 6.

Plastic integrity during processing, handling, usage and storage are governed by their mechanical properties. In the present work tensile behavior of the produced samples has been measured following the condition in section 2.2, confirming quantitatively the result from the panel test. In fact, as can be seen in Fig. 3 higher tensile stress at break (σ_b) corresponds to the employment of GL, reaching the highest value of 2.4 MPa for the sample GL1. A drop of σ_b value corresponds to a decreasing of the plasticizer content, for both GL and PEG400, whereas samples produced with plasticizer content <12wt%) show very similar values of σ_b (GL2 with GL3 and PG2 with PG3). This result confirms that at least 12 wt% of plasticizers (protein-based content) is necessary to achieve the highest results in term of tensile resistance at break, suggesting that, in other conditions, the microstructure configuration due to the additive content is not suitable to perform a strong binding between proteins due to poor chain linking density of the resulting polymer (Ganglani et al., 2002). Difference in GL and PEG400 behavior as plasticizer are consistent with their different functional groups that leads to a different protein-plasticizer interaction; more in details PEG 400 promotes the formation of more hydrophobic interactions that contributes to decrease the protein-plasticizer bonding (Knowles et al., 2015) whereas glycerol plays a chain linking action in the material's polymerization.

In fact, glycerol promotes its diffusion into proteins chain, because of its restrained molecular weight combined with high hydrophilic behavior, generating a restrained hydrogen-protein bonding (Awadhiya et al., 2016; Lunt and Shafer 2001; Martelli et al. 2006). Similar considerations can be done for the tensile stress at yield point (σ_y) that results lower than the tensile stress at break for all the

investigated samples indicating that the elastic component of the strain is strongly higher than the plastic one, leading to poor ductile materials (even if the standard deviations suggests a partial overlap of the confidence ranges). The results from mechanical tests can be compared with data obtained in very similar conditions from bioplastic from others animal sources, such as crayfish, keratin and albumen. In particular, Ramakrishnan et al. (2018) considers the keratin employment together with glycerol to produce bioplastic through casting technique, obtaining lower tensile strength at break with respect to the results of the present study. Similar consideration can be drawn taking in account Felix et al. (2014) that consider crayfish as bases for bioplastic production, employing glycerol as plasticizer through injection molding.

Therefore, an overall increase of the mechanical tensile resistance at break can be obtained if protein from BSF are employed with respect to other protein from animal sources, although not reaching the values of materials already available on the market and deriving from vegetables, such as starch, that shown tensile stress at break around 20MPa and strain at break over 200% (Niaounakis, 2015).

The degradation profile against time of the investigated samples has been measured as shown in Fig. 4. For all the investigated samples, except GL1, an almost linear inverse proportion among tensile stress at break and exposure time has been measured with similar rate of degradation. In fact, GL1 not only shows the highest values of σ_b with respect to others but remains the sample with the highest tensile stress at break during and after weathering test. This confirms that the protein-additive combination of this sample is the most favorable to the obtainment of a polymeric material more stable during exposure to the weathering agent. This result is consistent with an only partial degradation, as described in Fig. 4, since weathering agents should be taken in consideration together with burial in soil test to achieve a complete result of degradation.

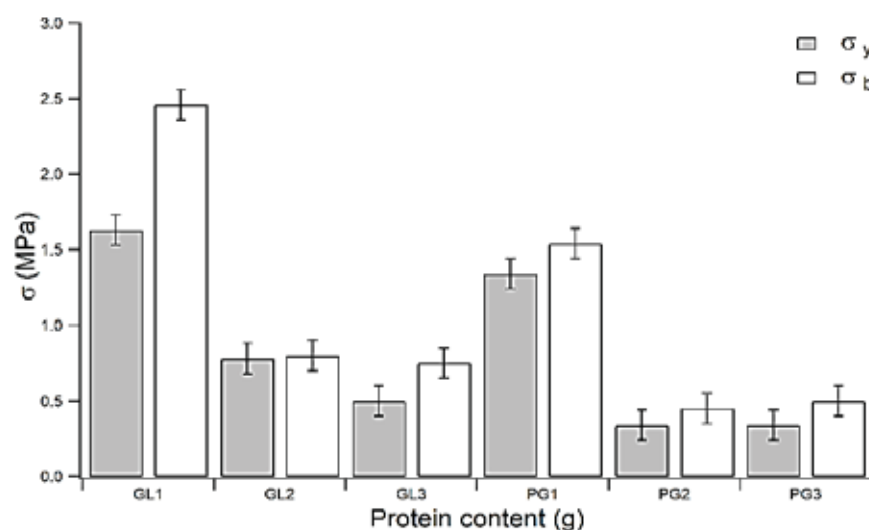


Fig. 3. Mechanical characterization

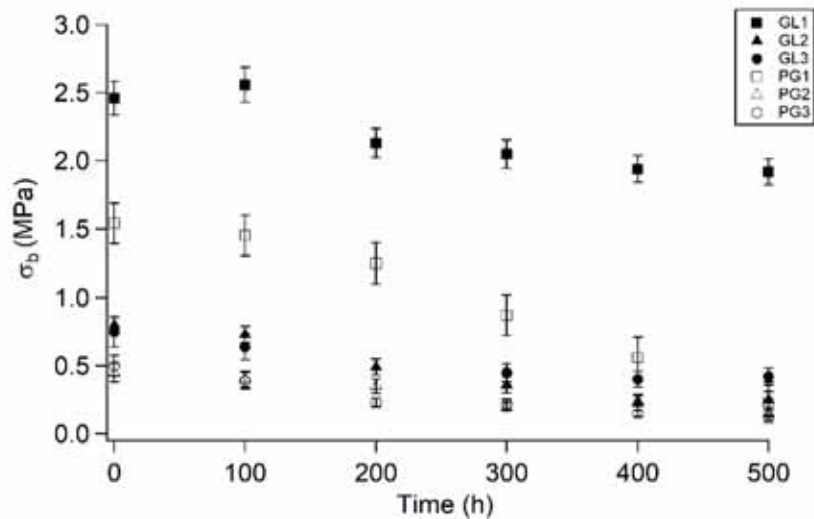


Fig. 4. Degradation profile

As shown in Fig. 5, the quantity of protein in plastic's formulation regulate both the moisture content and water solubility due to protein's high hydrophilic behavior.

Moisture content increases by decreasing the quantity of protein employed into material's formulation and this result is consistent with the strong hygroscopic behavior of the protein chains. This trend is true until 12 wt% of protein into the materials formulation, in fact after this value, considering the statistical error, samples have the same moisture content.

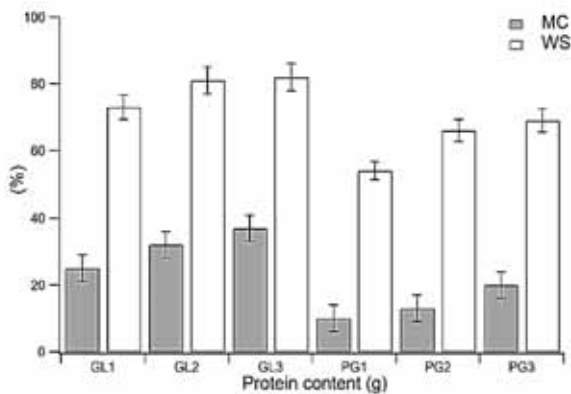


Fig. 5. Water absorbance and Moisture content comparison

Consequently, this protein quantity is a threshold for moisture content increment; In fact, over 12 wt% the packing of the protein chain becomes more open, due to restrained plasticizer's quantity, letting moisture be adsorbed more easily by the material.

The water solubility (WS), also known as total soluble matter (TSM), follows the same trend. As demonstrated in similar studies this parameter is strongly correlated with the amount of highly hydrophilic components into the materials formulation from which a high moisture content is derived.

High WS suggests also that proteins are not strongly bonded to the network structure (Felix et al., 2014). Similar trends can be associated with GL and PEG400 employment, even if lower values of WS and MC have been obtained with PEG400 due to the lower hygroscopic behavior if compared with GL. From these results the protein content > 12 wt% into material formulation promotes a higher content of nutrient not strongly bonded.

Therefore, in this condition higher is the protein content to be released in soil, acting as soil fertilizer during the plastic degradation (Chiellini et al. 2001; Gontard, 1994).

3.2. LCA

The analysis of results (Fig. 6) shows that the total damage associated to 1 gr bioplastic (following GL1 composition) production process is equal to 3.4296 mPt.

Furthermore, the main environmental impact is mainly due to energy consumption (63%), in particular related to the energy consumption of aspiration system (93%) needed for the drying of the specimens under aspiration hoods. In fact, the final part of the bioplastic production is related to the drying of the samples and it takes the greatest fraction of time of the overall process of production. Table 2 reports the environmental performance at end-point level (damage categories): Human health, Resources and Climate change categories afflict the total damage for 31.48%, 32.06% and 26.60% respectively.

In particular, in Human health category the major contribute to the total impact is due to particulates (<2.5 μm) emission in air; Resources category is mainly affected by natural gas and the emission that mainly contributes to the climate change is carbon dioxide. All these emissions are due to electric energy production employed by the aspiration system.

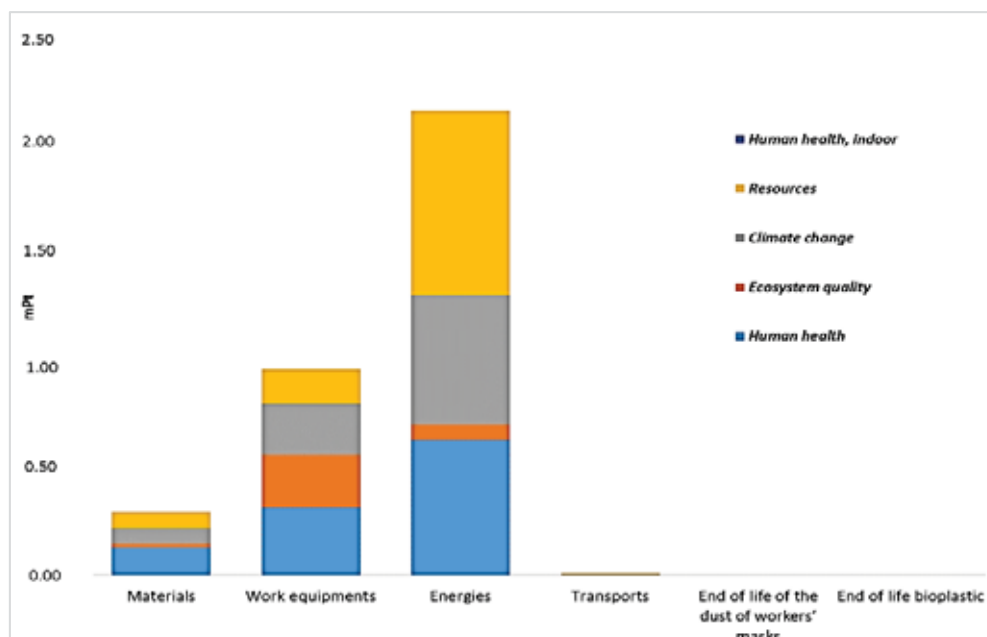


Fig. 6. Environmental damage of bioplastic production (1gr) by single score

Table 1. Environmental damage of bioplastic production process (1gr) by single score

Damage category	Materials	Equipments	Energies	Transports	End of life of the dust of workers' masks	End of life of bioplastic
Human Health (DALY)	1.29E-01	3.16E-01	6.32E-01	2.21E-03	1.60E-10	5.96E-06
Ecosystem quality (PDF*m2*yr)	1.93E-02	2.46E-01	7.16E-02	9.83E-04	2.05E-11	8.16E-07
Climate change (kg CO2 eq)	7.07E-02	2.37E-01	6.03E-01	1.89E-03	1.20E-10	3.61E-06
Resources MJ primary	7.70E-02	1.60E-01	8.61E-01	2.08E-03	5.54E-11	3.53E-06
Human Health Indoor (DALY)	3.32E-07	-	-	-	-	1.55E-07
Total [mPt]	2.96E-01	9.59E-01	2.167E+00	7.16E-03	3.57E-10	1.41E-05

“Occupation, forest, intensive” category affects Ecosystem quality damage (9.85% on the total damage) and it is generated by wood used for the activated carbon filter production (filter typology installed in the aspiration system).

These results are consistent with the available literature about life cycle assessment of possible new field of application for proteins that describe the energy consumption for air-condition/heating/drying as the category with the highest environmental impact. In particular, as already reported by Van Zaten (2015), the environmental impact is high nevertheless the rearing of the insect is made employing waste as livestock feed.

Moreover, the employment of Black Soldier Fly increases the energy consumption according to Salomone (2017) although insects are employed to produce plastic due to the lab scale condition of production, therefore in scale-up perspective the aspiration system employment should be strongly reduced or substituted.

4. Conclusions

This study confirms that proteins extracted from Black Soldier Flies prepupae can be employed to obtain bioplastic films, promising as bio-compostable plastics. The added value of these films is that they are generated by waste processing and reduction in volume by insects, increasing their positive effect in a circular economy perspective.

The addition of glycerol as plasticizer shows high potential due to favorable mechanical properties if the content of protein is near 12 wt% also after aging tests. Nevertheless, tensile stress at break must be increase in order to obtain values nearer to other bioplastic on the market, by employing other additives. PEG400 has shown very poor beneficial effect on film generation due to different functional group and bonding related to the interaction with protein with respect to glycerol.

LCA results show that the energy consumption of aspiration system is near to 63%, but this can be mainly attributed to a laboratory-scale production

process and related with the energy consumption of aspiration system (93%). This consumption should be reduced in order to minimize the environmental impact, even if this process step it is necessary for the drying of the materials. This environmental impact is perhaps due to the production process is a laboratory-scale process and not yet an industrial one. Therefore, these results will help to the eco-design of industrial production of innovative bioplastics in order to minimize these environmental issues.

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IMPACT OF EFFLUENTS FROM WASTEWATER TREATMENTS REUSED FOR IRRIGATION: STRAWBERRY AS CASE STUDY

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Abstract

This research is intended to study the possible transfer of the residual chemical contamination from treated wastewaters reused for irrigation purposes of *Fragaria x ananassa* strawberry (cv. *Camarosa*). Different sewages from urban, and mixed urban-textile origins treated according to different treatment trains were used for the irrigation of strawberry in pots. Organic and inorganic chemical contamination indicators, i.e.: PCBs, including dioxin-like congeners, PAHs, and Cr(VI), were monitored along the whole agricultural production chain (wastewater treatment effluents, soil and crop). Robust analytical procedures were specifically developed for the determination of contaminants in the above-mentioned matrices with quantitation limits (MQLs) ranging from 1.3 (Phe) to 11.0 ng/L (PCB169) for wastewaters; from 3 (PCB180) to 10 µg/kg (BbF1) for soils; from 1.0 (Phe) to 10.9 µg/Kg (PCB169) for strawberries. For Cr(VI), limits were 0.15 µg/L (waters) and 0.018mg/kg (soils). These performances fully satisfy limits fixed by Italian or European regulations on maximum admitted concentration of pollutants in treated water intended for irrigation, in soils and crops. Even if selected PAHs and PCBs were detected in wastewaters (highest concentrations observed for phenanthrene, 429 µg/L, and PCB52, 110 µg/L) their presence was not observed in soils and in strawberries above the MQLs. On the contrary, chromium content in strawberries and soils irrigated with TWs suggested a possible transfer of the metal during irrigation, which however does not represent a hazardous situation for consumers since calculated daily intake does not exceed the Tolerable Daily Intake of 300 µg/kg b.w.

Key words: food chain; reuse impact; strawberry; treated wastewater

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1. Introduction

Agriculture is characterized by a high-water demand; one third of the water use in Europe is addressed to the agricultural sector, most of it for crop irrigation (EEA, 2012). Italy is one of the Countries with the highest water footprint in Europe, 25% above

the European Union average, and, at a global level, 66% above the world average (Sartori et al., 2014). It is worth to be mentioned that agriculture in Italy accounts for about 85-89% of the water footprint of consumption.

The availability of fresh water (FW) is a problem of increasing concern in the world and

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climate change will likely introduce significant variations in water availability in the Mediterranean region, where aridity or droughts periods could originate conflicts among water uses. Water reuse is a possible solution to face water scarcity in view of preserving high-quality water that can be used for human consumption. EU is defining minimum requirements for water reuse, with the general objective to alleviate water scarcity across the EU, in the context of adaptation to climate change, notably by increasing the uptake of water reuse (European Commission, 2018), in particular for agricultural irrigation. In this context, treated wastewater (TW) might provide a reliable supply. Area under wastewater irrigation has increased in arid developing countries, with Tunisia having the largest experience in wastewater reuse in agriculture dated back to the early '60 (Haddaoui et al., 2016).

The reuse of municipal or mixed municipal/industrial TWs for irrigation could be an efficient tool of reducing water shortage, but can negatively affect plant growth and productivity, so that a number of concerns regarding environmental and health aspects should be taken into account (Norton-Brandão et al., 2013).

Among chemicals of high environmental concern, still potentially present in TWs, Cr(VI) and organic micropollutants arouse a great attention. Cr(VI) is a hazardous compound for human health, frequently used for chrome plating, dyes and pigments, leather tanning, and wood preserving. Cr(VI) is mobile in the environment and can easily penetrate the cell wall, exerting its noxious influence in the cell itself, being also a source of various cancer diseases [IARC Group 1] (Straif et al., 2009). Also polycyclic aromatic hydrocarbons (PAHs) are a frequently detected class of water pollutants (Bruzzoniti et al., 2010); they are mainly formed by the incomplete/inefficient combustion of organic material (anthropogenic source), by diagenesis and biosynthesis. (Boehm, 1964). It is well known that PAHs are recalcitrant and that they are mutagenic/carcinogenic pollutants. Hence there is serious concern about their presence in the environment, especially for their tendency for bioaccumulation in food chains (Boga et al., 2018; Yan et al., 2004), particularly due to their lipophilicity (Balducci, 2008). Similar behaviour can be pointed out for polychlorinated biphenyls (PCBs). PCBs were manufactured and used in industry as heat transfer fluids, hydraulic lubricants, dielectric fluids for transformers and capacitors, plasticizers, pesticide extenders and were detected in the environment since 1966 (Jensen, 1972). This class of pollutants is divided into dioxin-like (polychlorinated non-ortho and mono-ortho biphenyls) and non-dioxin-like PCBs. The higher toxicity of dioxin-like PCB compounds is ascribed to their role in the activation of the Ah receptor (AhR), responsible for gene expression (Giesy and Kannan, 1998).

Several recent studies showed that PAHs/PCBs are still found in TWs even after tertiary refinement

processes such as filtration by activated carbons (Dimpe and Nomngongo, 2016, Petrie et al., 2015). Cr(VI) can still be detected in wastewater effluents of textile districts. (Fibbi et al., 2012). If PAHs and PCBs are present in waters as a result of natural and anthropogenic processes which produce both point source and diffuse emissions, Cr(VI) is the result of point source emission mainly through textile waters. Based on the aforementioned considerations, the aim of this research was to investigate possible transfer effects along an agricultural production chain under irrigation with municipal and mixed textile TWs. *Fragaria x ananassa*, *Camarosa* cultivar was chosen as model plant, since it accounts for about 60% of the world's production and it adapts greatly to wide climate and growth conditions. A comprehensive monitoring approach was followed, considering the residual contamination of PAHs, PCBs, and Cr(VI) in: (i) TWs used for irrigation; (ii) soils where strawberry cultivars grown up and (iii) the strawberry crop.

For this purpose, suitable extraction protocols and analytical methods were positively developed. To the best of our knowledge, the use of industrial or mixed municipal/industrial TWs for crop irrigation or plant nursery is under-investigated (Gori and Caretti, 2008, Hashem et al., 2013, Lin et al., 2000, Sou et al., 2013, Vergine et al., 2017). In fact, in no case, studies about the fate and the accumulation of pollutants in the agricultural and food chain are as comprehensive as the one presented in this manuscript, since only a partial uptake evaluation (i.e. water-soil or water-crop transfer) is presented. Moreover, organic and inorganic pollutants fate was also not considered simultaneously (Amin et al., 2013; Arora et al., 2008; Khan et al., 2008; Kipopoulou et al., 1999; Song et al., 2006).

2. Material and methods

2.1. Treated waters and sampling campaign

Five types of TWs were used to irrigate strawberry plants. TW1: mixed urban/industrial wastewater treated by primary settling, biological oxidation, secondary settling, clariflocculation, ozonization. TW2: TW1+ clariflocculation, sand filtration, activated carbon, disinfection with hypochlorite. TW3: TW1+ clariflocculation, sand filtration, dilution with river water, disinfection with hypochlorite. TW4: as TW1 with sewage incoming composed by mixed urban/industrial, aerated septic tank wastewaters and by landfill leachate pre-filtered with a membrane biological reactor. FW: drinking water (control).

TWs were provided by the wastewater treatment plant GIDA (Prato) every fifteen days and stored in dark tanks in the irrigation site. Samples were withdrawn at the 1st and 8th day from the filling of the tanks, stored at -10°C until analysis. Physicochemical, chemical and microbiological (data not reported) characteristics of TWs and FW were monitored within the irrigation period (2017) which is labelled as: A,

May; B, June; C and D, July; E, August; F, September; G, October. Analyses were performed in triplicate.

2.2. Experimental cultivation plant

The experimental site (Fig. 1), implemented at the scientific campus of the University of Florence, consists of five rows, composed by a water tank for each TW studied and seven pots (each one containing 10 strawberry plants).

Certified plantlets of strawberry (*Fragaria x ananassa*, cultivar “Camarosa”), purchased from Vivai Fratelli Zanzi (Ferrara, Italy) were transferred in 80-liters pots (ten plantlets per pot) filled with commercially available top-soil for fruit and vegetable nursery. A porous, expanded commercial perlite (AGRILIT 3) with a grain size of 2 -6 mm, specifically developed to be used as a growing medium and/or soil improver, was added to each strawberry pot. Plantlets were irrigated with four TWs, and FW as control. Pots were covered with a plastic tunnel in order to protect the plants from animals and to avoid the direct contact with rain water, which interfere with the experimentation. Pots were regularly irrigated from

May to October 2017. Contamination of soils was evaluated before (April 2017) and after irrigation (November 2017). The post-irrigation substrate was collected in a depth range of 0-15 cm; for each TW and pot, three different substrate cores were sampled. For each TW irrigation line, the collected substrate portions were mixed, homogenized, freeze-dried and stored at -20 °C until analysis.

After the harvest, strawberries were washed, dried and frozen at -4°C, until analysis.

2.3. Reagents and solutions

The complete list of 13 PAHs and 14 PCBs analysed in this study is reported in Table 1. PAHs standards were from Sigma Aldrich-Merck (Darmstadt, Germany) and contain the priority compounds listed by EPA. PCBs were from LGC Standards (Milan, Italy), and were chosen according to the results of the main environmental monitoring campaigns carried out in Italy. The PCB congeners chosen represent chlorine (from mono to epta) substitution classes and include dioxin-like compounds (marked with an asterisk in Table 1).

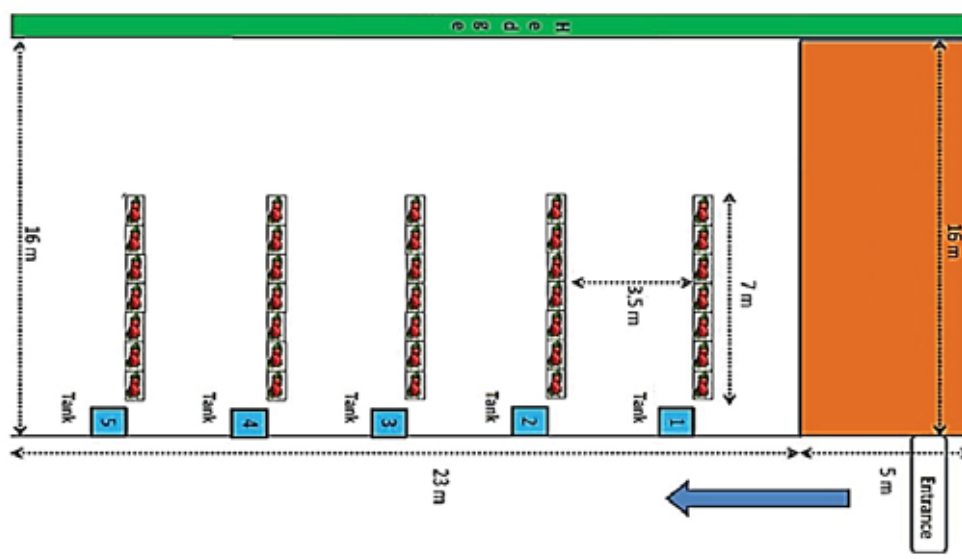


Fig. 1. The experimental site for strawberry cultivation

Table 1. Target PAHs and PCBs studied in waters, soils and strawberries, together with labelled compounds used as internal standards (IS) and surrogates

PAH ANALYTE	PCB CONGENER	SURROGATES AND IS
Acenaphthylene (AcPY)	3,3'-dichlorobiphenyl (PCB 11)	13BaA
Fluorene (Flu)	4,4'-dichlorobiphenyl (PCB 15)	13Chr
Phenanthrene (Phe)	2,4,4'-trichlorobiphenyl (PCB 28)	13BbFl
Anthracene (Ant)	2,2',5,5'-tetrachlorobiphenyl (PCB 52)	13BkFl
Pyrene (Pyr)	3,4,4',5-tetrachlorobiphenyl (PCB 81*)	13BaP
Benzo[a]anthracene (BaA)	2,2',4,5,5'-pentachlorobiphenyl (PCB 101)	13Ind
Chrysene (Chr)	2,3',4,4',5-pentachlorobiphenyl (PCB 118*)	13DBA
Benzo[b]fluoranthene (BbFL)	2',3,4,4',5-pentachlorobiphenyl (PCB 123*)	13BP
Benzo[k]fluoranthene (BkFL)	2,2',3,4,4',5-hexachlorobiphenyl (PCB 138)	¹³ C PCB 28
Benzo[a]pyrene (BaP)	2,2',4,4',5,5'-hexachlorobiphenyl (PCB 153)	¹³ C PCB 52
Indeno[1,2,3-cd]pyrene (Ind)	2,3',4,4',5,5'-hexachlorobiphenyl (PCB 167*)	¹³ C PCB 118
Dibenzo[a,h]anthracene (DBA)	3,3',4,4',5,5'-hexachlorobiphenyl (PCB 169)	¹³ C PCB 153
Benzo[ghi]perylene (BP)	2,2',3,4,4',5,5'-heptachlorobiphenyl (PCB 180)	¹³ C PCB 180
	2,3,3',4,4',5,5'-heptachlorobiphenyl (PCB 189)	

(*): dioxin like PCBs

Labelled isotope compounds from both categories, (Wellington Laboratories, Ontario, Canada), were used as internal standards and surrogates in order to obtain calibration curves and extraction recoveries, respectively. From chromium analysis, Cr(VI) standard solution was prepared from K_2CrO_4 (Alfa Aesar, Haverhill, USA). Reagent grade dichloromethane, 2-propanol, cyclohexane, acetone, Na_3PO_4 were from Sigma Aldrich-Merck (Darmstadt, Germany). High-purity water (18.2 M Ω cm resistivity at 25 °C), produced by an Elix-Milli Q Academic system (Millipore-Merck, Vimodrone, MI, Italy) was used.

2.4. Instrumentation

For PAHs and PCBs analysis, a gas chromatographic-mass spectrometric (GC-MS) method was optimized, moving from both EPA 8275A procedure (EPA, 1996) and Zhang et al. results (Zhang et al., 2007). A (5%-Phenyl)-methylpolysiloxane column (HP 5ms, 30 m x 0.25 mm x 25 μ m, Agilent) was used. Analysis was performed in Single Ion Monitoring (SIM) mode, selecting for each analyte its proper m/z ratio (m/z ratio available upon request). 2 μ L of each sample were injected using the Pulsed Splitless mode (pressure at 40 psi for 2.5 minutes). The oven ramp was set as follows: starting temperature: 40°C, hold for 2 min; ramp to 176 °C, 12 °C/min rate; ramp to 196°C, 5 °C/min rate, hold for 3 mins; ramp to 224°C, 12 °C/min rate; ramp to 244 °C, 5°C/min rate, hold for 3 min; ramp to 270 °C, 7°C/min rate, hold for 3 min; final ramp to 300 °C, 5°C/min, hold for 10 min to completely clean and restore the GC column. The total run time for the complete separation of PAHs and PCBs is 52 min. The determination of Cr(VI) was performed by ion chromatography (IC) with post-column derivatization and spectrophotometric detection, as developed by our research group (Bruzzoniti et al., 2017).

2.5. Extraction Procedures

Due to the comprehensive aim of the presented study, several matrices were analyzed (water, soil,

strawberries). For each matrix, extraction procedures were appositively optimized, as summarized hereafter.

Water samples. PAHs and PCBs were extracted from TWs and FW by solid-phase extraction (SPE) on a polymeric reversed-phase cartridge (STRATA XL, Phenomenex, Torrance, USA), as schematized in Fig. 2, and injected for GC-MS analysis. Cr(VI) was analysed by direct injection IC of filtered samples (nylon filters, 0.45 μ m).

Soil samples. PAHs and PCBs were extracted by microwave assisted extraction (MAE). The procedure, developed for PCB extraction (Bruzzoniti et al., 2012), was here tested for the extraction of PAHs as well. Briefly, 0.4 g of soil, previously sieved at 2 mm, were put in a disposable Pyrex vessel with 5 ml of a 3:2 acetone-cyclohexane solution. After a temperature ramp (0-10 min up to 130°C, 10-15 min T=130°C, 15-25 min decrease to 60°C) in microwave oven, the vessel was centrifuged at 3850 rpm for 5 min, acetone was evaporated by heating at 60°C and the solution made up at 5 mL with cyclohexane. Finally, 2 mL of H_2SO_4 were added, as a clean-up step, and 1 mL of supernatant was withdrawn and injected for GC/MS analysis.

Cr(VI) was extracted using a Na_3PO_4 solution (Bruzzoniti et al., 2017). In detail, two aliquots of the same soil were extracted at the same time. The first aliquot (0.5 g) was extracted with 50 mL Na_3PO_4 (10 minutes, 100°C), filtered with a 0.45- μ m nylon syringe filter and injected in IC system. To evaluate the extraction yield, the second aliquot (0.5g) was spiked with Cr(VI) to obtain 1 μ g/L in the final extract and then extracted as previously described.

Strawberry samples. PAHs and PCBs were extracted using the QuEChERS approach (Bruzzoniti et al., 2014, De Carlo et al., 2015). Briefly, 5 g of strawberries were put in a vial containing 10 mL dichloromethane, 400 mg $MgSO_4$ and 1 g NaCl. The tube was vigorously shaken and centrifuged at 1507 xg for 5 minutes. The supernatant was then transferred for clean-up in a new vial containing 50 mg Primary and Secondary Amine (PSA) sorbent and 150 mg $MgSO_4$. Again, the tube was shaken and centrifuged (7871 xg, 10 minutes). 1 mL of the supernatant was directly analyzed by GC-MS.

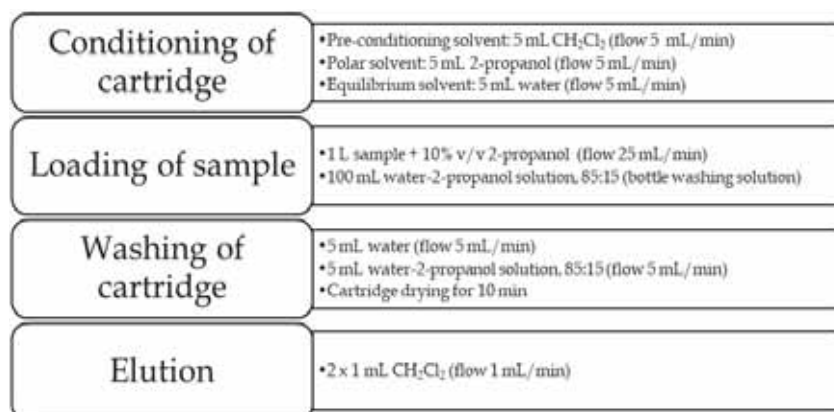


Fig. 2. Protocol for the SPE extraction of PAHs and PCBs

On the opposite, the determination of Cr(VI) on strawberry was not successfully achieved by transferring the soil extraction procedure to crops, due to a residual colour interference of the matrix. Hence, the determination was performed measuring total Cr by means of an acid MAE digestion (HNO₃/H₂O₂ mixture) followed by ICP-MS detection.

3. Results and discussion

3.1. Performances of analytical methods

To verify that the methods developed fulfil limits imposed by regulations (where present), extraction yields, methods detection (MDL) and quantitation limits (MQL) were tested.

For PAHs and PCBs extraction recoveries for each matrix were in the following ranges, waters: 60% (13BP)- 99% (13PCB52); soils: 45% (13BP) – 95% (13PCB52); strawberries: 62% (13Chr) – 102%

(13PCB118). For Cr(VI), recoveries were 85% (waters) and 30% (soils).

For PAHs and PCBs, MQLs for each matrix were in the following ranges, waters: 1.3 (Phe)- 11.0 ng/L (PCB169); soils: 3 (PCB180) - 10 µg/Kg (BbFl); strawberries: 1.0 (Phe) - 10.9 µg/Kg (PCB169). For Cr(VI), MQLs were 0.15 µg/L (waters) and 0.018 mg/kg (soils). These performances fully satisfy limits fixed by Italian or European regulations on maximum admitted concentration of pollutants in i) treated waters to be reused for irrigation (D. Lgs 185/2003); ii) private and commercial soils (D.Lgs 152/2006), since limits are not present for soils intended for agricultural aims; iii) fruit crops (CE Regulation 1881, 2006). These limits will be fully discussed in the subsequent sections.

3.2. Chemical characterization of TWs

PAHs and PCBs. Data for PAHs found in TWs and FWs are summarized in Table 2.

Table 2. Concentration (ng/L) and standard deviations in brackets of PAHs in TWs and FW, for each sampling period (see Materials and Methods section). Average data for the whole campaign is also shown

	<i>AcPy</i>	<i>Flu</i>	<i>Phe</i>	<i>Ant</i>	<i>Pyr</i>	<i>Chr</i>	<i>BaA</i>
FWA	1.42 (0.01)	12.54 (4.6)	66.23 (25)	10.73 (2.7)	2.91 (0.96)	1.30 (0.14)	0.79 (0.04)
FWB	3.47 (0.02)	50.68 (35)	33.9 (0.08)	6.53 (1.8)	2.00 (2.0)	25.01 (28)	10.8 (8.4)
FWC	16.34 (2.5)	38.90 (16)	44.3 (20.6)	19.99 (4.8)	12.87 (4.3)	6.49 (2.4)	9.2 (2.6)
FWD	24.49 (3.8)	98.08 (30)	633.7 (180)	45.03 (7.6)	9.31 (1.2)	1.00 (0.12)	0.86 (0.32)
FWE	11.3 (0.04)	38.21 (3)	201.3 (29)	24.18 (4)	11.26 (2.4)	2.54 (0.3)	2.65 (0.3)
FWF	0.49 (0.09)	7.21 (0.30)	4.39 (0.32)	nd	0.50 (0.15)	nd	nd
FWG	6.18 (0.15)	8.0 (1.2)	31.3 (0.89)	nd	1.63 (0.02)	nd	nd
AVERAGE	20.35 (1.45)	36.23 (7.02)	145.02 (26.45)	15.20 (1.48)	5.78 (0.79)	5.19 (4.71)	3.47 (2.62)
TW1A	57.79 (5.2)	157.18 (4.9)	858.93 (37)	79.72 (2.6)	39.82 (1.8)	1.75 (0.44)	17.6 (1.8)
TW1B	17.70 (0.5)	131.56 (39)	477.76 (142)	37.57 (3.4)	21.21 (3.4)	3.97 (0.4)	3.45 (0.52)
TW1C	10.79 (5.0)	101.13 (38)	391.89 (180)	17.86 (6.8)	15.20 (6.9)	1.39 (0.4)	1.23 (0.8)
TW1D	21.6 (7.4)	177 (28)	1160 (168)	83.2 (12.8)	15.70 (3.0)	1.29 (0.06)	nd
TW1E	7.12 (0.6)	24.19 (1.6)	103.06 (3.2)	8.48 (0.4)	2.57 (0.4)	nd	nd
TW1F	2.52 (0.19)	13.92 (0.56)	7.19 (1.13)	nd	1.80 (0.5)	nd	nd
TW1G	1.01 (0.03)	15.41 (2.35)	5.73 (0.32)	nd	nd	nd	nd
AVERAGE	16.93 (1.48)	88.53 (8.78)	428.85 (40.91)	32.17 (2.14)	13.48 (1.19)	1.2 (0.10)	3.18 (2.08)
TW2A	20.60	76.54	340.64	54.95	32.24	0.74	1.78
TW2B	1.18	4.97	22.45	7.39	5.0	1.83	4.1
TW2C	37.94 (4.1)	173.26 (14.8)	739.01 (94)	46.10 (6.7)	40.58 (3.0)	1.71 (0.3)	1.36 (1.08)
TW2D	31.99 (4.6)	130.85 (24)	477.80 (165)	65.17 (14.2)	15.01 (3.2)	2.34 (0.8)	1.13 (0.26)
TW2E	9.83 (0.5)	27.27 (1)	145.64 (5.6)	17.54 (2.4)	6.09 (1.0)	1.51 (0.3)	1.80 (0.56)
TW2F	1.03 (0.28)	5.04 (0.13)	2.40 (0.92)	nd	nd	nd	nd
TW2G	nd	2.16 (0.46)	1.44 (0.15)	nd	nd	nd	nd
AVERAGE	13.38 (0.82)	59.69 (5.85)	246.69 (27.95)	27.19 (2.48)	14.33 (0.76)	1.10 (0.13)	1.45 (0.5)
TW3A	0.76 (0.02)	2.90 (0.24)	13.73 (1.4)	2.10 (0.02)	0.55 (0.1)	nd	nd
TW3B	3.24	20.66	51.38	2.19	0.45	nd	nd
TW3C	24.07 (2.8)	67.11 (8.8)	328.11 (43)	33.62 (1.8)	9.55 (0.3)	nd	nd
TW3D	10.2 (0.48)	34.65 (2.1)	181.27 (21)	13.46 (1.8)	3.36 (0.6)	nd	nd
TW3E	12.95 (2.4)	39.15 (9.8)	201.92 (54)	16.15 (4.6)	4.06 (1.0)	nd	nd
TW3F	1.53 (1.05)	3.89 (2.24)	13.3 (1.23)	nd	1.44 (0.09)	nd	nd
TW3G	nd	3.04 (0.32)	nd	nd	nd	nd	nd
AVERAGE	7.54 (0.63)	24.74 (1.99)	112.61 (10.48)	9.65 (0.75)	2.77 (0.17)	nd	nd
TW4A	65.64 (17)	169.36 (40)	926.91 (224)	93.07 (17.7)	45.24	1.29 (0.2)	24.2 (5.2)
TW4B	24.4 (0.24)	92.83 (7.8)	468.21 (24)	35.24 (0.92)	13.50	1.14 (0.1)	nd
TW4C	34.85 (0.6)	106.35 (2.0)	594.65 (7.4)	50.38 (2.1)	18.94	1.41 (0.16)	0.68 (0.14)
TW4D	6.76 (3.6)	23.93 (10.2)	112.84 (51)	69.13 (4.6)	1.40	nd	nd
TW4E	9.83 (0.34)	39.75 (10.2)	251.75 (8.4)	19.19 (0.7)	10.00	0.50 (0.06)	0.22 (0.04)
TW4F	1.05 (0.22)	9.19 (0.07)	nd	nd	nd	nd	nd
TW4G	nd	nd	nd	nd	nd	nd	nd
AVERAGE	20.36 (2.48)	63.92 (6.27)	335.86 (35.08)	38.14(2.63)	12.73	0.45 (0.03)	3.58 (0.74)

It is interesting to observe how PAHs with higher aromatic ring number (BbFL, BkFl, BaP, DBA, BP and Ind) were never detected in TWs; moreover, a variability of PAH concentrations among the different months is also present for all the analytes. The absence of several PAHs in TWs demonstrated the efficacy of the WWTPs in the removal of PAHs from wastewaters. In fact, previous studies demonstrated that high molecular weight compounds are better removed during the treatment, and probably transferred to sludges (Yan et al., 2016), in respect to low molecular weight ones, which still remain in the final effluent. Results are also in good agreement with previously obtained studies on TWs (Mezzanotte et al., 2016, Yan et al., 2016), even if concentrations of Flu and Chr here detected are higher than data collected in the above-mentioned works.

Some concentration trends could be highlighted. PAH concentrations in all TWs reached a maximum in the summer months (July and August), for decreasing in September and October. This trend could be explained by summer storms phenomena affecting Mediterranean countries, included Italy, where concentration level of PAHs in wastewater could greatly increase up to 10–100 fold (Blanchard et al., 2001). Another trend is highlighted for AcPY, Flu, Phe and Ant concentrations which are higher in TW1 and TW4. This behaviour can be correlated with the Total Suspended Solid (TSS) parameter that for TW1 and TW4 was 1.5 order of magnitude higher than the other TWs (21.1 mg/L and 19.8 mg/L against an average of 1.2 mg/L). These PAHs with low aromatic ring number are known to slightly interact with particulates (Li et al., 2010): therefore, for high TSS contents in the water, these compounds are expected to be present in higher concentrations (Sangster, 1989).

As previously discussed, admitted concentrations of PAHs in treated waters reused for irrigation are included in the D.M. 185/2003, that, however, regulates only the presence of Benzo[a]pyrene, to a fixed limit of 10 ng/L (Italian Republic, 2003). In this regard, it is important to highlight the absence of benzo[a]pyrene in the four TWs and in the control. To follow a precautionary approach, data in Table 2 were also compared to the more strict EU Directive on waters intended for human consumption (98/83/CE), which regulates not only benzo[a]pyrene (10 ng/L), but also benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(ghi)perylene and Indeno[1,2,3-cd]pyrene, whose sum should not exceed 100 ng/L (European Council, 1998). Again, none of the above-mentioned compounds was detected in TWs and FW.

PCB concentrations (data not shown but available upon request) for all the samples ranged from below detection limits (1-2 ng/L) to about 200 ng/L (PCB52) with concentrations in FW lower in respect to TWs. PCBs are not included in the D.M. 185/2003 and, therefore, no comparison with

maximum admitted concentrations could be discussed. As for PAHs, the removal of PCBs in WWTPs is strongly dependent on their sorptive behaviours, affected by their octanol-water partition coefficients. However, contrary to what evidenced for PAHs, no seasonal trend could be observed. It is important to underline that most PCB presence is expressed by PCB11 and PCB52; in particular, the predominance of the PCB11 species (which however is not a dioxin-like PCB) is in good agreement with data on WWTPs effluents, (Balasubramani et al., 2014, Yao et al., 2014). Its presence can be explained with the wastewater treatment processes, as a result of dechlorinating of heavier PCBs into lighter PCBs (Balasubramani et al., 2014). Furthermore, the presence of PCB11 can be ascribed also to the peculiarity of the influent (domestic/textile waters). In fact, PCB11 is known to be produced in the manufacture of diarylide yellow pigments, used in textile industries (Grossman, 2013). Pigments are a point source of other dioxin-like congeners, such PCB167, thus justifying its detection even if sporadic and at trace levels, in the October sampling.

Chromium(VI). Hexavalent chromium concentrations found in irrigating waters are summarized in Fig. 3. Samples withdrawn in September and October (F and G) are not presented in figure, due to fail of storage conditions during transportation to our laboratory. The colour of some TW samples interfered in Cr(VI) determination by IC at concentrations lower than 1 µg/L, hence for these samples, 1 µg/L was assigned as a precautionary value. The presence of Cr(VI) in the TWs effluents is not surprising, since it is well known how traditional removal treatments could be affected by incomplete removal of heavy metals (Barakat, 2011), especially in textile districts. It should be remarked, however, that for all the tested samples, Cr(VI) concentration is under the limits set by Italian decree 185/2003 on the reuse of treated waters for irrigation (5 µg/L).

TW3 sample exhibits a higher Cr(VI) contamination than TW1, although it derives from a further refinement of TW1 (see Material and Methods section). This behaviour could be ascribed to the treatment process that relies on dilution of TW1 with an adjacent river. Indeed, it is not unusual to detect hexavalent chromium in natural waters basins (Hemmatkhan et al., 2009, Vasilatos et al., 2008).

3.3. Soil chemical characterization

PAHs and PCBs. The presence of PAHs, PCBs and Cr(VI) was evidenced in TW samples. Therefore, the analysis of soils irrigated with TWs is fully justified to evaluate whether a contamination took place or not from waters.

Soils used for the cultivation of strawberry plants were analysed before the irrigation period started (to obtain a “zero level” characterization) and at its end (Fig. 4).

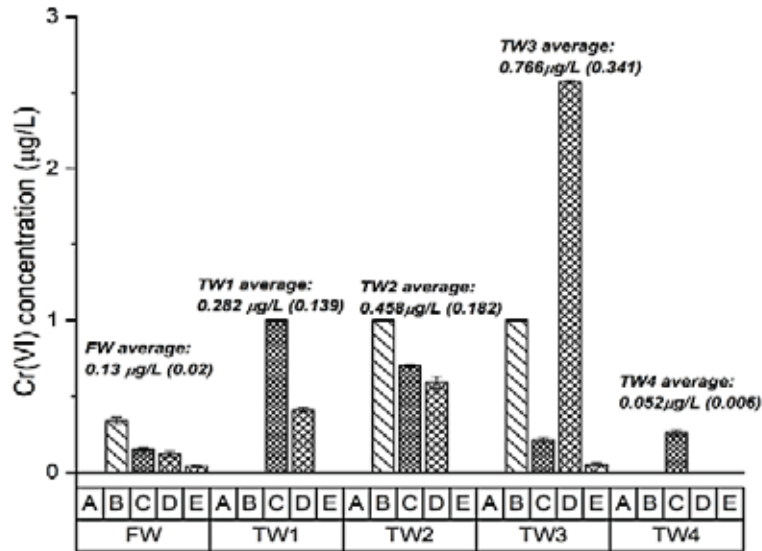
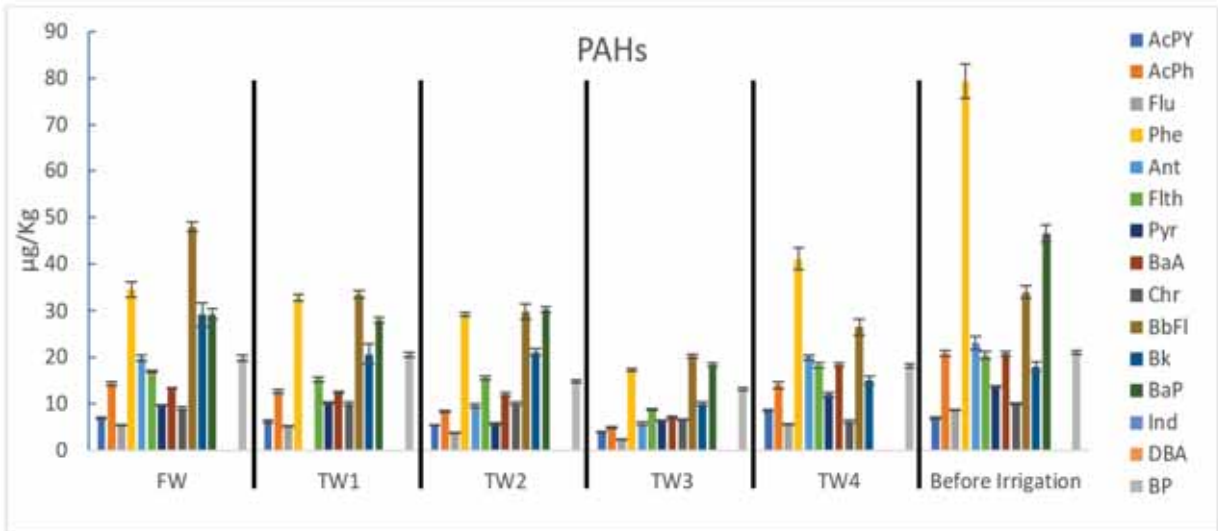
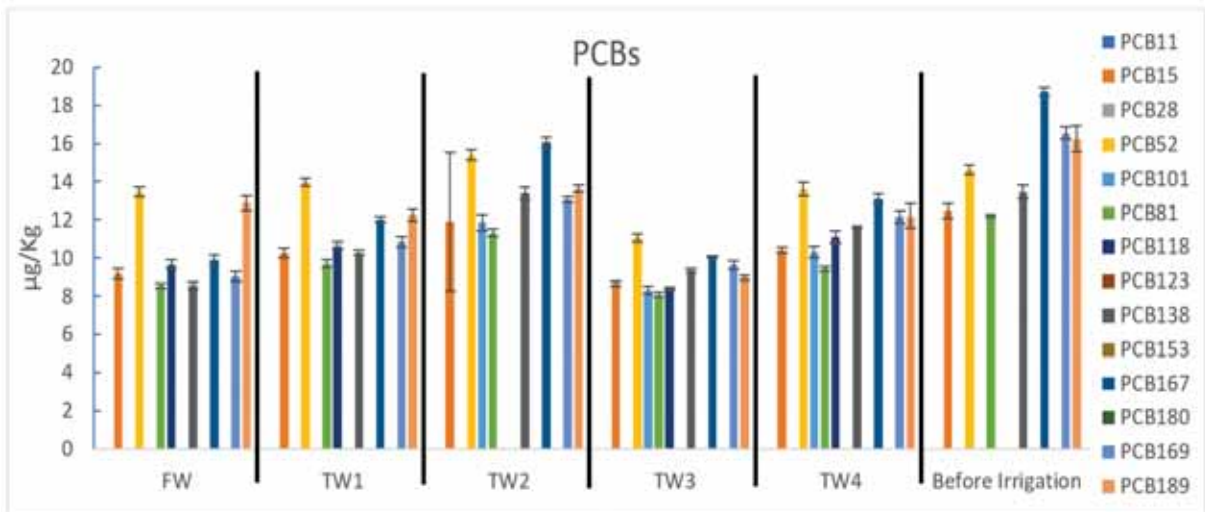


Fig. 3. Concentration of Cr(VI) in TWs and FW, for each sampling period (A-E); average value for the whole campaign is also indicated, together with standard deviation in brackets



(a)



(b)

Fig. 4. Concentration (µg/kg) of (a) PAHs and (b) PCBs in soils, before and after the irrigation period

Both for PAHs and for PCBs, concentration levels before and after the irrigation period are very similar. The total amount of PAHs in the pre-irrigated soils is 320 µg/Kg (data range: 6.85 µg/Kg AcPY - 80 µg/Kg Phe). Toxic equivalency (TEQ) value was also evaluated as follows (Eq. 1):

$$TEQ = \sum (TEF_{PAHi} \cdot [PAH_i]) \quad (1)$$

where: $[PAH_i]$ is the concentration of the i -th PAH congener and TEF_{PAHi} is the toxicity equivalent factor of the i -PAH congener (Jimenez et al., 2014). TEQ value is 54.5 µg/Kg. Based on literature data, the sum of PAHs matches values observed for agricultural soil (Zheng et al., 2014); however, TEQ value suggests that soil used for strawberry cultivation exhibits toxicity properties more similar to urban soils, due to the high contribution of BaP in the TEQ calculation (85%) (Soukarieh et al., 2018).

The total amount of PAHs in soils irrigated with TWs ranged from 125 µg/Kg (TW3) to 320 µg/Kg (FW). These concentrations are more than one order of magnitude lower than the limit imposed by Italian regulations (D.Lgs 152/2006), which is set at 10 mg/kg for the sum of specific PAHs (BaA, BaP, BbFl, BkFl, Chr, DBA, together with Benzo(g,h,i)terylene, Dibenzo(a,e)-, Dibenzo(a,l)-, Dibenzo(a,i)pyrene, not investigated in this work) for soils in public-green, private and residential areas, and at 100 mg/kg for soils intended for commercial aims.

According to the data obtained, a slightly higher contamination of soils by PAHs before irrigation was observed in respect to the end of the experimental trial. This behaviour could be explained by the contribution of perlite (present in the soil substrate) to partially retain PAHs, as demonstrated by other authors (Bjorklund and Li, 2015, Turan et al., 2009). Additionally, contribution of microbial PAH degradation in soil should not be excluded (Cardak et al., 2007). It should be remarked that, even if possible retention of PAHs by the strawberry plant could occur, the evaluation of the uptake by different parts of the plant (excluded the fruit) is out of the scope of this work.

Also, for PCBs, concentration in soils before and after irrigation indicated the absence of impact from TWs. Their presence in the original soil substrate can derive from atmospheric deposition (Glüge et al., 2016). PCB concentrations reaching about 60 µg/kg were observed in rural soils (Meggo and Schnoor, 2013), confirming the ubiquitous presence of these compounds. Despite the fact that, to the best of our knowledge, PCBs in soil are not regulated at a EU level, in Italy concentration limits for PCBs are set by the D.Lgs 152/2006, at 0.06 mg/kg and 5 mg/kg for soil intended for private or commercial aims, respectively.

Cr(VI). As done for organic compounds, the presence of hexavalent chromium was monitored both before and after irrigation. Results (Fig. 5), showed

that only a marginal increment in Cr(VI) concentration occurred after irrigation for all samples. The highest increment observed for soil irrigated with TW3 is in agreement with the higher Cr(VI) content observed for this treated water (see previous paragraph).

Data obtained fully satisfied the limits declared by the Italian decree D.Lgs 152/2006 (2 mg/kg) and are in good agreement with previous studies on urban soils (Jankiewicz and Ptaszynski, 2005).

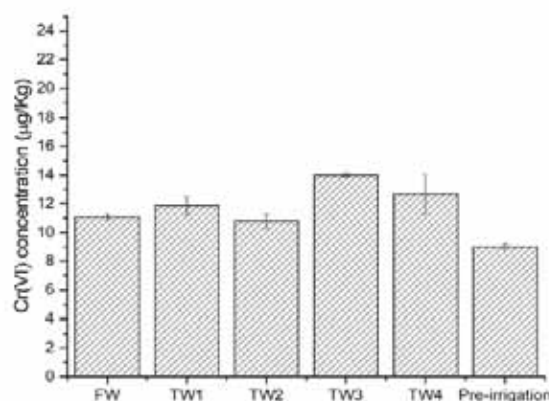


Fig. 5. Concentration of Cr(VI) in soils before and after irrigation by TWs and FW

3.4. Strawberries

3.4.1. PAHs and PCBs

Strawberry crop grown up under irrigation by TWs represents the final part of the agricultural chain. Several studies demonstrate that PAHs and PCBs (Lovett et al., 1997, Paris et al., 2018), as well as metal ions (Khan et al., 2015) could be detected in fruit and vegetables depending on the contamination of crop area.

To what concern PAHs and PCBs, none of the above-mentioned molecules was detected at a quantifiable level, apart from BaA in strawberries irrigated with TW1. In fact, BaA was observed at 1.14 µg/Kg (MQL 1.17 µg/Kg); this value agrees with PAHs found in fruit and vegetable cultivated in rural areas (Camargo and Toledo, 2003). EU regulation on food contamination (CE) N. 1881/2006 (European Council, 2006) does not set a limit for PAHs and PCBs in fruits. BaP, which however was not detected in strawberries, is the only PAH for which a limit ranging from 2 to 10 µg/kg is set according to the type of food considered (infant food excluded).

For strawberry irrigated with TW1, TEQ is 0.114 µg/kg. This value is comparable to those estimated for other Italian food products, such as cheese, bread and eggs (Lodovici et al., 1995). If a precautionary intake of strawberry is fixed to 100 g/day for an adult person, the Bench Mark Dose Lower Confidence Limit of BaP (100 µg BaP/kg bw/day, concentration producing a predetermined change in the response rate of an adverse effect, established as carcinogenicity in mice orally dosed with a mixture of

representative genotoxic and carcinogenic PAH present in food) is fully respected (FSANZ, 2005).

3.4.2. Chromium

The IC analysis of Cr(VI) in strawberries suffered for residual colour interference. In addition, even after the direct spike of Cr(VI) in the fruit at concentrations as high as 500 µg/L, the method was not able to quantify Cr(VI), presumably due to the high content of anti-oxidant species in strawberries (Doumett et al., 2011) which reduced Cr(VI) to Cr(III). Hence, total chromium determination was performed (Fig. 6).

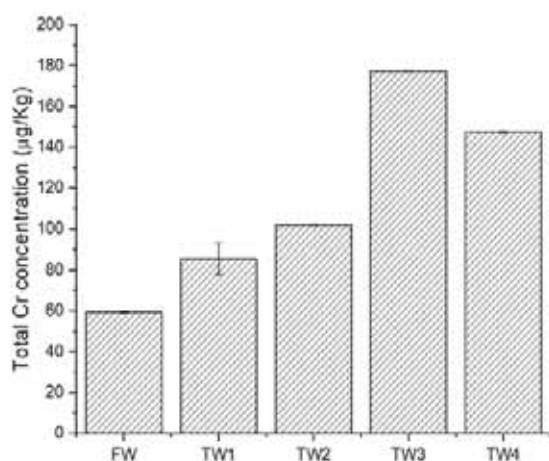


Fig. 6. Total Cr determination in strawberries irrigated with TWs and FW

Differently from what observed for soils, strawberries irrigated with TWs have a higher Cr content, in respect to the control, thus suggesting the transfer of the metal during irrigation. The highest concentration of Cr in strawberries grown up under irrigation with TW3 are in agreement with the highest concentration of Cr(VI) detected in the TW3 sample.

Cr concentrations are from two to five times higher than those detected in other strawberries, as presented in the EFSA Scientific Opinion of 2014, EFSA Panel on Contaminants in the Food Chain (CONTAM, 2014). Nevertheless, assuming the same premises previously presented for PAHs (100 g/die consumption for an adult over 50 kg bw), Cr content in crop irrigated with TW3 results in a daily intake of 0.4 µg/kg bw, which summed with the estimated daily intake expressed as Cr(III) (0.6-5.9 µg/kg bw) does not exceed the TDI of 300 µg/kg b.w-EFSA Panel on Contaminants in the Food Chain (CONTAM, 2014)

4. Conclusions

For the first time, the possible chemical contamination by organic (PAHs, PCBs) and inorganic compounds (chromium) in the agricultural chain of strawberries irrigated with different kind of reclaimed waters was assessed. All the treatments considered are capable of guaranteeing the levels set

for PAHs and Cr(VI) by Italian legislation for wastewater reuse for irrigation.

Although PAHs, PCBs were detected in waters, their presence was not observed in strawberries, except for BaA at amounts comparable with quantitation limit of the method. Irrigation with these TWs does not impact the quality of the soil that exhibits similar PAHs and PCBs content before and after irrigation. On the contrary, chromium content in one of the strawberry crops (which however does not represent a risk for consumer) presumably derives from the original residual contamination of treated water.

The results observed within this study seems in agreement with a negligible impact of lipophilic compounds and a possible transfer of inorganic water-soluble compounds (metals) in fruits of high-water content such as strawberries.

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INVESTIGATING CIRCULAR ECONOMY URBAN PRACTICES IN CENTOCELLE, ROME DISTRICT

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Abstract

The embedment of circular economy principles in cities is an opportunity to reduce the urban waste production and resource consumption towards closed-loop systems. In particular, the adoption of circular economy strategies as regeneration, optimization, sharing and loops can overcome waste and inefficiencies of linear economy. This paper presents some circular economy practices bringing an integration of circular strategies at urban scale. This work was funded by the Research into Electrical Systems Italian National Programme aimed at implementing a Smart District Models. The research project has been carried out in an Italian demonstrator quarter: Centocelle located in the city of Rome. Thanks to the smart solutions employment, a collaborative process has been established involving citizen. As a result, several circular economy urban practices have been identified supporting the smart community growth. The combination of smart community experience and circular economy principles application has resulted in several mutual advantages. In particular, the identified circular practices, as community gardens, co-workings, local and km0 production systems, recycling centres and other experiences of waste management, have achieved an urban transformation in terms of regeneration, sharing economy experiences, resource optimization, closed-loop systems. Therefore, circular economy practices have positively and actively influenced the urban community supporting the transition towards circular economy models.

Key words: circular economy, smart district, smart community, urban regeneration, waste prevention

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1. Introduction

Approximately the 70% of the European Union population live in urban areas (EC, 2016). The share of population in cities continues to grow, and it is likely to reach more than 80% by 2050 (World Bank, 2019). Population density, urbanization process and economic activities intensify both environmental and

social issues (ISWA, 2018). Cities are therefore facing correlated and complex challenges in terms of climate change, environmental pollution, waste production, economic development, occupation, demography, migration etc. (World Bank, 2010). But urban areas are also placing of innovative transformation, incubators of new concepts and ideas, steering both global and local economy and furthermore creating

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opportunity for job and wellbeing. Cities are so dynamic places where change can happen on a larger scale and at a more rapid pace, and they are also privileged places.

This paper focuses on urban transition towards Circular Economy (CE). Actually, there is the need to prioritize a systematic transition from the linear paradigm of production and consumption towards a circular economy model (EMF, 2017). According to EMF (2019), CE in cities should be based on the following three principles:

1. regenerating natural system, preserving and enhancing natural capital;
2. keeping products and materials in use and reducing virgin material pressures interactions;
3. designing out waste and pollution.

CE at urban level could act as a restorative and regenerative process. In particular, the first principle will encourage conditions for regeneration, such as soil for renewable resources. The second principle consists of keeping materials and products ‘circulating’ in the techno-sphere (McDonough and Braungart, 2009), through the efficient use, reuse, repair and sharing of goods (Geissdoerfer et al., 2017; Korhonen et al., 2018). Finally, the third principle is aimed at eliminating waste and reducing carbon emissions and pollution. In fact, CE promotes waste prevention one of the main strategic objectives of European waste management policy (EC Communication, 2015). According to the European Directive 2018/851 (EC Directive, 2018), which updated and replaced the original 2008/98/CE Directive (EC Directive, 2008), a waste hierarchy in the legislation and policies around waste management has been established and waste prevention has a priority position. In addition, a specific European waste prevention strategy is devoted to reduce plastic waste. Other CE consequences are related to climate change. In particular, it also renown that CE facilitates cutting down on GHG emissions, by reducing the amount of energy needed by production processes (Enkvist and Klevnas, 2018) and transforming primary raw materials into usable products (Iacovidou et al., 2017). Therefore, the application of CE principles in cities will allow fostering the challenge of transforming cities into circular aggregates. The CE principles adoption can be supported by the ReSOLVE model introduced by Ellen MacArthur Foundation (EMF, 2015) and consisting of the following six strategies: Regeneration, Sharing, Optimization, Loops, Virtualization and Exchange. In particular, regeneration concerns a broad set of actions that maintain and enhance the earth’s biocapacity. Sharing strategy is aimed at getting a full use out of goods eliminating waste and duplication. Optimization is related to remove waste and inefficiencies in the life cycle phases of goods, such as manufacture, distribution, use, etc. Loop strategy is intended to process resources in order to close the cycle and put them back into the economy, rather than lost through landfill. Virtualization promotes a dematerialization of goods and products. Finally,

exchange is the process of swapping in new technologies, upgrading or replacing older ways of doing things.

This study aims to investigate the application of CE strategies in a Smart Community through the collaboration of citizens and other urban stakeholders. In particular, the experimental activities have been carried out in the framework of an Italian national strategic research project funded by the Fund for Research into Electrical Systems (RSE project, 2019). One of the RSE project final objective (Project D.7 PAR 2015; Project D.6 PAR 2016-2017) is the development Smart Urban District integrated model and the implementation was carried out in a demonstrator district of the city of Rome, called Centocelle.

2. Experimenting CE in Smart Community: the Centocelle Case study

2.1. The role of Smart Community

According to Granelli (2012) a Smart City represents "a new kind of common good, a large technological and immaterial infrastructure that makes people and objects connect with each other, in a way that it integrates information and generates intelligence, it fosters inclusion and improves everyday life". In a smart city social, economic and environmental sustainability are closely related. The Italian Digital Agenda under article 20 of 221/2012 Act (Italian Law, 2012) defines a smart community as “a community built to form a connective structure, (open, aware and focused), and, at the same time, an adaptive structure, capable of generating data and knowledge and making one's behavior evolve”. Furthermore, thanks to its local dimension, sustainability practices are more applicable in smart communities, whereas cannot be guaranteed in non-smart communities (Bifulco et al., 2016). Broader understanding of smart cities also highlights the use of modern technologies but sees them more as an enabler for better quality of life and decreased environmental impacts (IEEE, 2014). As an example, Marsal-Llacuna et al. (2015) suggest that smart city initiatives aim, by using data and information technologies, to “provide more efficient services to citizens, to monitor and optimize existing infrastructure, to increase collaboration amongst different economic actors and to encourage innovative business models in both private and public sectors”. Another body of literature highlights – in addition to new technologies – the role of human capital in developing smart cities with improved economic, social and environmental sustainability (Ahvenniemi et al., 2017). This more holistic understanding suggests that smart cities allow the conjunction of governance, society and technology in order to bring the smartization process into urban economy, environment, mobility, lifestyle and population (IEEE, 2014).

This paper analyses some experimental activities contributing to a local Smart Community

establishment in Centocelle. In a Smart Community, citizens are able to act for the co-governance of the neighbourhood and to actively participate in the collective life (De Nictolis et al., 2017). In fact, several factors have an influence on pro-environmental behaviour (Buenrostro et al., 2014). In particular, the importance to build attitudes of belonging and co-responsibility has been highlighted (Hasan and Idris, 2014). Moreover, collaborative models offer opportunities to overcome linear approach (Sposato et al., 2017). In the RSE project, a co-creation process has been mainly developed related to innovative and smart solutions, based on Social Urban Network (SUN). In particular, the SUN consists of a coordinated set of interventions that has been developed both on the ICT technologies (social networks, web portal) and on the urban scene (interactive installation, local initiatives). This paper focuses on local initiatives based on the application of circular economy principles implemented in Centocelle through a strong citizen engagement.

2.2. CE practices in Centocelle District

Adopting a user-centric innovative approach based on Urban Living Labs (ULL) approach (McCormick and Hartmann, 2017), a collaborative process where citizens were actively engaged has been carried out within the Centocelle urban district (Cappellaro et al., 2019). Thanks to interactive workshops and interviews, 14 CE practices have been identified. In particular, a first screening on CE practices has been realized during the ULL activities consisted of working groups and co-design activities coordinated by researchers. Further data were also gathered thanks to a questionnaire and personal interviews to CE practices reference person. In Table 1 a description of the identified CE practices is provided. This study aims to analyze CE practices identified in Centocelle district. Details related to each CE practices are reported in the paragraphs below.

Table 1. Circular economy practices in Centocelle District

CE practices	Centocelle local initiative
Community green garden	“Giorgio De Chirico” Park
	“Villa Flaviana” community garden
	“Centocelle” Park regeneration
	“Tor Sapienza” community garden
	“100 e a capo” community garden
	“Centocelle” community garden
Co-working	“ZappataRomana” Urban map of community gardens in Rome
	Coworking “L’Alveare” Coworking “FusoLab”
Zero-kilometre practices	Km0 restaurant “DOL – Di Origine Laziale”
	Local purchasing grup “GAS L’Alveare”
Water house practices	Water House “ACEA”
Smart waste management models	Second-hand market “Capannelle”
	“AMA” recycling centres

2.3. Community green gardens

Several CE practices identified in Centocelle are related to urban regeneration of green spaces. Actually, thanks to the Rome Municipality authorization, citizens are allowed to convert these areas into a garden where to grow fruit and other vegetables, as plants and flowers. During the RSE project, three community gardens have provided data on their area dimension, as reported in Table 2.

Table 2. Data collection of Centocelle Community gardens

Community gardens	Short description	Dimension of green area (m ²)
“100 e a capo” community garden	Small community garden with plants and flowers	900
“Centocelle Public park”	Public park with a small education garden	6.000
“Tor Sapienza” community garden	Small community garden with fruits and vegetables	840

2.4. Co-workings

Other CE practices identified during the RSE project have been related to co-working. Coworking is becoming a new way of conceive work, especially in the larger urban centers which allow an economical and shared use of workspaces (individual workstations and meeting rooms) within an equipped and organized facility (internet access, printers, kitchens, relaxation areas, etc.). In Centocelle, two co-workings have been identified: L’Alveare and Fuso-Lab. Data on workstations provided by both Centocelle co-workings has been collected in Table 3.

Table 3. Data collection on Centocelle Co-workings

Co-working	Number of working stations	Other facilities
L’Alveare	30	2 offices 1 meeting room 1 baby space 1 local food purchasing group 1 kids clothes exchange service
Fuso-Lab	20	1 single private room, 1meeting room, 1 space for training courses, 1 conference call-room 1 room for event, 1 cafeteria with relax area, 1 self-managed food point, 1 info-desk outdoor spaces.

2.5. km0 practices

Other CE practices identified in Centocelle are based on the zero-kilometer (Km0) culture, an approach that promotes local products ensuring distribution transport reduction. In particular, the RSE project has identified a Km0 restaurant and also a local purchasing group. The main characteristics of a Km0 restaurant are the food quality (controlled and recognized), the respect of food seasonality and the promotion of local products.

In this study, a Centocelle Km0 restaurant, called DOL (Di Origine Laziale), has been investigated. According to data provided by the DOL restaurant owner in an interview, most suppliers are located within 170 kilometres and about 60 meals are served every day. In particular, DOL restaurant promotes the quality of Lazio Region food and wine and spreads the concept of local and short supply chain. Even local purchasing groups are Km0 practices arranged directly by consumers who cooperate to favor small local producers, product quality and respect for the environment. These groups in Italy are known as Gruppo Acquisto Solidale (GAS). As described in Table 3, L'Alveare co-working provides a service of GAS.

In this group, the most sold products are especially local fruit and vegetables. According to data collected during an interview, a typical shopping composition is: 70% vegetables, 10% fruit, 10% dairy products and 10% meat. GAS suppliers are located no more than 100 kilometres distance and the amount of purchased products is 150 kg per week.

2.6. Water house

Another CE practice is the water house: a modern fountain delivering water from local water supply. Currently, 77 Water Houses have been installed in Rome, and they are managed by ACEA (ACEA, 2018): 22 water houses are located in the city districts and 55 water houses are across the province of Rome. Centocelle is located in the V Municipality district of the city of Rome. Unfortunately, in the V Municipality there is not a water house, but for our study we refer to the water houses in the closest municipalities: IV, VI and VII municipalities, whose data are reported in Table 4.

Table 4. Water House data collection

Water houses	Litres of supplied water (l)
IV municipality (177.000 inhabitants)	7.350
VI municipality (257.534 inhabitants)	2.100
VII municipality (307.607 inhabitants):	750

2.7. Recycling centres

Finally, other CE practices identified in Centocelle were related to waste reduction. Through

these CE practices, waste are not to be disposed, but they are re-introduced in a new economic cycle (Mihajlov et al., 2015). Examples are recycling centres. According to Panaitescu and Bucuroiu (2014), recycling centres allow to reduce the amount of recyclable waste landfilled minimizing the environmental impacts.

Recycling centres are generally managed by municipality. In Rome, AMA, the municipal waste management agency, has opened recycling centers for bulky waste and WEEE to be sent for recovery. Moreover, AMA also organizes a special Sunday bulky waste collection, arranging special recycling points in all Rome City Districts, including Centocelle.

In this study, data on the recycling center of V Municipality of Rome, which includes the Centocelle district, has been analyzed. In 2017, the Recovery Centre collected 3.947 tons of material. Moreover, Sunday every 2 months, AMA is present in the V Municipality of Rome for the special collection of bulky material, wood and metal. During the RSE project, the municipal company AMA provided data on the type and quantity of waste materials collected in the Centocelle district and it emerged that in 2017, 67 tons of material were collected. In Table 5, details on collected waste composition, and quantity are reported. Details on waste destination are not currently available.

Table 5. Waste data for recycling center in V Municipality of Rome

Waste type	Waste amount (tons/year)
C&D waste (cement, brick and ceramic aggregates)	1.884
Wood	1.779
Metal	185
Biodegradable waste	72
Paper and cardboard	65
Bulky goods (doors, beds, sofas and furniture)	29
Total	4.014

Other CE practices reducing waste are second-hand market or reuse centre. In these centres, citizens who no longer need a good, instead of turning it into waste, they can leave or donate them. In Centocelle, a Second-hand market is named Capannelle and managed by an association but currently no data are available for this practice.

3. Results and discussion

From the previous paragraphs several CE practices have been emerged in Centocelle district: community garden, co-working, Km0 restaurant, local purchasing group, water house, recycling centre. In order to better explain how the application of CE principles affects positively the urban transformation, the identified CE practice can be connected to the six ReSOLVE strategies (EMF, 2015). In Fig. 1, CE

practices distribution on the basis of ReSOLVE strategies is described.

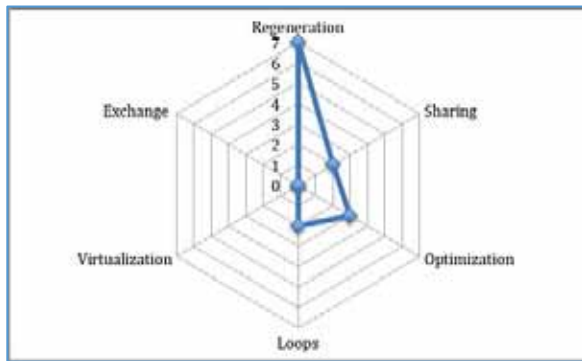


Fig. 1. ReSOLVE strategies application in Centocelle

From Fig. 1, it emerges that the majority of ReSOLVE strategies applied in Centocelle are related to regeneration, followed by optimization, sharing and loops strategies. No practices have been identified for virtualization and exchange. In the following, first results on CE benefits have been described for each CE strategies. According to the data collected in Section 2 and literature data derived by scientific papers and web sites, benefits in terms of CO₂eq emissions avoided, CO₂ emissions absorbed, waste reduction and prevention have been described.

3.1. Regeneration strategies

Most of Centocelle CE practices concern community gardens, which are initiatives generally aimed to regenerate urban degraded areas and transform them in green areas. Actually, green areas are the biggest carbon sinks in urban area. Carbon sinks are natural systems that suck up and store the greenhouse gas carbon dioxide from the atmosphere. According to a study carried out by Monito del Giardino (GR, 2014) the absorption of CO₂ in the Italian urban area each hectare of green area (consisting of a standard mixture of trees, shrubs and meadows of medium and high height) has an average absorption of 4.2 tons of CO₂ per year.

This value is within the range of other studies (Elmqvist et al., 2015). Therefore, the quantity of CO₂ emission absorbed by each community garden has been calculated (Table 6).

Table 6. CO₂ emission absorbed in community garden

Community gardens	CO ₂ emission absorbed per year (kg)
“100 e a capo” community garden	378.0
“Centocelle Public park”	2.520.0
“Tor Sapienza” community garden	352.8

The community garden environmental benefits have been calculated on the basis of CO₂ emissions absorbed by the green area dimension

(square meter). As a consequence, community gardens are CE practices allowing the enhancement of natural system in urban area.

3.2. Sharing

Another CE strategy applied at urban level is sharing and two co-workings are identified in Centocelle. Thanks to the sharing use, several benefits can be achieved by co-working compared to a traditional office. Different studies highlight these benefits in terms of CO₂eq emissions avoided for each working station service (Bolici et al., 2015; Swift et al., 2014). Data are summarized in Table 7. For the sharing use, it is assumed 1 equipment for 15 coworkers.

From the Table 7, the CO₂eq emissions avoided in a coworking are mainly due to a reduction in the demand for work mobility, mainly thanks to a greater proximity between users home and coworking (Padovani et al., 2017). Other coworking advantages are related to the space use optimization, which implies a reduction of heating and electricity consumption (Rangone and Bertelè, 2012). Moreover, the sharing of equipment leads to significant reductions in energy consumption.

Table 7. Estimation of CO₂ emission avoided for co-working workstation

Coworking service advantages	CO ₂ emission avoided for workstation [kg CO ₂ eq/year]
Sharing use of: wifi, printers, coffee machine	123
Energy saving	180
Desk use increased (20%)	700
Carpooling from home to work	887
Total	1.890

3.3. Optimization

Other significant results have been achieved thanks to km0 practices, as restaurant and local purchasing groups (in Italy GAS). These CE practices apply optimization strategy through the promotion of local products. Generally, in supermarkets and other traditional stores, several products come from all over the world. It has been estimated that only the 20% of the energy needed to produce and market agri-food products is due to the agricultural sector and most of energy consumption are actually absorbed by processing, packaging, refrigeration, transport and distribution phases (DEFRA, 2017; Tukker et al., 2006). In particular, food transport has significant and growing impacts and it is an important factor affecting the environmental impact of food.

According to a study carried out by the main Italian farmers’ organization (Coldiretti, 2011), in Italy a meal travels an average distance of 2,000 kilometers before reaching to the consumers table. According to these studies, km0 practices allow optimizing and reducing food transport impacts. As a

consequence, DOL km0 restaurant allows reducing about of 90% the food transport of a traditional restaurant.

Another “zero-kilometre” practice identified in Centocelle is water house. This CE practice applies the optimization strategies utilizing local water instead of mineral water. Different advantages are afferent to water house practices (Botto et al., 2011; Nessi et al., 2012). In this study the reduction of mineral water plastic bottles uses and, consequently, urban waste prevention has been emphasized for their connection to CE principles.

In particular, supplied water liters have been identified as reference flow for water house, and then it has been calculated the quantity of PET bottles (1.5 litres) avoided in the urban waste. In table 8, an estimation of waste prevention connected to the three water houses identified in Rome Municipalities close to Centocelle is reported. Data has been calculated on the basis following three assumptions:

- no water leaks are accounted, so the total amount of water supplied by water houses has been assumed for drinking use;
- avoided water bottles are considered of 1.5 liters PET bottles, according to Bevitavia (2018) it is the most packaging used in Italy;
- weight of PET water bottle is assumed of 28,7 g (Diercxsens, 2014).

Table 8. Plastic PET bottle and waste avoided by water house

<i>Water houses</i>	<i>Number of 1.5L PET Bottle Avoided</i>	<i>Plastic waste avoided (kg/year)</i>
IV municipality (177,000 inhabitants)	4.900	140.63
VI municipality (257,534 inhabitants)	1.400	40.18
VII municipality (307,607 inhabitants):	500	14.35
TOTAL	6.800	195.16

3.4. Loops

Finally, other CE practices identifies in Centocelle are related to loops strategy. In particular, thanks to recycling center practices, collected waste can be reused or transformed into secondary raw materials, reducing waste-related impact such as resources consumption, energy use and GHG emissions. Moreover, second-hand markets favor to reuse products that are in good condition and working. In this case, the goods life is extended avoiding the production of waste. In RSE project, only for the recycling center practices data were available, therefore the total amount has been calculated on the basis of the data collected and resulted in 4,014 tonnes of waste avoided.

3.5. Discussions

From the previous consideration, this study made a first attempt to identify benefits related to CE practices at urban level. This study has carried out a first screening based on data from literature related to climate change and waste production issue. For a deeper understanding of the whole impacts of cities’ activities (i.e. water consumption, atmospheric aerosol loading, chemical pollution, eutrophication, etc.), a detailed analysis according to decision support models (Ghinea and Gavrielescu, 2010) is needed in the future.

As final consideration, it can be affirmed that CE practices based on ReSOLVE strategies has revealed an opportunity for urban transition towards circular economy and in this process the Smart Community engagement process has played a crucial role. In particular, the Smart Urban network SUN has stimulated the local community in sharing information and expressing their ideas on CE practices. Moreover, applying CE at urban level has shown further implications in terms of urban district quality of life improvement and urban services effectiveness and efficiency. In particular, several socio-economic aspects are connected to CE application (Carolina et al., 2017; Ghisellini et al., 2016; Mihelcic et al., 2003). An example is the CE practice of community garden projects that were originally born especially for social purposes with the aim to aggregate citizens. Actually, community gardens are usually managed by citizens’ associations and several social activities are organized in them such as: gardening education, garden cultivation and recreational, sporting activities. Also, co-workings are characterized by services and facilities that can be related to social aspects. For example, in Centocelle, L’Alveare co-working offers also vocational training projects and social inclusion projects for migrant women living in the neighborhood.

Even the other Centocelle co-working, Fusolab, is managed by a social promotion association that promotes educational, cultural, technological and social projects for the benefit of the local community. Furthermore, second-hand markets are often connected to socio-economic aspects. In particular, second-hand spaces are generally managed either municipality or voluntary associations. In some case, volunteers can sell goods and earnings finance social projects. In other case, goods are selling for free or directly donate to poorer people. Moreover, the km0 practices can also produce important social-economic benefits, such as the promotion of local agricultural, culinary tradition and support to the local economy. Finally, other advantages are related to the water house practice.

Not only the plastic bottles use is avoided, but also, they produce economic advantages. Studies are estimating about in 130,00 euro/year for an average family (ISTAT, 2018). Therefore, the introduction of CE practices allows urban communities saving both

economic and environmental resources and also meeting social needs.

Conclusively, urban transition is a complex and systemic process and it cannot be achieved by any single actor. It will require collaborative efforts across the urban community, involving individuals, the private sector, different levels of government and civil society. Therefore, transforming cities into circular aggregates requires not only top-down traditional innovation approach, but also a collaborative approach based on user experience and co-creation. CE practices can promote systemic and collaborative change in society through the sharing of resources, the development of new skills and the co-creation of new urban communities.

4. Conclusions

Circular economy in cities is a challenging opportunity and this paper has described an experimental project carried in Centocelle Rome district aimed at implementing CE within a Smart District Model. In particular, co-creation interventions have been explored in collaboration with citizens and thanks to this experimental and collaborative approach, CE practices have been selected and studied. A particular focus was on the initiatives based to CE principles (as natural system regeneration, keeping materials in use and reducing waste and environmental pressures) developed in a Smart Community.

Thanks to the crucial role of citizens, placed at the centre of the innovation, the adoption of CE strategies, based on ReSOLVE model, were revealed more effective. As a consequence, positive results have been identified both in terms of environmental issues, and new socio-economic interactions. In particular, the application of ReSOLVE strategies such as regeneration, optimization, sharing and loops has brought benefits in terms of waste and carbon emission reduction. Moreover, CE practices at urban scale can also support the growth of new business opportunities and the development of new skills in urban community.

Therefore, CE urban practices have positively and actively influenced the local community real life, fostering a systemic transition towards circular economy.

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OPTIMIZING BIOREMEDIATION OF HYDROCARBON POLLUTED SOIL BY LIFE CYCLE ASSESSMENT (LCA) APPROACH

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Abstract

In the former Carbochimica site of Fidenza, a bioremediation approach was developed using the technique of biodegradation of pollutants thanks to a selected autochthonous bacterial-fungal consortium. The soil was heavily polluted up to values of total hydrocarbon equal to 1800 mg/kg. The consortium was selected from the microorganism living in the Fidenza soil, bioaugmented and finally reinoculated in the bio-pile for soil treatment. The approach is absolutely innovative, due to the presence not only of bacterial strains but also for the use of fungal strains operating in synergy with the bacteria. The first data from the trials show an effective soil remediation performance. The LCA analysis allowed to make a global assessment of the environmental impacts of the bio-pile remediation treatment scenario compared to the no-action scenario. Impacts were assessed on 18 impact categories at the midpoint level according to the ReCiPe method. For the bio-augmented bio-pile remediation, the results showed a value in the climate change category of 10 kg CO₂ for each ton of remediated soil, and at the same time improvement in the categories relating to the toxicity at the local level.

Key words: bio-augmentation, bio-remediation, hydrocarbon, LCA

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1. Introduction

The deperation of soil in contaminated industrial areas is a crucial practice to reduce significant hazards for health and environment. The remediation of soil answers to the need of restoring the natural ecological functions: geopedological nutrient cycling, primary production of food and feedstock, water deperation and biodiversity saving (WHO,

2005). Several case studies showed that the ecological restoration of soil resulted in a significant increase in biodiversity and in the restoration of critical ecosystem functions (Benayas et al., 2009).

According to the European Environmental Agency, the EU territory counts 340,000 contaminated sites (EEA, 2014, Panagros et al., 2013) of which, at least 290,000 have never been treated. Among the soil polluted by hydrocarbons, the

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recurring compounds are mineral oils, polycyclic aromatic hydrocarbons and mixtures of low weight volatile hydrocarbon (EEA, 2014).

If, on the one hand, the restoration of the natural function of the soil is a social priority at the local level, on the other hand, every intervention of remediation brings environmental costs, which always have a global dimension (greenhouse gas emissions, particulate matter formation), unlike the effect of site pollution. In order to evaluate the effectiveness of a bioremediation treatment, it is necessary to make an analysis of the environmental impacts of all the steps included in the remediation process and to have an idea of the consequences of inaction (no action scenario).

Up to now, the criteria used to choose a soil remediation technology are based on technical standards or cost constraints (Cappuyns, 2013), and yet it is not common practice to perform a global analysis of the environmental consequences at the local and global level for the different technologies and in different environments. The life cycle assessment (LCA) is a widely accepted and applied method for evaluating and quantifying the environmental impacts associated with the life of a product or service, from the input of resources to end-of-life treatment.

Thus, LCA delivers a systemic and global perspective on the service and can be usefully applied to the evaluation of technologies that produce ecological benefits (Hauschild, 2005). LCA is now increasingly used to evaluate the environmental pros and cons of different options for the remediation of contaminated sites (Beames et al., 2015, Hauschild, 2005, Toffoletto et al., 2005).

LCA in the case of contaminated sites ideally aims to account for primary impacts, associated with the state of the site pollution, (i.e. it describes the local impacts related to the pollutants bound in the soil and released during the considered timeframe of evaluation, in addition to the pollutants left in the subsurface during and after remediation); secondary impacts, associated with resources use and emissions arising in the remediation project, (i.e. the use of fuel, electricity and chemicals for the soil clean-up) and finally when possible, LCA should account for tertiary impacts, associated with the effects of the reoccupation of the site: they cover future consequences and impacts of the clean-up process and are linked to the restoration of ecosystemic services, economic benefits and social issues of the future use of soil. In this work, primary, secondary and tertiary impacts were considered, and the work highlighted the impacts at local and global level.

This assessment, therefore, provided a global tool to consider the pros and cons of a bioremediation intervention based not only on the implementation costs or the need for treatment, but also on the consideration of impacts of intervention and non-action.

Finally, the use of LCA allowed pinpointing hotspot in the management of bio-pile that were

critically discussed and represent the basis for a more sustainable approach to bioremediation and bio-pile running.

2. Materials and method

2.1. Description of the contaminated site and remediation technique

The polluted soil evaluated in this work was from the Carbochimica industrial area, a national interest site (SIN) of Fidenza municipality (PC), Italy. The area was occupied for over 50 years by petrochemical companies, and currently the soil is heavily polluted with PAH and BTEX up to a value of total hydrocarbon of 1800 mg/kg. Within the frame of LIFE BIOREST, EU life project, a bioremediation approach was developed using the technique of biodegradation of pollutants thanks to a consortium of selected autochthonous bacteria and fungi. The consortium was selected from the microorganisms living in the Fidenza soil on the base of their ability to degrade different hydrocarbon compounds and to be competitive in the contextual conditions. The selected microorganisms were bio-augmented (Jiang et al., 2016, Mrozik and Piotrowska-Seget 2010, Pino et al., 2016, Spina et al., 2018) and finally reinoculated in the soil for treatment in the dedicated bio-pile. The approach is entirely innovative, due to the presence not only of bacterial strains but also for the use of fungal strains operating in synergy with the bacteria. A detailed description of the bioaugmentation approach used is provided in Spina et al. (2017) and Spina et al. (2018).

The experimental bio-remediation trial was performed on 600 tons of soil, using the facility and the management procedure actually used in the SIN, except for the use of the selected consortia of microorganism.

Excavation of soil, the first phase, was followed by the inoculation step, i.e. the soil was supplemented with rice husk, the carrier of the fungi-bacteria inoculum, nutrients were added (nitrogen and Phosphorus) and finally mixed. The soil was then positioned in a closed vessel on a waterproof platform made by a geo-membrane of high-density polyethylene (HDPE), the bio-pile. Air was forced for a total of 12 hours a day into the pile to support the composting process. The temperature of the biomass and the oxygen concentration in the outlet flow were monitored during the process.

2.2. LCA Goal and scope

The objectives of this study are a) to evaluate, via attributional LCA methodology, the potential environmental impacts of the remediation process for hydrocarbon-contaminated soils, as developed in the BIOREST project (bio-augmentation of fungi-microbe consortium), b) to compare the effects of bioremediation to that of the no-action scenario, c) to highlight possible points for improvement of the

environmental performance of the bioremediation and model an optimized scenario.

2.3. System boundaries

The boundaries considered in this work include the inputs of material and energy for all the production steps and the capital goods, i.e. the building of the bio-pile device, the production of the fungi-bacteria inoculum for the bioaugmentation, the operation of the facility (energy and fuel, nutrients and water supply), and the disposal of waste (filter, active carbon disposables). Production steps considered to list the inputs are reported in Fig. 1.

The functional unit (FU) provides the reference to normalise all the data in the assessment. The FU for remediated soil should consider not only the final quality but also the starting point of the pollution level. In this work, to take into consideration the clean-up level and the contamination of the soil at the starting point, the FU is set as the amount of soil (1 ton) coming from 0 to 3 m depth, remediated to a level of 50%, i.e. almost 800 mg kgTS⁻¹ of total hydrocarbons have been degraded.

2.4. Inventory

Life Cycle Inventory (LCI), is where the energy and material inputs and outputs (including products, co-products, wastes and emissions) are identified and quantified to provide the basis for impacts evaluation. It is based on the identification of system boundaries (Fig. 1) and the quantification of the inlet and outlet flows.

Primary data coming from the bio-pile were used both for inputs and outputs of the core module of the analysis (structure for bioremediation and managing, level of contaminants at the end of the process). Upstream module data, such as the

manufacturing of products and goods used in the facility and produced elsewhere, transportation of raw materials, extraction and refining of raw materials, come from the database Ecoinvent 3.3.

Emissions of volatile hydrocarbons from bioremediation facilities may impact air quality or human health. In the considered process, the treatment is performed by positioning a bio-pile in a closed vessel under negative pressure, and carbon filters treat all the air before discharge. Thus, the only possible emission in the atmosphere refers to the excavation phase and is accounted in the inventory according to the findings of Ausma et al. (2011) during the landfarming.

Opposite the air emission referring to the situation as it is (soil not remediated) is estimated based on surface volatility of the compounds present in the soil and according to the amount of them likely exposed to the soil/air interface (Nishiwaki et al., 2009).

The pollution level in the soil is the primary impact. In this work, the amount of hydrocarbons in the soil in the reference scenario (no action) and the amount of the residual hydrocarbons in the remediated soil is considered. The evaluation of pollutants' transfer for the soil-groundwater pathway requires the evaluation to be based on primary local data or modelling based on laboratory leaching tests. In this work, the emission to the soil in the reference scenario (no action) and the emission to the soil of the residual content of pollutant in the remediated soil (remediated scenario) is considered. Different conservative models have been reviewed to consider the mobility of pollutants to groundwater, since that is one of the main impacts. Main models and references used to model the leaching into groundwater are Zand et al., 2009, Kalbe et al., 2008. The resume of primary data of the inventory of this work is reported in Table 1.

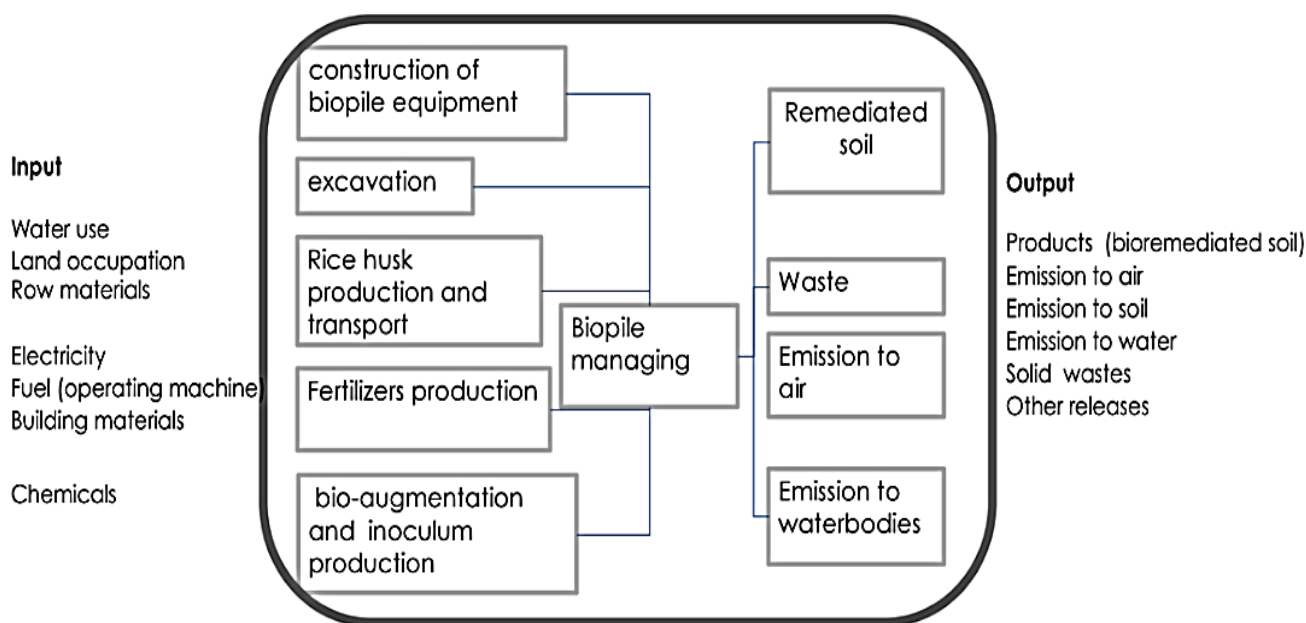


Fig. 1. System boundaries of the bio-remediation technology

Table 1. Data used for calculation of the inventory

<i>Parameter</i>	<i>Unit</i>	<i>Value</i>
Soil treated in a bio-pile	ton	600
Time of treatment	days	150
Average number of treatments performed in the structure (bio-pile vessel) in one year	n	3
Bio-pile equipment, lifetime	years	3
Bio-pile surface	m ²	385
Destination of soil use: urban industrial	m ²	1307
Height of bio-pile	m	1.3
Bio-pile structure		
HDPE vessel	kg	15600
HDPE pipe for water drainage	kg	900
Total HDPE	kg	16500
Steel (equipment/pumps)	kg	100
Average distance from the suppliers of building equipment	km	60
Bio-pile running		
Diesel consumed for excavation in the site and bio-pile production	litre diesel	500
Electricity consumed in bioremediation trial	kWh	10500
Electricity modelled for the optimised scenario		
Air supply	m ³ h ⁻¹	200
Water added to bio-pile	kg	1500
Rice husk added (% of soil volume)	%	10
Urea added to the bio-pile	kg	180
Market phosphorus fertiliser added to the bio-pile	kg	36
Average distance for commercial fertilizer supplier	km	100
Amount of slurry used in the optimised scenario	ton	45
Average distance for slurry supplier	km	20
Bioaugmentation process, inoculum production (for at least 100 bio-pile)		
Lab selection		
Fuel for soil sampling operation: 600 km (return travel with a small vehicle)	km	600
Electricity for lab operation (economic allocation)	kWh year ⁻¹	43200
Gas for lab operation (economic allocation)	m ³ year ⁻¹	2000
Inoculum production, industrial scale		
Fermenter production capacity	litre	200
Heat needed for fermenter (gas to keep 25 degrees for three days) methane gas	m ³	2.0
Electricity	kWh	216
Amount of inoculum needed for one bio-pile	litre	1

2.5. Scenarios considered

The scenarios considered in the analysis are described below. 1) No-action Scenario: no remediation is implemented, and the primary impacts of the pollution of soil are considered to persist for 50 years (land use is prevented, and emission to waterbodies continues); 2) Bioremediation by bio-augmentation Scenario: bioremediation is performed according to the model proposed in the Biorest project and described in the section System Boundaries, data come from a full-scale trial; 3) Optimized Bioremediation Scenario: on the basis of highlighted hotspots and data recorded on-site, an optimized scenario is modelled to overcome the weak points of the process. The optimized scenario is modelled according to conservative assumptions: the key differences with respect to the trial performed in scenario 2, are the use of local fertilizers in place of commercial ones and the lower time of active aeration. Lower aeration can be safely assumed as working mode, considering the very high levels of oxygen recorded in the outlet airflow during the whole test (oxygen level of the outlet airflow always almost equal to 21%v/v).

2.6. Method for the Life Cycle Impact Assessment

In the Life Cycle Impact Assessment (LCIA), emissions and resource data identified during the Life Cycle Inventory (LCI) are translated into environmental indicators. In this work the ReCiPe 2008 method Hierarchist perspective (Goedkoop, 2009; Huijbregts et al., 2017) was used to calculate indicators at midpoint level. The software Open LCA was used for the computational implementation of the inventories.

3. Results and discussion

3.1. Soil characterization

The soil considered in this work was a silty clay soil (45% clay, 23% silt), rather low in carbon content. The soil texture data suggest that it is a severe soil to treat: the macroporosity is low, the movement of oxygen and water is hampered, and pollutants are firmly bound to the soil colloids (De Andrade Lima, 2018). Typically clay soil is less aerated and more prone to the conservation of carbon (Hassink, 1997, Jagadamma et al., 2010); thus, the remediation action

is very challenging and demanding in time and energy. The characterization of samples collected at the start and at the end of the remediation process are reported in Table 2

The addition of nutrients to the bio-pile supplies large availability of nitrogen so that no limiting conditions are experienced by the selected consortia to start the degradation of pollutants. The Carbon to Nitrogen ratio (C/N) is equal to 5 at the starting of the activity of the bio-pile and reaches 6.5 at the end of the treatment. The total carbon of soil is coming mainly from the hydrocarbons present in soil (50%, i.e. 1.6 g kgTS⁻¹) and from the addition of rice husk as inoculum carrier. During the process, almost 0.8 g kgTS⁻¹ of total hydrocarbon were degraded, corresponding roughly to 0.31 g kgTS⁻¹ of carbon, i.e. a large amount of the overall decrease of organic carbon in the soil of the bio-pile (0.5 g kgTS⁻¹). These values prove that remediation activity was effective and specific for pollutant degradation.

3.2. Impact assessment

In the no-action scenario (Table 3), the only impact categories listed are the ones related to toxicity (for human and ecosystem), due to the pollutants that are released year by year in water bodies and air, affecting the quality of the environment for years to come (primary impacts). All these categories describe the persistence and accumulation of hazardous chemicals and are all expressed as the amount of 1 kg

of 1,4-dichlorobenzene (1,4-DB) equivalent. Photochemical oxidant formation describes the air pollution consequent to the reaction of sunlight with emissions from the contaminated soil (or combustion of fuel) as it is formed by the NO_x and Non-Methane Volatile Organic Compounds (NMVOCs); it is expressed as non-methane volatile organic compounds equivalent. In the no-action scenario, the emission of NMVOCs due to the soil pollution exceeded the emission caused by the remediation activity (fuel combustion for remediation activity).

LCA categories dealing with toxicity are complex to be described and resumed in one number. The toxicity indicators are related to many compounds that may disperse into different environmental compartments, with different kinetics: thus, they are finally very difficult to model. Research on these categories is still at an initial stage, so these are no categories as robust as the categories related to climate change, fossil depletion or eutrophication. ERA, Environmental Risk Assessment is the tool to specifically address the evaluation of the potential damage to humans and the ecosystem at the local level, while LCA by now, studies the system as a whole and aggregates a large number of impacts on different geographical and temporal scales altogether; it allows to draw the big picture but at the same time lessens the focus on the local scale of the problems, thus may underestimate the local and specific impact of toxicity.

Table 2. Characterisation of the soil used in the bio-pile tests

		<i>Start of bio-pile running</i>	<i>End of bio-pile running</i>
pH		6.82±0.05	6.83±0.03
Hydrocarbon (sum of C<12 and >C12 compounds)	mg kgTS ⁻¹	1615±248	843±150
Carbon content in soil	g kgTS ⁻¹	3.24±0.09	2.74±0.12
Nitrogen	g kgTS ⁻¹	0.67±0.04	0.42±0.06

Table 3. Characterisation at the midpoint level according to Recipe (H) method of the scenario no-action and remediation by bioaugmentation

<i>Impact category</i>	<i>Unit</i>	<i>No- action scenario</i>	<i>Remediation by bioaugmentation</i>
Climate change	kg CO ₂ eq	0	9.69
Ozone depletion	kg CFC-11 eq	0	7.72E-07
Terrestrial acidification	kg SO ₂ eq	0	4.49E-02
Freshwater eutrophication	kg P eq	0	1.64E-03
Marine eutrophication	kg N eq	0	3.74E-03
Human toxicity	kg 1,4-DB eq	1.27	1.69
Photochemical oxidant formation	kg NMVOC	0.04	0.03
Particulate matter formation	kg PM10 eq	0	0.02
Terrestrial ecotoxicity	kg 1,4-DB eq	0.30	4.87E-03
Freshwater ecotoxicity	kg 1,4-DB eq	0.16	0.12
Marine ecotoxicity	kg 1,4-DB eq	0.01	0.07
Ionising radiation	kBq U235 eq	0	0.68
Agricultural land occupation	m ² a	0	0.49
Urban land occupation	m ² a	17.9	0.07
Natural land transformation	m ²	0	1.15E-03
Water depletion	m ³	0	0.24
Metal depletion	kg Fe eq	0	0.38
Fossil depletion	kg oil eq	0	4.60

Bioremediation scenario, on the contrary, displays impacts in the categories related to the operation of bio-pile venting and equipment construction, belonging to the secondary impacts. The emission of CO₂ equivalent (Climate Change), that is a widespread indicator of environmental performance, is equal to 9.69 kg for each ton of treated material (FU).

It is difficult to compare the performance of the remediation implemented in this work with other reference data, as most of the published LCA studies on bioremediation express the impacts as aggregate indicators at endpoint level (Cadotte et al., 2007, Suer et al., 2011; Toffoletto et al., 2005). In this work for clarity and simplicity, we chose to use only indicators at the midpoint level, i.e. numbers that have a stronger relation to the real quantity of the environmental flows. The endpoint indicators describe the effect of damage produced, i.e. provide sharper information on the environmental relevance, but they bring more uncertainty than the midpoint indicators (Hauschild and Huijbregts, 2015). Anyhow, to better frame the relevance of the impact of remediation, we can compare CO₂ emission to numbers more comfortable to manage: 1100 tons of remediated soil cause the same CO₂ emission of only one inhabitant equivalent for one year according to ReCiPe normalization factor. The emission of CO₂ for remediation is mainly due to the use of electricity for bio-pile venting (23%) and to the construction of the device (HDPE for the containment of bio-pile in a closed vessel) that accounts for another 21%. The excavation phase, in which the soil is excavated from the site (up to 3 meters depth) and is positioned into the bio-pile, contributes for 17% of the CO₂ emissions, including the fuel used and all the emission caused by the engine. Finally, 16% of the total CO₂ equivalent is caused by the production of chemical fertilizers (N addition) used in the bioremediation.

The same four categories are, as expected, the main contributors to the category of fossil depletion, that quantifies the total use of fossil fuel; in this category the higher contribution is due to the use of HDPE for the bio-pile equipment.

Terrestrial acidification quantifies the deposition of nitrogen oxides (NO_x), ammonia (NH₃), and sulphur dioxide (SO₂) to the soil in acidifying forms. (Huijbregts et al., 2017). Terrestrial acidification, in the remediation scenario, depends mainly on the use of nitrogen fertilizers supplied to the bio-pile and on the use of rice husk, that is related to agricultural activity (and to nutrients management). Finally, also the use of electricity and fuel brings a smaller contribution to these categories. The category of freshwater eutrophication quantifies the increase of phosphorus in the freshwater environment, where phosphorus is the limiting factor for biomass production, leading to increased biomass productivity and biodiversity reduction. In this remediation activity, the category of freshwater eutrophication is mainly fed, once again, by the combustion of fossil

fuel and the deposition of P contained in the fuel (Wang et al., 2014).

Marine eutrophication refers to the amount of N that will end up in coastal water, causing an increase in primary productivity, as N is the limiting factor for eutrophication in the marine environment. For nitrogen release, in the remediation option, the main contribution is provided by the rice husk added as inoculum carrier. The production of rice husk is related to nitrogen management in soil, leaching and final release to the coastal water bodies. The bioremediation scenario, as the no action, shows an effect on the category of human toxicity, linked to the burning of fuel for the production of electricity and the production of fertilizers (N and P): the impact is almost equally due to the three activities. It is anyhow to highlight that, even if the category of human toxicity is the same as the no-action scenario, the geographical location of pollution is entirely different. The impact of hydrocarbon emission is concentrated explicitly on-site and close to the site, opposite to the pollution and the effect on human health due to the production of electricity and fertilizers that is somewhere at the national level, where the energy production occurs and even further in the case of urea and phosphate fertilizers production. Again, the weights of the impacts for stakeholders are different. Particulate matter formation refers to the emission of NO_x, NH₃, SO₂, or PM_{2.5} to the atmosphere, followed by an atmospheric transformation in the air. It is expressed as PM₁₀ equivalent. Particulate matter formation, as in previous categories, is due to the combustion of fuel for electricity and fertilizers production. The specific bioaugmentation approach used in the trials linked to the specific activity of isolating and producing fungi-microbial inoculum contributed only for 0.03% to the category of Climate change, and the contribution was even lower in the other impact categories.

Finally, LCA can partially model tertiary impacts and then provide some quantification. In the case of no-action scenario, there is an estimation of how many square meters of soil for a year will not be available for use and for the eco-systemic services, such as water depuration and biodiversity reservoir. It is more difficult in this case to provide an explicit "positive" quantification of the eco-systemic services that the soil will provide once the restoration occurs. In this work, it is assumed that the remediated soil will be transformed from an industrial area with no vegetation (low eco-systemic services) to a higher quality ranking area with vegetation.

Comparisons of scenarios highlight the topic of the weighting between primary (local) impacts and secondary impacts, that are often global or far from site impacts (CO₂ emission or ozone depletion). To address this point, somehow lacking in LCA, Diamond et al., (2009) suggested to include in the LCA evaluation, some impact categories that are specific for the effect and damage at local level, such as changes in soil quality parameters, changes in

aquifer and changes in the level and quality of ecosystem regeneration.

3.3. Modelling an optimized scenario

In this work, the results showed, for the remediation option, an impact at the global level (global impact categories such as Climate change), due to the energy demand for soil treatment, but at the same time a definite improvement in the categories relating to the local toxicity, i.e. pollution of the air and water sectors. At the same time, hotspots and possible enhancements are clearly outlined: treatment in bio-augmented bio-pile has its primary source of impacts in the consumption of electricity for aeration and in the use of chemical fertilizers. The amount of oxygen in the exhausted air was always in the range of 21%; thus, excessive ventilation was performed in the bio-pile. If we consider the total carbon amount in soil (Table. 2) and the oxygen consumption related to this degradation, we find the average oxygen rate demand (Table 4) for the complete degradation of all the compounds present in the soil, considering an average value of 3.4 g of oxygen for the complete biodegradation of 1 g of hydrocarbon (Huesemann and Truex, 1996). On the other hand, we can consider the average range of soil respiration rate, when the optimal condition of temperature and nutrients level is assured (Curiel Yuste et al., 2007; Huesemann and Truex, 1996). Considering these numbers (Table 4) it appears that the aeration provided to the bio-pile is 700 times higher than the average soil respiration rate, and 7 times higher than the oxygen supply rate needed to consume all the organic carbon in the soil in 150 days,

carbon from soil organic matter, from rice husk and from pollutants. If we then consider the real amount of oxygen needed for the biodegradation that actually occurred in soil, we find a value quite close to the respiration of well-amended soil, and again the oxygen supplied is significantly in excess compared to what needed. Based on the data discussed, it is possible to model an alternative precautionary scenario, where an on-off aeration system based on O₂ concentration feedback could reduce by one third the electricity consumption all around the lifecycle of the treatment, assuring a more than suitable oxygen concentration for biodegradation process. The opportunity of this reduction is also confirmed by the fact that the setup of aeration of high rate composting facility for waste stabilization is set to assure concentration of oxygen not below 16%, a ceiling that is considered suitable to support the activity of aerobic microorganism (Petric et al., 2012).

Finally, the use of recovered nutrients (slurry and manure) in place of fertilizers, when they are available on-site as it occurs in Fidenza district, could as well decrease the total impacts on the category of GHG and acidification. In this case, the production of slurry is considered burden-free, while it is considered the transport and the emission during use. Values of impacts of the new optimized scenario are reported as comparisons in Fig. 2. The optimized scenario, thanks to the saving of fossil fuel consumption for electricity and fertilizers, allows to decrease the values of the toxicity categories such as human, terrestrial and freshwater, of 68, 99 and 61% with respect to that of the remediation scenario and the CO₂ eq emission drops to values of 5.8 FU⁻¹.

Table 4. comparisons of the oxygen supplied in the bio-pile to the rate of oxygen consumption of soil and to the rate of oxygen consumption needed to consume all the organic compounds in the bio-pile in 150 days

<i>Air supply</i>	m ³ h ⁻¹ Mg soil ⁻¹	0.29
<i>Oxygen rate supplied in the biopile</i>	mg O ₂ kg soil ⁻¹ s ⁻¹	0.024242
<i>Average oxygen rate consumed by soil respiration (Huesemann and Truex, 1996, Yuste et al., 2007)</i>	mg O ₂ kg soil ⁻¹ s ⁻¹	0.000035
<i>Average oxygen rate needed to biodegrade all the carbon in the soil in 150 day</i>	mg O ₂ kg soil ⁻¹ s ⁻¹	0.003498
<i>Average oxygen rate needed to degrade the carbon actually degraded during the trial</i>	mg O ₂ kg soil ⁻¹ s ⁻¹	0.000035

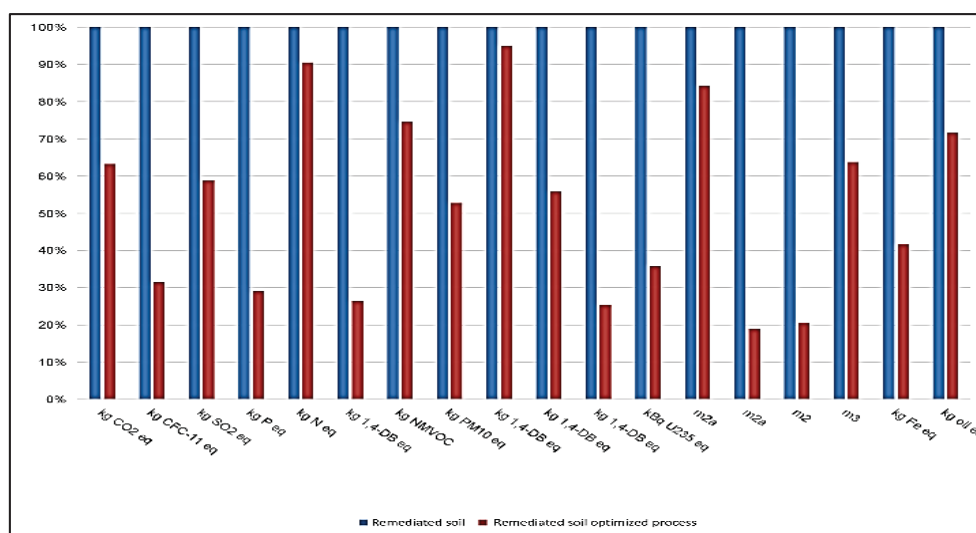


Fig. 2. Comparison between the bio-remediation scenario and the optimized bio-remediation scenario Impacts assessment calculated according to ReCiPe midpoint (H) V 1.12 method

4. Conclusions

Bio-remediation by bioaugmentation model proved to be effective in remediation, as it costed only 9.69 kg CO₂ eq per ton of soil, i.e. 1100 tons of remediated soil cause the same CO₂ emission of 1 inhabitant equivalent for one year according to ReCiPe normalization factor. The use of bioaugmentation technique contributes only to 0.003% of the total CO₂ emission. The management of oxygen supply for biodegradation should be carefully dimensioned, as the electricity used for ventilation is the main contributor to the environmental impacts of the bio-remediation treatment. Bio-remediation by bioaugmentation can be optimized, by LCA perspective, thanks to a more efficient management of air pumping and the use of recovered nutrients, to drastically reduce the value of toxicity categories and achieve a CO₂ eq emission values of 5.8 FU⁻¹.

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BIOMASS EXPLOITATION FOR ENERGY SUPPLY AND QUALITY COMPOST PRODUCTION. AN EXEMPLARY CASE OF CIRCULAR ECONOMY IN THE NORTH EAST OF ITALY

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Abstract

The goal 12 of the 2030 Agenda for Sustainable Development takes into consideration the responsible consumption and production in the perspective of circular economy. The agri-food sector is more actively involved in these initiatives, because it offers the possibility to exploit waste and by-products, by adopting suitable biotechnologies. Such processes can be carried out either under aerobic conditions, for the production of compost, or anaerobically, for the production of biogas. In this work the case of a plant managed by Desag Ecologia, located in the municipality of Sedegliano, in the North-East of Italy, is presented. The plant started up in June 2016. Its main activity consists in exploitation of the organic fraction of municipal solid waste and urban forestry green waste coming from separate waste collection. The basin of provenance of collected materials consists not only of the province of Udine, but also of other areas of the Friuli Venezia Giulia region and other northern Italian regions. The plant ensures the production of both biogas (used in a cogeneration installation for producing electricity and heat) and quality compost, which can be used in agriculture, after submission to physico-chemical analyses to verify the end-of-waste status. In this way, the reduction of waste disposal in landfill is ensured. Thermal energy is partially recovered for the production of hot water to heat the anaerobic digester, the leachate collection tank and the plant rooms. Approximately 10% of electricity is self-consumed for the needs of the anaerobic facility, the remaining amount is fed straight into the public electricity network.

Key words: biogas production, cogeneration, compost production, integrated anaerobic-aerobic plants, organic waste management

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1. Introduction

The European Commission has adopted the revised Best Available Techniques (BAT) conclusions for waste treatment, published on August 17th, 2018, giving to the national authorities the guidelines for technical installation. The document contains BAT conclusions for the most common waste treatments, including mechanical, biological and physical-chemical treatments and treatment of water-based liquid waste (EU Decision, 2018).

The Directive 2009/28/EC fixed the guidelines for waste management, recycling and recovery, in

order to reach a separate waste collection (SWC) of 50% by 2020 (EC Directive, 2009). According to the “Report on Municipal Solid Waste (MSW), 2017 Edition”, (ISPRA, 2017), Italy is not so far from this target, even if the situation is different among the regions. In 2016, Italian production of municipal solid waste (MSW) was 30.1 million t, with a 2% increase in respect of 2015. The percentage of separate waste collection (SWC), calculated as the ratio between the amount of materials collected by separate waste collection and the amount of materials collected as unseparated MSW x 100, was 52.5%, with an increase of 5 points if compared with 2015.

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SWC per capita was 261 kg at the national level, 328 kg in the North of Italy (+38 kg more than in 2015), 266 in the Centre (+28 kg more than in 2015) and 169 in the South (+20 kg more than in 2015). From 2011 to 2016, SWC variation was 62 kg per capita. In 2016, in the Friuli Venezia Giulia (FVG) region, in the North-East of Italy, SWC was 67.1%, with an increase of 6.7% in respect of 2015.

The EC Communication 2015/614/EC promoted SWC in order to decrease landfill disposal, by introducing economic incentives for technological solutions (EC Communication, 2015). In Italy, the Ministerial Decree of June 23, 2016 fixed exact dispositions for plants fed by biomasses, biogas and sustainable bio-liquids production (MD, 2016). According to the LD, (2003), biomass is the biodegradable part of the residues of forest sector, agricultural and food industry sectors, zoo-technical sector, organic waste (residues of green and scraps of food)

Every year, the report on the State of Green Economy intervenes on national and European debate on the sustainability of economic recovery and job occupation. In particular, the General States of the Green Economy 2012 organized a national strategy for the revival of biomass supply chain: "Biomass potential in Italy is very high, but there are many obstacles for its exploitation. It will be necessary to develop the second and third generation of biofuels, the biogas/bio-methane supply chain and the energetic valorization of biodegradable fraction of waste, taking into account the respect for the European hierarchy" (Ronchi and Morabito, 2012).

In 2016, SWC of organic fraction (consisting of the wet fraction of kitchen scraps and of waste coming from the management of gardens and parks and ornamental green) in Italy was 6.5 million t, considering also the quantity destined to domestic composting, which corresponds to more than 220,000 t; an increase of almost 450,000 t (+7.3 %) was observed compared with 2015. Also, in 2016, the plants of mechanical-biological treatment processed, in Italy, more than 10.8 million t of waste, with a 4.4% increase in respect of previous year. About 5.7 million t were recovered in these plants at the end of the process (+ 10% in respect of 2015). Almost 3.4 out of 5.7 million t were addressed to composting plants, about 2 million t to integrated anaerobic/aerobic treatment plants and little more than 249,000 t were worked in anaerobic digestion plants.

In 2016, 326 plants (309 in 2015) were working in Italy. More in particular, 274 plants (263 in 2015) were devoted only to aerobic treatment (composting), 31 plants (26 in 2015) to integrated anaerobic/aerobic treatment (ISPRA, 2017) and 21 plants (20 in 2015) to anaerobic digestion (Bacchetti et al., 2013; Bozano et al., 2012). 26 integrated plants were located in the North, in particular 2 in the FVG region, 2 in the Centre and 3 in the South. In these plants, biogas is produced by anaerobic digestion and compost by aerobic degradation of digestate and other organic waste. The composition of the treated substrate is

made up of 69% of wet fraction, 10% of green fraction, 15% of sludge and 6% of other waste: the organic fraction is 91% of the total waste managed by integrated plants (ISPRA, 2017).

Exploiting the bio-waste involves many associated activities: the collecting services, the technical effort for the plant project and realization, the activities for the valorization and the use of compost. The affair volume of the supply chain for the collection-treatment of bio-waste is about 1.8 billion euro/year (ICC, 2018).

The integrated plants, built to control the odorous emissions and to stabilize the biomasses, are constituted of sequential treatment lines to recover renewable energy under biogas/bio-methane form, and to transform the digestate, plus other organic waste, by aerobic treatment, in quality compost for use as fertilizer in the agricultural sector (ISPRA, 2017).

An integrated plant is a sustainable plant, from all the points of view, giving benefits to:

- the economy of the territory, by assuring work to the community and by producing profits (economic and social support for the society);
- the community that is sure that the collected organic fraction is re-cycled (to produce renewable forms of energy and compost);
- the environment from which fewer primary resources are taken, through the use of renewable materials (organic waste); moreover, burning biogas is less polluting for the environment than burning fossil fuels (Arthur et al., 2011). Furthermore, the produced compost is suitable to fertilize fields, when the results of the chemical analyses assure its qualification to be used in the agronomic sector (ICC, 2018).

Biogas composition mostly depends on the type of decomposed material 50–85% CH₄ (methane); 20–35% CO₂; H₂, N₂ and H₂S form the rest (Pastorek et al., 2004; Salomon and Lora, 2009; Vasmara and Marchetti, 2018).

Biogas production can be considered a sustainable process for simultaneous treatment of organic waste and generation of renewable energy (Angelidaki et al., 2018; Mateescu et al., 2008). Biogas is a clean fuel used for heat, electricity and transport (Scarlat et al., 2018). If biogas is used as fuel, it is necessary to remove carbon dioxide to increase the percentage of methane and subsequently its calorific power.

In Europe, considering the national consumptions of natural gas, the contribution of biogas is 4% on average, but it is widely different depending on the country considered, reaching the maximum of 12% in Germany. Germany, United Kingdom, Italy and Denmark are the leader producers (Scarlat et al., 2018). In Italy, the LD (2010a) defined the conditions that must be met by organic waste to stop the waste status at the end of the composting process. As a consequence, three types of qualified amender can be obtained: composted green amender, composted mixed amender and composted amender with sludge, depending on the typology of waste treated (LD, 2010a).

Afterwards, the LD (2010b) defined quality compost as “the product obtained by organic waste composting under specific technic rules, to be adopted by the State, finalized to define contents and use compatible with environmental and health protection and, in particular, to define quality levels”.

The aim of this study is to present a successful integrated plant, managed by Desag Ecologia, located in Sedegliano, in the province of Udine, in the North-East of Italy, that contributes to circular economy, by exploiting biomass (household organic and green waste) for the anaerobic production of biogas, and subsequently aerobic production of quality compost.

2. Material and methods

An in-depth analysis of successful projects of waste management in the FVG region allowed to find out a recent plant designed to treat the organic fraction of MSW and green waste on the basis of anaerobic/aerobic digestion: Desag plant, located in Sedegliano, in the province of Udine. For getting eloquent data and information, we visited the plant and interviewed the Chief Executive Officer (CEO) of the facility to collect data and information. Furthermore, some e-mails and calls were necessary to improve information on the characteristics and production aspects of the plant. Data and information collected are presented in Tables 1-5.

3. Case study presentation

Desag Ecologia S.c.a.r.l., founded more than 10 years ago, is a special purpose entity set up with 100% private capital, of which leading private companies are part: De Vizia Trasfer S.p.A. and Sager S.r.l.; De Vizia Trasfer S.p.A. is specialized in heavy lifting and industrial installations assembly sectors, Sager S.r.l. is specialized in integrated waste management. In the case studied, Desag Ecologia signed a concession contract with A&T 2000, the grantor society for the building of a plant for renewable energy and compost production, for a total period of 25 years in a project financing operation where the public authorities require private capitals for the realization of work for public use in accordance with current legislation (Italian Law, 1998). A&T 2000 is a public society founded in 1998, as a natural evolution of an aggregation of municipalities of the province of Udine, with the aim of implementing economic and operational strategies in the field of municipal waste management. At present, it incorporates 50 municipalities and has a catchment area of 200,000 inhabitants.

Within the project financing, A&T 2000 is the grantor subject, that is to say who provides the good, in this case waste. A&T 2000 deals only with ensuring the commodity (waste) at a specific tariff agreed with Desag Ecologia. The construction and operation of an integrated plant of anaerobic digestion and composting were charged to concession holder, Desag Ecologia.

The Desag Ecologia plant is located in Pannellia, in the municipality of Sedegliano, in the province of Udine, in the North-East of Italy; its construction started in 2013 and the plant was put into service in June 2016. A schematic representation of the stages of the production process is given in the Results and discussion section (Fig. 1).

After a first phase of waste acceptance, aimed at selecting only compliant waste, a storage phase follows, in specific inner paved areas, coated with anti-wear anti-acid lining and provided with a liquid conveying system. During these phases, specific measures ensure the least pollutant or odorous emissions leakage. Accepted waste is classified into four categories: organic fraction of municipal solid waste and similar, mowing materials/small wood waste, large wood waste and sludge. Purity of incoming materials is verified by random sample analysis, visual check and bulky waste removal. The production process starts with a waste pre-treatment phase: waste pass through a machine devoted to bag opening (the machine is also equipped with a system aimed at detecting foreign materials). Large wood waste are subjected to a possible volumetric reduction (chipping).

Anaerobic digestion. Anaerobic digestion is performed in a single-stage, batch feed, dry fermentation way, at 37°C for a duration of 28 days. The plant consists of 8 fermenters, that allow to treat approximately 830 m³ of mixture. The fermenters are equipped with a heating system installed on the bottom and the walls, which is ensured by the heat recovered by the group of biogas cogeneration. This system allows to maintain the process temperature in fermenters.

The fermenters are uploaded (and emptied after a fixed reaction time) in different days, properly scheduled, in order to ensure continuity to the process and to properly distribute downloading, mixture preparation and material uploading for the following cycle. In order to ensure the conditions of constant moisture and controlled temperature, the leachate generated from biomass is partly collected and sprayed once again on medium in a controlled way.

Biogas Production. Anaerobic digestion process enables to obtain biogas, which is conveyed into two co-generation groups (499 kWe each), for combined heat and electricity production. Before combustion, biogas is submitted to pre-treatments: filtration (on activated carbon), dehumidification (by means of a condenser) and compression at 80 mbar.

The Desag Ecologia plant is equipped with a safety torch, which is activated only in case of the start-up phase or of servicing or failure of the cogenerator, or biogas overproduction. In order to ensure security, in the case that the torch is not sufficient, three emergency chimneys have been set up to ensure immediate leakage of excess biogas. Combustion cogenerative engines are provided both with a system that allows the removal from biogas of sulphur compounds, before introduction in combustion chamber, and with a system for absorbing

nitrogen oxides in exhausted gases resulting from combustion.

Composting. Composting of the mixture is carried out inside 8 independent bio-tunnels, isolated from the external environment. The static heap technique is adopted. The bio-tunnels are supplied with a floor aeration equipment, a system of suction of the exhausted air (conveyed to the treatment section in order to eliminate odorous emissions), a device of collection of the process liquids (stored in a devoted tank) and an automatized system of monitoring and control of the process parameters (temperature, moisture, oxygen and carbon dioxide concentrations).

The composting process provides for the previous preparation of the mixture, which is made up by:

- material digested following anaerobic fermentation (about 50% by weight),
- wood waste after chipping and sludge,
- material already subjected to composting and recycled,
- material subjected to primary ageing which is not yet aged,
- intermediate fraction obtained by compost refining, with size between 10-15 and 100 mm,
- possible compost which proved to be not compliant with the criteria established to define the end-of-waste status, and for which the suitability to be recycled in the process has been positively evaluated.

Previous preparation of the mixture is carried out in a covered area in front of the bio-tunnels. The bio-tunnels are loaded in sequence, at pre-defined time intervals, to assure the process continuity and the correct management of the time for the steps of downloading, mixture preparation and material loading for the subsequent cycle. The composting step has the aim of metabolizing the most easily biodegradable materials; it lasts 14 days and is carried out at the temperature of 55°C. At the end of the process, compost is transferred to the primary ageing step, during which the most complex materials are degraded. The primary ageing step lasts 28 days. Then the material is transferred to the area of secondary ageing, located under a roofing, during which the material is periodically turned over. This step lasts 20 days. The final step of refining has the aim of separating possible extraneous fractions which can be present in compost, that is, plastics, iron components and materials with an unsuitable size (greater than 10-15 mm).

On the obtained compost, sampling and chemical-physical analyses are carried out to verify the end-of-waste status. In particular, the employed criterion is the respect of the limits provided for by the enclosure No 2 to the LD 2010a relative to the mixed composted amender (LD, 2010a). In case of not compliance with the aforesaid criterion, compost can either be managed as waste and sent to authorized plants, or recycled in the production process for the preparation of the mixture to be subjected to

composting, or used as filtering bed during the bio-filtration process.

Systems for shooting down of emissions into the environment and control systems

The Desag Ecologia plant uses systems under continuous improvement for reduction of diffuse and fugitive emissions. All operations that can generate dust or odor are located inside buildings, in enclosed spaces. The plant is endowed with a suction system, which conveys air to bio-filters which are filled with a mixture of aged compost, bark and sawdust.

The facility is equipped with separate networks of collecting, treatment and discharge of wastewater. The plant is fitted with an automation system, which allows monitoring of process parameters and their dynamic setting, notification of operating faults, activation of correction or emergency procedures, actuation of equipment, plant programming and control.

4. Results and discussion

In this paper, the case of the plant located in the municipality of Sedegliano, in the province of Udine, in the North-East of Italy, is taken into account. The plant is devoted to processing by anaerobic digestion both of the waste coming from separate collection of the organic fraction of MSW and of sludge, and to subsequent composting of the digestate.

In the plant, anaerobic digestion of the raw materials is carried out to produce biogas, which is then transformed into both electric power by two cogeneration engines of the whole potential of 998 kW, and thermal power, used to heat the fermenters in order to keep the process temperature of about 37°C. The potentials of the plant are 31,000 t/year of incoming waste, 3 million Nm³/year of biogas production and 10,300 t/year of compost production (Table 1).

Table 1. Plant potentials

Incoming waste	99 t/day
	31.000 t/year
Generative groups electric power	2 groups (499 kW each)
Biogas production	3.000.000 Nm ³ /year
Compost production	30 t/day
	10.300 t/year

Anaerobic digestion. Anaerobic digestion is carried out after a previous preparation of the mixture, which is made up by:

- 50% of waste represented by the organic fraction of MSW and by green waste (mowing and trimming materials, small wood pieces);
- 50% of digestate produced by the anaerobic fermenters and recycled into the process, to allow the development of a suitable bacterial population.

Percent composition of the mixture for anaerobic digestion is shown in Table 2. In Table 3 the biogas production characteristics are presented.

Table 2. Mixture composition for anaerobic digestion

Material type	Percentage
Recycled digestate	50.0
Organic fraction of MSW	42.9
Moving and trimming materials/small wood pieces	7.1
Total	100.0

Table 3. Characteristics of biogas production

Biogas produced per year	3,000.000	Nm ³ /year
Production hours	8.760	hours/year
Biogas flow per hour	342	Nm ³ /hour

Composting

The percent composition of the composting mixture is shown in Table 4. The following fractions are obtained:

- fraction with a size smaller than 10-15 mm, representing compost, on which plastic suction and separation of iron materials are carried out. On this fraction, sampling and chemical-physical analyses are carried out to check the end-of-waste status;

- fraction with a size between 10-15 and 100 mm, mainly made up by woodchips, which can be recycled for the preparation of the mixture to be subjected to composting;
- fraction with a size larger than 100 mm, made up by rejected materials which are managed as waste in authorized plants.

The quality compost obtained is certified by Italian Consortium of Composters for use in the agricultural sector. The mass balance flow-chart is shown in Fig.1. The output of materials obtained in 2017 was 3,408 m³ of biogas, 10,293 t of refined compost, 2,030 t of waste and 2,630 m³ of leachate.

Table 4. Mixture composition for composting

Material type	Percentage
Digestate from the plant	58.5
Not aged compost	30.3
Big wood pieces/waste	6.7
In-between fraction from refining	4.5
Total	100.0

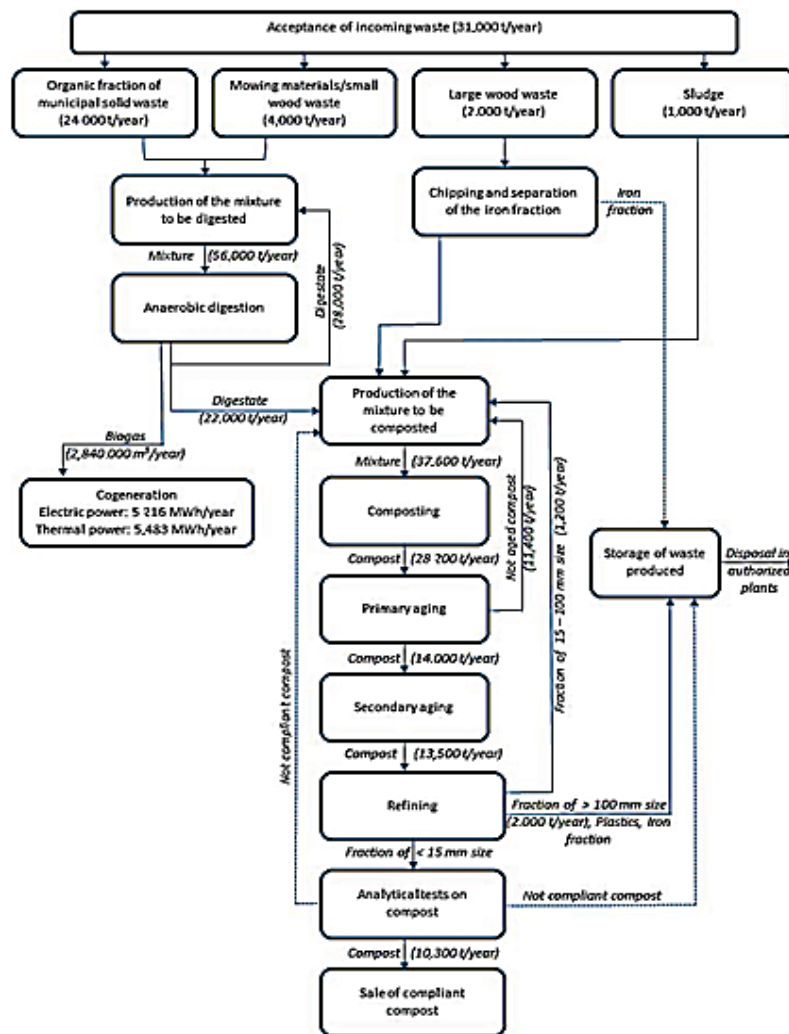


Fig. 1. Mass balance flow-chart for Desag Ecologia plant

At present, 75% of the raw materials treated by the plant comes from the FVG region, while the rest from other basins, but in the future the plant manager will extend SWC to the whole region. In fact, even if the plant started its activity in 2016, the management expects to enlarge the plant potential, by a new area of waste reception, already authorized by the Environmental Integrated Authorization (EIA), and for which other building licenses are required, starting works in 2019. EIA, regulated by the II part of the LD (2006), authorizes the running of plants that carry out activities cited in annex VIII, forcing measures to avoid or reduce air, water and soil emissions (LD, 2006).

Energy balance. The cogeneration engines allow the production of electric and thermal power. About 10% of the electric power obtained is employed for the needs of the plant itself, whereas the remaining amount is distributed by the Enel network. The thermal power obtained is partly recovered by the use of heat exchangers to produce hot water at the temperature of about 85°C. Hot water is employed to heat the digesters, the tank of leachate collection and the technical rooms. The total amount of gas oil employed for collection and transport of raw materials to the plant was calculated on the basis of the average distance covered by the lorries, of the yearly number of transports and of the average gas oil consumption of each lorry, and by considering that the gas oil caloric power is 9.85 KWh/L.

The total amount of gas oil employed for disposal of waste and of part of the leachate produced by the plant was calculated in a similar way. The amounts of energy involved in the management of the plant in 2017 are shown in Table 5.

Table 5. Plant energy balance in 2017

	<i>Thermal and transport power (MWh/year)</i>	<i>Electric power (MWh/year)</i>
Production	5.792	5.500
Own consumption	870	550
Amount sold to a third party	-	4.950
External supply	-	3.965
Dissipated thermal power	4.922	
Gas oil employed for collection and transport of raw materials	2.108	
Gas oil employed for waste and leachate disposal	78	

4. Conclusions

The waste management EU policies act in order to reduce the environmental and health impact. Waste production is increasing because population and consumes are growing, so their production is unavoidable, but it is important to improve the

technologies for recycling the materials that can be transformed in renewable forms of energy and/or products to be used in different sectors.

To increase the SWC percentage at the national level, by shortening the differences among the Italian regions, is a duty of national policies, by reducing the gap between the collected and the recycled waste.

Anaerobic digestion is a suitable, cheap and simple technology that combines biofuel production with a sustainable waste management, with the aim of controlling also the organic waste smell.

In the future, bio-methane will play an important role because of its versatility in substitution of natural gas; in particular, it can be produced close to the point of use, minimizing the problems related to the transit of gas.

In Italy the integrated treatment plant sector (anaerobic/aerobic) has been constantly growing, involving investments, creating new jobs and generating several positive effects on the national economy.

Desag Ecologia plant efficiency is assured by the clean raw materials used, in particular by the organic fraction coming from separate waste collection and sludge. The plant under examination adopts the best technologies for obtaining biogas and quality compost, consequently the outgoing extraneous materials are very low. Furthermore, the environmental impacts (air emissions and wastewater production) are minimal.

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SECONDARY TREATED WASTEWATER AS A SUPPORT STRATEGY FOR TREE CROPS IRRIGATION: NUTRITIONAL AND PHYSIOLOGICAL RESPONSES ON APPLE TREES

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Abstract

Wastewater for irrigating tree crops may act both as water and mineral nutrients source, offering potential agronomical and environmental advantages. This work investigated the effect of an entire season supply of secondary treated wastewater (STW) on the nutritional and physiological responses of 3 year old apple trees. Trees (Gala /M9) were individually grown on 40-L pots filled with a sandy-loamy soil and drip irrigated with: 1) Tap water (TW) (without any mineral fertilizer inputs); 2) Tap water plus mineral fertilized inputs (TW+MF) and 3) STW (without any mineral fertilizer inputs). Each treatment was applied to five individual trees. Daily leaf carbon assimilation rates were promoted by STW, compared to TW trees, although TW+MF trees showed the highest values. Although STW provided a “fertigation-like” effect, the tree nutrient demand was only partially fulfilled. Leaf mineral concentration resulted mostly in the optimal range for STW and TW+MF, except TW, which showed nutritional deficiencies, especially on leaf rather than on fruit tissues. No heavy metal contamination was recorded in STW leaves nor in fruit tissues. A decrease in STW-irrigated tree stem water potentials suggested a moderate salinity stress that indirectly improved fruit quality parameters. Irrigating with STW did not enhance shoot growth compared to TW+MF, promoting instead fruit yield. Results indicate how STW may be suitably reused as a precious resource for supporting the traditional fresh-water supplies in irrigating fruit tree crops. Moreover, the application of STW could allow to partially save tree mineral fertilization needs, thanks to its nutritional inputs.

Key words: fertilization, nitrogen, plant water relations, water reuse, water scarcity

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1. Introduction

Recycling treated municipal wastewater for agricultural irrigation purposes may reduce the water volumes extracted from natural water sources especially in areas facing water shortages. This practice could contribute to recycle nutrients and reduce the amount of pollutants discharged into the waterways (Hanjra et al., 2012). For instance, through wastewater irrigation practices, most of the eutrophication-related elements (i.e. N and P) could be

conveniently reused as fertilizers rather than lost in fresh-water bodies.

Nowadays, the reuse of treated wastewater in agriculture is highly encouraged as the amount of collected and treated wastewater is likely to increase considerably with population growth and urbanization. However, treated wastewater must be carefully managed to protect the environment and public health. Scientific knowledge of such practice on both annual and perennial crops intended for human consumption are highly required (Pedrero et

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al., 2010), especially when its use in agriculture is increasing in the Mediterranean Countries.

Compared to freshwater, treated wastewater has a higher mineral and organic matter (OM) concentration, representing a precious source of nutrients to “fertigate” crops which in turn can provide benefits on plant physiological and nutritional status (Khurana and Singh, 2012). Literature mostly confirms that tertiary treated wastewater (TTW) can be suitably reused as water resource to irrigate tree crops in water-scarce Mediterranean areas (Mendoza-Espinoza et al., 2008; Pedrero and Alarcon, 2009; Pedrero et al., 2013; Petousi et al., 2015; Vivaldi et al., 2013). In Europe a univocal legislation regulating the reuse of treated wastewater in agriculture is currently missing and each Country adopts its own regulation. In Italy, for instance, the reuse of secondary treated wastewater (STW) for irrigation purpose is still not admitted.

The fertilization effect of STW on cultivated crops remains underestimated. Indeed, STW supplies significant amount of OM as well as plant-available nutrients (Chen et al., 2008). Thus, a large-scale utilization of STW to irrigate crops would reduce the need of chemical inputs in agriculture. The use of wastewater in agriculture has been demonstrated to positively affect soil fertility and productivity (WCED Report, 1987). However, most of these studies were addressed using TTW, in which the amount of nutrients, especially nitrogen (N) and phosphorous (P), were significantly depleted as a consequence of the cleaning treatments (Pescod, 1992), while the effect of the STW is only beginning to be explored. On the other hand, although the agronomic validity would need to be demonstrated, the use of STW in agriculture implies environmental (i.e. soil pollution, phytotoxicity), food safety risks and social acceptance obstacles as well (Bernstein, 2011; Fatta-Kassinos et al., 2009; Muchuweti et al., 2006).

Although irrigation of fruit tree crops normally does not wet the plant canopy (preventing external contamination), investigations on the potential consequences of STW irrigation on the tree-root absorption pathway are required before its diffusion on a large scale.

Finally, these studies are of extreme importance to support the legislator in promulgating new regulations about treated wastewater reuse in agriculture. Among the irrigated fruit tree crops, apple is within the most important cultivated species in Italy with a total area over 57.000 ha and an annual production of about 2.4 Mt (Istat, 2018), the most important European producer after Poland.

The aim of this work was to investigate the effect of STW (treated according to the Italian Decree (DME, 2006) as irrigation water on the nutritional and physiological responses of bearing apple trees.

2. Material and methods

2.1. Experimental set up

We carried out a 1-year experiment at the experimental farm of the University of Bologna located in Cadriano (BO), on 15 bearing 3-year old apple trees (*Malus domestica* Borkh) cv. Gala grafted on M9. Trees were grown in 40-L pots each, filled with an alkaline, poorly fertile sandy-loamy soil (USDA classification) and maintained under a shading hail net. Trees were trained as slender spindle, irrigated by four drippers per tree of 2 L h⁻¹ and managed according to the local Integrated Standard Crop Management practices (ICM, 2010) for pruning, thinning, pest and disease management. Climate of the region is temperate sub-continental with warm and humid summers and cold winters. The average annual temperature was 14.1 °C, while annual precipitation was equal to 750 mm.

Starting 48 days (May 15th) after full bloom (DAFB) till 174 DAFB (September 5th), three irrigation treatments were set up, with 5 replicates (single tree) each: 1) irrigation with tap water (TW) 2) irrigation with tap water and fertilization with mineral inputs (TW+MF) and 3) irrigation with secondary treated wastewater (STW). Trees irrigated with STW did not receive additional fertilizer sources. STW (DME, 2006) was provided by the local urban wastewater treatment plant, managed by HERA S.p.a (Italian multi-utility). Along the season, TW+MF trees received 7.83, 1.56, 5.97, and 0.49 g tree⁻¹ of N, P, K and Mg as commercial mineral fertilizers, respectively, split in 3 interventions starting from 48 DAFB. Trees were irrigated twice a day to balance crop evapotranspiration (ET_c) rate.

2.2. Irrigation water chemical and microbiological characterization

Samples of STW and TW were collected at two weeks intervals throughout the irrigation season for chemical analyses, then stored at 5°C. Mineral concentration was determined by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) (Ametek Spectro Arcos EOP, Kleve, Germany) on liquid samples as such. pH was measured with a pH-meter XS PH510 (Eutech Instruments, Singapore) whereas electrical conductivity (EC) was determined by a conductimeter (METERLAB, CDM 210, Radiometer Analytical, France). Finally, total organic carbon (TOC) and total dissolved N were measured in the water samples by an elemental analyzer TOCVepn- TNM1 (Shimadzu Corp., Kyoto, Japan).

The abundance of *E. coli* and *Salmonella* spp., was determined on STW samples by the membrane filtration method. Briefly, for *E. coli* enumeration, membranes were placed onto a Chromocult ES (VWR) agar and incubated at 37 °C for 24 h. *Salmonella* spp. Relative abundance was performed according to procedure UNI EN ISO 19250:2013.

The annual nutrient input was calculated multiplying the concentration of the dissolved elements in TW and STW water by the amount of water provided throughout the season. The TW+MF

annual nutrient input is the contribution of the TW annual nutrient input plus the mineral fertilizer supply.

2.3. Tree nutritional status

Leaf mineral concentration was assessed on ten fully expanded leaves per replicate, randomly selected from annual shoots on the second half of July. Petioles were removed, then leaf limbs were washed, oven-dried, weighed, milled and analysed. N was determined by the Kjeldahl method (Schuman et al., 1973) while P, K, Ca, Mg, S, Fe, Cu, B, Na, Zn, Mn were determined by ICP-OES after digestion with nitric acid (HNO₃) by a microwave lab station (Ethos TC-Milestone, Bergamo, Italy). The same procedure was adopted to assess mineral concentration of fruit peel and fruit pulp on fruit sampled at commercial harvest.

2.4. Vegetative growth and daily photosynthetic assimilation rates assessment

Three shoots per tree were selected and their length was recorded at 34, 41, 48, 54, 60, 68, 76, 83, 93, 104, 128 and 157 DAFB. Furthermore, for each tree leaf net assimilation rate (A) was measured at about 9:00, 13:00 and 16:00 hours at 174 DAFB using a portable gas analyser (Li-COR 6400, LI-COR, Lincoln, Nebraska, USA). Measurements were carried out on one fully-expanded leaf per plant. Light intensity inside the cuvette was maintained constant as recorded by the photosynthetic photon flux density (PPFD) sensor immediately before the measurements. Cumulative daily photosynthesis ($\sum A$) (from 9:00 to 17:00) was then calculated as described by Tozzi et al. (2018) using the following equation (Eq. 1):

$$\sum_y = \sum_{i=t_0}^{t_1} \left(\frac{y_{t_0} + y_{t_1}}{2} \right)_i \sum_{j=t_1}^{t_2} \left(\frac{y_{t_1} + y_{t_2}}{2} \right)_j \quad (1)$$

where: y is the variable A whereas t_0 , t_1 and t_2 correspond to the A values recorded at 9:30, 13:30 and 16:30, respectively. Cumulative daily photosynthesis ($\sum A$) was then multiplied by the total leaf area per tree. Leaf area was estimated by multiplying the total leaf number by the average leaf area, measured by a leaf area meter (LI-3000 A, LI-COR, Lincoln, Nebraska, USA) on three replicates per tree, each of 30 grams of leaves.

2.5. Tree leaf and stem water potential

The daily patterns of leaf and stem water potentials (WP) were assessed at 115 (pre-harvest) and at 150 DAFB (post-harvest). Measurements were performed at 6:00, 9:00, 13:00 and 16:00 hour using a Scholander pressure chamber (Soil Moisture Equipment Corp., Santa Barbara, CA, USA). Leaf water potential was measured on well exposed leaf per tree following the recommendations of Turner and Long (1980). Similarly, stem water potential was measured on leaves previously covered with

aluminium foils and placed in plastic bags for at least 90 minutes prior measurements, to allow equilibration with the stem (Naor et al., 1995).

2.6. Fruit growth rate, tree yield and fruit quality

The diameter of 8 fruit per treatment, randomly chosen, was recorded at 60, 68, 76, 83, 94, 104, 117 and 128 DAFB, using a digital caliper provided with an external memory (<http://www.hkconsulting.it/>). At commercial harvest, yield was assessed for each tree. Fresh weight, dry matter content, flesh firmness, soluble solids content (SSC), skin lightness (L*) and a* b* colour components were assessed on the harvested fruits. Flesh firmness was assessed by a 53220 FTA Fruit Texture Analyser (T.R. Turoni srl, Italy) equipped with a 11 mm plunger. Soluble solids content was determined on the fruit juice by a digital refractometer (ATAGO CO., LTD, Japan) and peel colour was measured using a Minolta CR-400 (Konica Minolta Sensing Americas, Inc, USA).

2.7. Statistical analysis

Shoot length and fruit growth were analysed using a linear mixed model function. A one-way ANOVA followed by a Tukey HSD test using R software (www.r-project.org) was used to establish differences among treatments for daily leaf and stem water potential, daily cumulative photosynthesis and fruit quality parameters. Data of the tissue mineral concentration were analysed according to a complete randomized block design. When the analysis of variance showed a statistical effect, means were separated by the SNK Test (SAS 9.0, SAS Institute Inc., Cary, NC, USA).

3. Results and discussion

3.1. Water quality

As expected, mineral concentration was lower in TW than STW (Table 1). This latter showed a moderately alkaline pH and a relatively low EC and SAR indexes, indicating low risks of soil salinization. Values were even within the Italian legal thresholds for a direct utilization of treated wastewater sources, as TTW, in the agricultural sector (DME, 2003). TOC was almost 10-fold higher in STW compared to TW, with potential benefits on soil microbial activity, CEC and nutrient availability (Beutler et al., 2014). Dissolved mineral nutrients supplied through STW irrigation allowed to save 50.3 % and 75.1 % of N and P, respectively, compared to the reference mineral fertilized treatment (TW+MF). Similar results were achieved in open field by Vivaldi et al. (2017) and Pedrero et al. (2012) on nectarine trees. *E. coli* mean concentration in STW was 4 CFU 100 mL⁻¹, below the Italian *E. coli* threshold for irrigation water (DME, 2003). No *Salmonella* spp. were detected in STW water samples. Ag, Al, As, Be, Cd, Co, Cr, Hg, Mo, Sb, Sn, Ti, Tl, and V concentration either in STW or TW was below the instrumental detection limit (dl).

Table 1. Chemical and microbiological parameters of tap water (TW) and secondary treated wastewater (STW) (n=7 ± SE) and estimated annual nutrient inputs supplied through the water source (TWni, STWni) and from the mineral fertilizers (TW+MFni)

Chemical parameters	Irrigation water		Nutrition elements	Annual nutrient inputs		
	¹ TW	² STW		³ TWni	⁴ STWni	⁵ TW+MFni
pH	7.43 ± 0.04	8.31 ± 0.92				
EC (dS m ⁻¹)	0.47 ± 0.01	1.21 ± 0.04				
SAR	0.63 ± 0.03	1.85 ± 0.04				
N (urea 46%)			N (urea 46%) (g tree ⁻¹)			5.52
NH ₄ -N (mg L ⁻¹)	0.02 ± 0.01	1.02 ± 0.09	NH ₄ -N (g tree ⁻¹)	0.02	0.37	1.88
NO ₃ -N (mg L ⁻¹)	3.28 ± 0.60	11.6 ± 0.74	NO ₃ -N (g tree ⁻¹)	1.18	4.17	1.63
P (mg L ⁻¹)	0.03 ± 0.01	3.28 ± 0.33	P (g tree ⁻¹)	0.01	1.18	1.57
K (mg L ⁻¹)	4.81 ± 1.10	23.2 ± 0.68	K (g tree ⁻¹)	1.73	8.32	7.70
Ca (mg L ⁻¹)	57.3 ± 5.91	72.8 ± 3.7	Ca (g tree ⁻¹)	20.6	26.21	20.6
Mg (mg L ⁻¹)	17.2 ± 1.92	26.2 ± 2.12	Mg (g tree ⁻¹)	6.11	9.43	6.60
S (mg L ⁻¹)	17.1 ± 1.24	28.7 ± 1.25	S (g tree ⁻¹)	6.11	10.3	6.11
Na (mg L ⁻¹)	20.7 ± 0.73	82.9 ± 1.04	Na (g tree ⁻¹)	7.43	29.8	7.43
Cu (µg L ⁻¹)	6.08 ± 1.10	15.9 ± 1.39	Cu (mg tree ⁻¹)	2.18	5.72	2.18
Fe (µg L ⁻¹)	6.00 ± 0.50	22.9 ± 2.31	Fe (mg tree ⁻¹)	2.16	8.24	2.16
B (µg L ⁻¹)	83.7 ± 4.71	180.7 ± 6.47	B (mg tree ⁻¹)	30.1	64.8	30.1
Zn (µg L ⁻¹)	10.3 ± 1.70	42.9 ± 7.20	Zn (mg tree ⁻¹)	3.70	15.4	3.70
TOC (mg L ⁻¹)	1.13 ± 0.21	10.4 ± 1.71	TOC (g tree ⁻¹)	0.40	3.74	0.40
<i>E. coli</i> (CFU 100 mL ⁻¹)	0	4 ± 2				
<i>Salmonella</i> spp.	0	0				

¹Tap Water. ²Secondary Treated Wastewater. ³TWni Tap water annual nutrient input. ⁴STWni Secondary treated wastewater annual nutrient input. ⁵TW+MFni Tap water plus mineral fertilized annual nutrient input

3.2. Tree nutritional status

Trees irrigated with TW (without fertilization) exhibited a leaf N concentration far below the optimal threshold (Table 2), while the overall values of leaf mineral concentration found in TW+MF and STW irrigated trees were close to the optimal range for the same variety (Cheng and Raba, 2009). Leaf and fruit N concentrations were statistically enhanced by the TW+MF, despite a larger canopy development. Intermediate values were recorded in trees irrigated with STW (Table 2). This indicates that N exclusively provided by STW (< 6.0 g tree⁻¹) was not enough to satisfy tree nutrient requirements and in line with what reported by Pereira et al., 2011 on citrus tree nutrition. Indeed, leaf N concentration of mineral-fertilized trees were significantly higher as a consequence of higher N inputs (9.0 g tree⁻¹).

On the contrary, leaf P and Ca concentration in TW+MF trees were significantly decreased, likely due to the dilution and partitioning effect induced by a larger vegetative biomass. An opposite trend was exhibited in the TW trees, with higher concentration for P and Ca (Table 2). Concerning micronutrients, TW+MF increased leaf Fe and Mn concentrations while no effects was detected on leaf Cu, B and Na concentrations, regardless of the irrigation treatment (Table 2). An increased concentration of Fe, Cu and Mn was induced by TW+MF in fruit peel and pulp (Table 2). Instead, a decreasing trend was detected in P, Ca, B and Na concentrations from TW+MF to TW (Table 2). The reiterate supplied of STW as irrigation water likely promoted an increased in the soil microbial biomass, due to the naturally high microbial abundance and biodiversity of this water source.

Thereby, other than the direct nutritional contribution provided by the STW (nutrients under mineral forms dissolved in the water), the effect of the STW-derived microorganisms on the native soil OM on tree uptake, cannot be disjointed (Smith, 1991). The availability of these elements could then allow a significant reduction in fertilizer application while still partially meeting tree nutrient requirements (Pereira et al., 2011), as it has been reported from other studies on fruit trees (Pedrero et al., 2012; Petousi et al., 2015; Segal et al., 2011; Vivaldi et al., 2017).

It is worth to mention that heavy metals accumulation in STW vegetal tissues (i.e. leaves and fruits) was not observed, excluding potential contamination risk for human health. Similar results were observed in olive trees irrigated with reclaimed wastewater by Petousi et al. (2015).

3.3. Vegetative growth and daily photosynthetic assimilation rate

TW+MF shoot length was characterized by a fast increase until 60 DAFB, while afterwards shoot growth rate was much slower and proceeded until 157 DAFB (Fig. 1). Shoots on TW+MF trees were statistically longer from 60 DAFB on, compared to the other treatments, reaching an average length of 32.0 cm shoot⁻¹ at the end of season. Shoot growth on STW and TW irrigated trees showed comparable growth patterns, with limited and slow growth rates, reaching a maximum length of 15.2 cm shoot⁻¹ and 9.9 cm shoot⁻¹, respectively (Fig. 1). It has to be taken into account the different yield of the treatments, that was the highest in the STW treatment, penalizing the STW vegetative growth (Grappadelli et al., 1994).

Table 2. Leaf, fruit peel and pulp macro and micronutrient concentration in TW+MF, STW and TW irrigated trees

Tissue	N	P	K	Ca	Mg	S	Fe	Cu	B	Na	Zn	Mn
Treatment	g kg ⁻¹					mg kg ⁻¹						
Leaf												
TW	11.9 c	2.41 a	14.5 a	13.5 a	2.56 a	0.70 b	45.8 b	8.80 a	27.0 a	63.4 a	15.4 a	23.0 b
TW+MF	19.9 a	1.15 b	14.0 a	10.9 b	2.27 a	0.98 a	90.0 a	9.80 a	25.2 a	52.8 a	10.8 b	34.4 a
STW	16.6 b	2.65 a	13.5 a	13.3 a	2.32 a	0.93 a	54.4 b	9.40 a	24.6 a	62.2 a	14.6 a	31.2 a
Significance	***	***	ns	*	ns	***	***	ns	ns	ns	*	**
Fruit Peel												
TW	2.00 b	0.30 b	3.50 c	1.34 a	0.89 a	0.14 c	37.5 a	1.90 a	24.6 a	27.1 a	2.44 a	5.80 c
TW+MF	2.72 a	0.31ab	3.85 a	0.82 c	0.81 b	0.28 a	44.1 a	2.10 a	11.9 c	19.6 a	2.44 a	7.46 a
STW	2.31 b	0.33 a	3.70 b	0.99 b	0.84 ab	0.22 b	50.3 a	1.40 a	14.8 b	23.1 a	2.73 a	6.55 b
Significance	*	*	**	***	*	***	ns	ns	***	ns	ns	***
Fruit Pulp												
TW	1.01 b	0.61 a	6.05 a	0.36 a	0.22 a	0.08 c	6.20 c	1.76 b	31.8 a	39.0 a	1.75 a	1.13 c
TW+MF	2.72 a	0.43 c	5.12 b	0.23 c	0.21 a	0.14 a	11.0 a	2.34 a	10.0 c	22.6 b	0.70 a	1.70 a
STW	1.34 b	0.56 b	5.40 b	0.29 b	0.22 a	0.11 b	8.40 b	1.75 b	14.5 b	25.5 b	2.15 a	1.44 b
Significance	**	***	**	**	ns	***	***	**	***	**	ns	***

ns, *, ** and ***: effect not significant or significant at $p \leq 0.05$, $p \leq 0.01$ and $p \leq 0.001$, respectively. Within the same tissue, means followed in column by the same letter are not statistically different ($p \leq 0.05$, SNK Test)

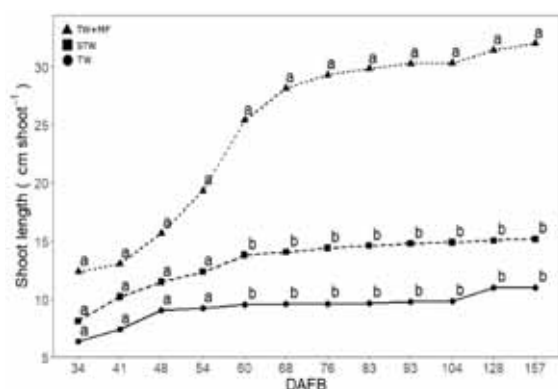


Fig. 1. Seasonal pattern of shoot growth (n=15) for TW+MF, STW and TW. Different letters indicate significant differences with P value <0.05

The different shoot growth rate is a direct consequence of the total N that trees received in the different treatments. TW+MF trees received a higher amount of N, which likely sustained tree growth. This indicates that irrigation with STW may partially contribute to partially fulfil plant nutrient requirements. Therefore, nutrients supplied by STW should be taken into account in the fertilization schedule. In our conditions, results suggest that the use of STW cannot replace traditional fertilization for young apple trees and mineral nutrients must be integrated by alternative sources. On the other hand, irrigation with STW was not detrimental to plant growth (Petousi et al., 2015; Segal et al., 2011) and nutritional status, indicating that STW is a potential water source to irrigate apple trees. Treatments significantly affected tree photosynthetic daily assimilation rate (Fig. 2). Compared to the TW-irrigated trees, irrigation with STW more than doubled the cumulative amount of assimilated C estimated at the end of the season (Fig. 2) with values of 13.4 and 5.04 g CO₂ d⁻¹ in STW and TW irrigated trees, respectively. However, the C assimilated in TW+MF trees was the highest, with a value of 19.8 g CO₂ d⁻¹

(Fig. 2). These differences are likely the consequence of the different nutrient supplies and canopy areas among the irrigation treatments. Tree canopy area was on average 0.68 ± 0.07 m² tree⁻¹, 0.29 ± 0.04 m² tree⁻¹ and 1.42 ± 0.09 m² tree⁻¹ for the STW, TW, and TW+MF irrigated trees, respectively.

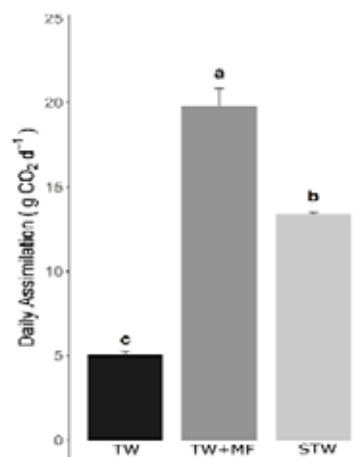


Fig. 2. Effect of the irrigation treatment on the cumulative daily canopy CO₂ assimilation (n=5; Avg. ± SE) measured at the end of the season. Columns with different letters indicate significant differences at P <0.05

3.4. Water relations

Leaf and stem water potentials (WP) showed a decreasing pattern from early morning to afternoon on both the day of measurements (Fig. 3). In pre-harvest (115 DAFB) leaf WP on STW and TW+MF trees were statistically more negative in comparison to TW trees (Fig. 3, b). This difference seems related to the higher water demand of STW and TW+MF trees, which can be mainly attributed to their larger leaf area and fruit yield (Chapter 4.5) compared to TW. No difference was found among treatments on the post-harvest leaf WP (150 DAFB), except at 9:00 A.M. In this case, STW trees showed slightly more negative water potentials.

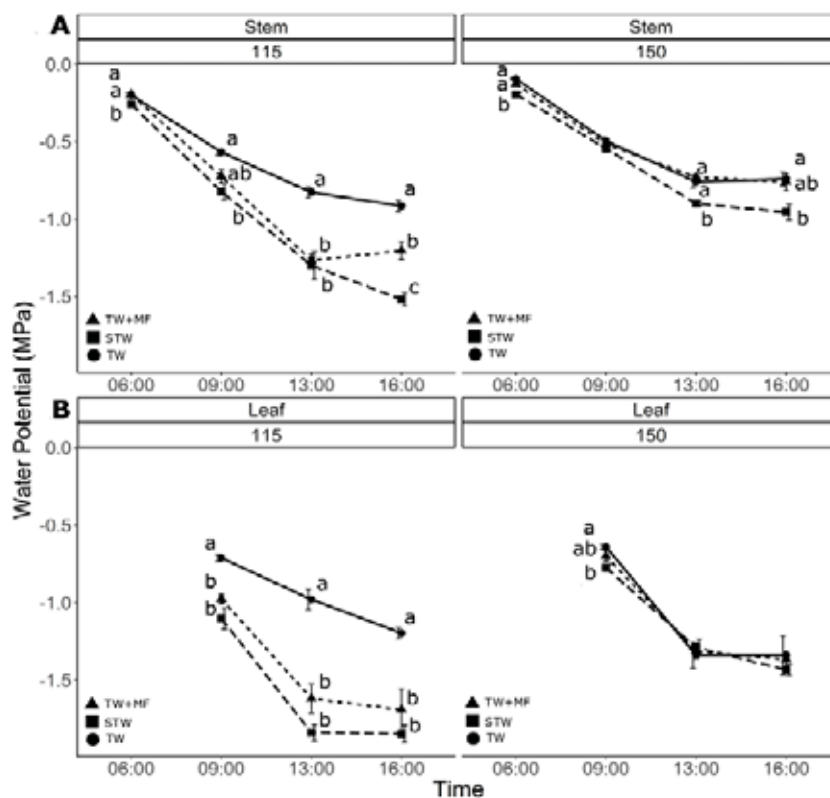


Fig. 3. Daily patterns of stem (A) and leaf water potentials (B) in TW, STW and TW+MF irrigated trees, measured at 115 and 150 DAFB. Each point represents the mean of 5 measurements. Within the same time, values with different letters indicate significant differences at $P < 0.05$

Stem WP at 115 DAFB revealed more negative values on STW trees compared to the other treatments (Fig. 3a) during the whole day, except at midday. This result may indicate a slight salinity stress (Acosta-Motos et al., 2017; Segal et al., 2011) induced by the irrigation with STW that is strengthened by the stem pre-down (i.e. 6:00 AM) data, as a direct indicator of the water soil availability (Van Zyl, 1987). Apple tree is considered among the most sensitive tree crops to soil salinity (FAO, 2002). Such effect was confirmed on the post-harvest measurement, at 150 DAFB, when trees are characterized by a physiological recovering process (Fig. 3), due to the fruit unload (Naor et al., 1997). Indeed, STW trees showed lower stem WPs if compared to TW+MF and TW treatments, except at 9.00 A.M. This is line with the salinity stress hypothesis observed during pre-harvest conditions.

3.5. Fruit growth, yield and quality

The seasonal pattern of fruit growth was not statistically different among treatments (Fig. 4). STW trees showed slightly higher values in fruit diameter for almost all the season compared to TW and TW+MF with a double yield if compared to the TW+MF. The fruit crop load was $1.2 \pm 0.4 \text{ kg tree}^{-1}$, $0.6 \pm 0.2 \text{ kg tree}^{-1}$ and $0.2 \pm 0.1 \text{ kg tree}^{-1}$ for the STW, TW+MF, and TW treatments, respectively. Nicolás et al. (2016) and Pedrero et al. (2013; 2014) found that the use of STW increased yield in mandarin and grapefruit trees,

respectively. Data indicate that STW did not negatively affect seasonal fruit development, despite the higher crop load.

Treatments affected most of the fruit quality parameters (Fig. 5). Fruit from TW+MF treated trees showed statistically higher b^* and lightness (L^*) values compared the other two treatments. Concerning dry matter and soluble solid content, TW+MF and STW trees showed statistically higher values if compared to TW trees. Skin lightness, as well as the b^* colour component, was significantly increased in mineral-fertilized fruits, while similar values were measured for the other strategies (Fig. 5). Conversely a^* component was statistically higher in the fruit skin of STW trees, followed by TW and TW+MF, respectively (Fig. 5). Fruit firmness was higher in TW trees compared to the other two treatments. No difference was detected in the fruit weight (data not shown).

The higher dry matter and soluble solid contents in STW-irrigated fruit, which were characterized also by a higher crop load, could be related to the chemical element concentrations and EC of the STW water (Table 1). In fact, many plants adapt to salt stress by enhancing the concentration of sugars, organic acids, proteins and amino acids which act as osmolytes to maintain plant turgor under salt stress. The presence of these metabolites often increases the nutritive quality and marketability of fruit and vegetables (Ahlem et al., 2011; Ahmed et al., 2009).

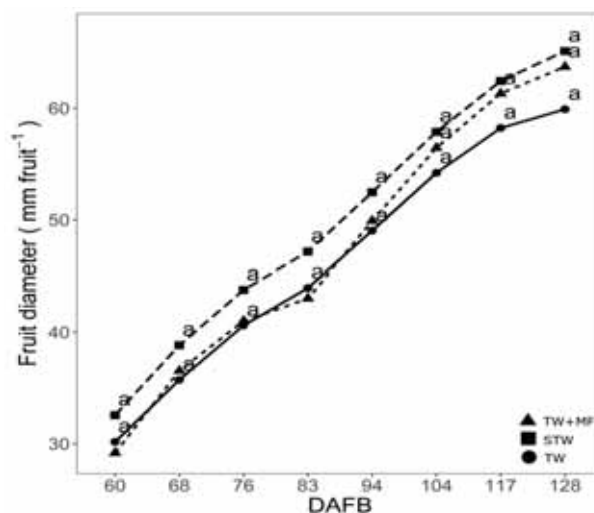


Fig. 4. Seasonal pattern of fruit growth (mm fruit⁻¹) of TW+MF, STW and TW irrigated trees (n=8)

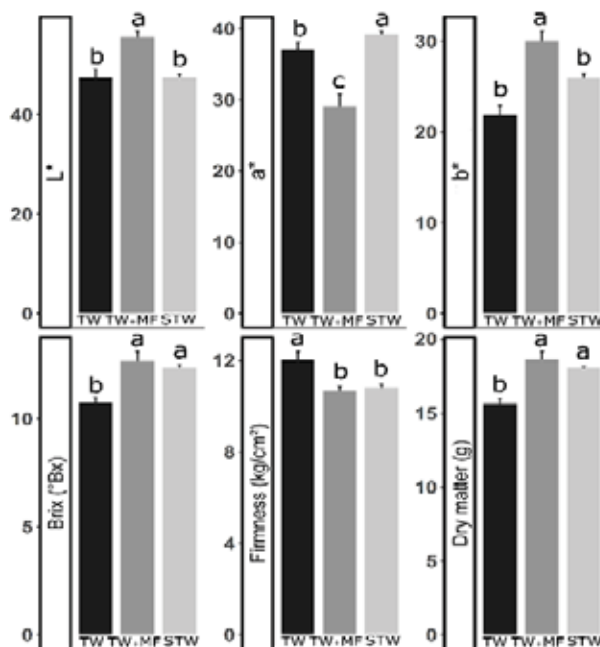


Fig. 5. Effect of the irrigation treatment on the fruit skin lightness (L^* , a^* ; b^* , soluble solid content, flesh firmness, and dry matter) at commercial harvest (n=12; Avg. \pm SE). Black, dark-grey and grey bars indicate TW, TW+MF and STW trees, respectively. Within the same parameter, columns with different letters indicate significant differences at $P < 0.05$. (L^* indicates skin lightness (black=0 while white=100) level; a^* indicates redness-greenness component (red=100 while green=-100) and b^* indicates yellowness-blueness (yellow=100 while blue=-100) component)

It has been demonstrated on different crops (tomatoes, muskmelon, and cucumber), that fruit quality parameters such as soluble solid content, improved in fruits irrigated with reclaimed water (Basiouny, 1984; Biernbaum and Argo, 1995; Crisosto et al., 1994; Lurie et al., 1996; Pedrero et al., 2012). Our data indicate that even if the STW used was not highly saline, fruit quality parameters were positively affected by irrigation with this water.

4. Conclusions

Results suggest that STW can be adopted as a water source in the orchard irrigation management. In

addition, this strategy conveniently contributes to fulfil tree nutrient requirements, with positive responses on the plant nutritional and physiological status. Recycling STW in agriculture allows to recover minerals (i.e. N and P) with positive ecological (e.g. limiting eutrophication problems) and agronomical (e.g. saving mineral inputs) implications.

In our conditions, irrigation with STW did not increase heavy metal concentration both in leaf and fruit tissues, indicating limited risks for human health. We observed a moderate plant water stress induced by the STW, likely induced by salinity. Nevertheless, this response may be associated with the improvement of the fruit quality parameters.

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THE ROLE OF WASTE COLLECTION CENTERS IN A CIRCULAR ECONOMY SCENARIO: AN EMPIRICAL STUDY ON THE CITIZENS' PERCEPTION

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Abstract

At a global level, due to the large diffusion of plastic and electronic products, people are disposing growing quantities of municipal solid waste (MSW), and its composition is more complex than ever before. Effective waste management behaviors, policies and technologies enable a variety of benefits in the multi-dimensional perspective of social, environmental and economic sustainability. Urban waste collection centers represent fundamental incentives for the separate collection of municipal waste, aimed at promoting actions that facilitate the contributions by citizens. In this context, the present study has a twofold purpose: to shed light on the current situation of separate collection initiatives and waste collection centers in Italy by analysing, in a preliminary phase, reference public data concerning six representative cities; to investigate the awareness and perception of quality of these initiatives in the perspective of citizens-users as well as their accuracy and frequency of use. Data collected through an online survey with a sample of 164 respondents were statistically analyzed through multiple regressions and allowed to test the predictive models concerning respectively the correlations among the variables of information and accuracy in the separate collection, and quality of service and frequency of use of collection centers. The results showed that the perceived accuracy is better predicted by information variables rather than by quality evaluation variables. Conversely, the frequency of use variable is better predicted by quality evaluation variables rather than by information variables.

Keywords: circular economy, citizens' perception, European Union regulations, municipal solid waste management, separate waste collection, waste collection centers

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1. Introduction

Municipal solid waste (MSW) is one of the prominent consequences of modern cities and lifestyle. Solid waste reflects the culture of the place where it is produced and its management directly affects the health of the people and the environment surrounding it (Buenrostro et al., 2014; Cocarta et al., 2009; Ionescu et al., 2015; Ortiz-Rodriguez et al., 2018; Vergara and Tchobanoglous, 2012). Rapid urbanization and economic growth largely impact on the generation of municipal solid waste through a continuous process and at an unprecedented rate.

Waste management prediction, options as well as conversion technologies are currently popular topics for discussion among policy makers, regulators, scholars and waste management industries (Buratti et al., 2015; Kolekar et al., 2016; Parkes et al., 2015; Shekdar, 2009; Singh and Ordoñez, 2016; Troschinetz and Mihelcic, 2009).

The amount of waste generation represents an indicator of the urbanization, industrialization and socio-economic development of a country. For instance, due to the recent high economic growth and rapid urbanization in China, the generation of this type of waste is a significant concern for the local

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government in order to protect public health (Chhay et al., 2018; Ghinea et al., 2016).

By considering the total waste generated in the EU, urban waste accounts between 7% and 10% according to the European Commission (Eurostat data). However, it is one of the most complex fraction to be managed and the overall organization and management methods generally represent good indicators of the quality of the entire waste management system of a Country, impacting at the same time on its health and environmental quality levels (Warunasinghe and Yapa, 2016). Urban waste is difficult to be managed due to its characteristics represented by the following factors: (i) extremely complex and inhomogeneous composition; (ii) close proximity to citizens; (iii) high public visibility and (iv) impact on the environment and human health. As a consequence, urban waste management requires a highly structured organization that includes an efficient collection system, an effective sorting system and adequate tracking of waste streams. In addition, the active involvement of citizens and businesses, as well as adequate infrastructures for the specific composition of waste based on a stable financing system are needed.

1.1. Municipal waste management in the European Legislation

Countries that established efficient municipal waste management systems generally achieve better results in overall waste management, including achieving high recycling targets (EU Directive 851, 2018 Of the European Parliament and of the Council of 30 May 2018, amending EU Directive 98 2008 on waste.). In this context, growing attention, both in the fields of science and policy making, has been recently given to the concept of Circular Economy (Bartolacci et al., 2017; Fava et al., 2018; Ferronato et al., 2019; Rada et al., 2017; 2018).

Although both scholars and practitioners have presented it as a novelty, it is worth nothing that it builds on the legacy of predecessors and reference models such as waste recycling and separation, industrial ecology, green economy and Life Cycle Assessment. Some concepts find their origin in the 1980's, such as the concepts of waste hierarchies (i.e. 3R's, 4R's etc.). The 3R's concept, for instance, has become commonplace in many international and national waste regulations (Reike et al., 2018). The waste hierarchy establishes a priority order from prevention, preparation for reuse, recycling and energy recovery through the disposal, such as landfilling. This principle aims to encourage the options that deliver the best overall environmental outcome (European Commission, 2015).

The aim of the new directives of the European Union package (i.e. Directives (EU) 2018/849,850,851 and 852) is to tackle the problem of waste recycling, typical of a linear economy model, through specific measures. On this topic, Directive (EU) 2018/849 states that "waste management in the

Union should be improved, with a view to protecting, preserving and improving the quality of the environment, protecting human health, ensuring prudent, efficient and rational utilization of natural resources and promoting the principles of the circular economy" (European Commission, 2018). The report "Closing the loop - An EU action plan for the Circular Economy" published by the European Commission (2015) sets the guidelines for the conversion from the current economic model of production to a new one that maximizes the efficient use of resources and reduces waste, through specific objectives and policies. One of the main drivers of this transformation comes from the rise of the recycling targets for urban and packaging waste. Consistently with this objective, Directive (EU) 2018/851 introduces the obligation to achieve at least the recycling of the 55% by 2025, 60% by 2030 and 65% by 2035 of all urban waste produced during the year (Directive (Eu) 2018/851, p. 21 – (c), (ii) (e)). Thus, incentives for the separate collection of municipal waste becomes a fundamental choice aimed at promoting actions that facilitate the contributions by citizens, sustained through the dissemination on the territory of facilities such as urban waste collection centers. The sorting of waste through these Collection Centers - defined in the DM 8 April 2008 of the Italian Ministry for Environment forms the basis of the integrated urban waste management system, and represents an indispensable tool to increase the visibility of these policies on the territory Collection Centers, by functioning as an intermediate point, represent in the waste management system the place where MSW already differentiated by users at home, is further sorted by specialized employees prior to be transferred to the recovery centers. For this reason, central governments and local administrations need to strongly encourage the use of these centers and provide current and prospect users with widespread informative actions and by enhancing the quality of these services.

A complete information must illustrate on one hand the functionality of the service provided (i.e. location of the centers, opening hours, what fractions can be conferred, methods of use, regulation, etc.); on the other one, it must highlight the main benefits deriving from these practices in terms of social, environmental and economic impacts (according to the traditional *triple bottom line* framework at the base of sustainable development strategies (Elkington, 1994; Slaper and Hall, 2011).

1.2. Research questions and objectives

As illustrated, among the priorities in the area of municipal solid waste management (MSWM) appears the increase in separate waste collection up to 65%, to be pursued through the preferential recourse to one or more of the following mechanisms: door-to-door collection; the promotion of waste collection centers; the implementation of incentive systems for service users/citizens; the preparation of guidelines to standardize the waste collection on the territory;

users/citizens training and information (ISPRA, 2018). Based on these priorities, this study focuses on investigating, through the analysis of both secondary and primary data, what is the current state of implementation of separate collection as well as the creation of waste collection centers in the major municipalities in the North, Central and South of Italy. The main purpose as well as final contribution of this work is to understand and evaluate how this implementation is perceived by the citizens of these municipalities in terms of quality of services and completeness of the information received. Starting from these challenging objectives, the research questions that guided the development of the present work were formulated as follows:

- RQ1: What is the current situation concerning the differentiated waste management and the implementation of Waste Collection Centers in the most representative Italian cities?
- RQ2: What is the overall evaluation, in terms of quality of services, completeness of information, accuracy and frequency of use of separate collection facilities, based on the perception of citizens from the most representative Italian cities?

In order to answer these questions, this manuscript is based on a two-phases study. The first phase is based on the elaboration of data concerning differentiated waste management and waste collection centers in Italy searched through the public databases of ISPRA (Institute for Environmental Protection and Research) and Istat (National Institute of Statistics). The study examines data from a sample of 6 selected cities (Turin, Milan, Bologna, Rome, Naples and Palermo) based on their size (> 200.000 inhabitants) and their geographical position (the macro-areas of North, Central and Southern Italy).

Despite the great importance of this topic of interest at European and global level, represented by several specific innovations in the European regulation and industrial policies, the impact that the implementation of tools aimed at increasing separate waste collection in a context of change from a linear to a circular economy paradigm have in the perception, attitude and behavior of citizens appear to have received limited investigation by the existing literature (Gutberlet et al., 2017; Lakatos et al., 2018; Ragazzi et al., 2017). Indeed, on the one hand the existing studies on this topic are mainly represented by extensive reports – as the ones carried out respectively by the European Commission on the attitudes of Europeans towards waste management and resource efficiency (European Commission, 2014), and ISTAT about the behaviors and satisfaction of a sample of Italian families concerning differentiated waste collection and municipal policies (Istat, 2018) – which although rigorous and generalizable, analyse the phenomena investigated only on a descriptive level. In addition, the authors did not find any report focused on the perception of citizens regarding waste collection centers and related policies. On the other hand, the academic literature on these specific settings presents a limited number of interesting studies

investigating similar issues but in very different contexts as well as countries (outside the EU), therefore characterized by dissimilar regulations, cultures and habits. For instance Folz and Giles (2002), investigated the impact of "Pay-As-You-Throw" policies on household waste disposal and recycling behaviors among the population of U.S. cities based on average quantities of materials disposed or recycled in these municipalities; more recently Warunasinghe and Yapa (2016) carried out a survey with 50 households examining the status of solid waste management (SWM) household level in a peri-urban area of Sri Lanka and obtaining evidence on the willingness of the people in the participation and their level of awareness about the environmental and health hazards associated with disorganized management of solid waste; Starovoytova and Namango (2018) conducted an empirical case study on SWM at a University college in Kenya, obtaining relevant insights on the level of knowledge, attitudes, and practices of students and vendors concerning SWM, which can be improved through significant and sustained behavioral change, achievable by environmental education. Other studies analyzed the topic of environmental awareness from the perspective of the enterprises (discussed into details in section 3).

The aforementioned body of literature represents a fundamental starting point and source of inspiration for the present work. Its specific contribution and added value reside in the understudied context considered (Italian municipalities), in the novelty of directives and policies of reference, and in the specific focus on urban collection centers. To achieve this result, the second quantitative phase (which builds on the preliminary results of the first one) was developed through an online survey with citizens from the 6 representative Italian cities taken into account. The questionnaire was structured on the basis of the research objectives as well as the models presented in the methodology section.

The remainder of the paper is structured as follows: The next section presents a review of the literature according to the topics of circular economy and environmental awareness. Section 3 describes the conceptual models and the methodology used to build the questionnaire items and to reach the final sample for the online survey. Next, the results obtained are presented and discussed in section 4. The paper is finalized with conclusions and final remarks.

2. Circular economy and environmental awareness

The circular economy (CE) concept is of great interest to institutions, scholars and practitioners because it is viewed as an operationalization for businesses and governments to implement the much debated concept of sustainable development (Fortuna et al., 2012; Ghisellini et al., 2016; Merli et al., 2018; Murray et al., 2017; Prieto-Sandoval et al. 2018; Sihvonen and Ritola, 2015).. As observed above, the

concept of circular economy focuses on the 3Rs of “reducing”, “reusing” and “recycling” materials and energy. The various R frameworks are considered by many authors as the “how-to” of CE and thus a core principle of it (Kirchherr et al., 2017; Reh, 2013; Zhu et al., 2010a; Zhu et al, 2010b). Accordingly, the core European Union Waste Framework Directive was structured based on the 4R framework, which introduces ‘Recover’ as the fourth R. This focus on the multiple “Rs” of the circular economy was found to have a close relationship with environmental awareness (which is connected to the degree of information obtained/owned) and behavior (influenced by many cultural and contextual factors). Liu and Bai (2014) in their study about environmental awareness and the behavior of firms in developing the circular economy reported that environmental awareness has been described in the literature as a multi-dimensional construct (Maloney and Ward, 1973).

With reference to enterprises, Zsóka (2008) showed that the dimensions of environmental awareness include environmental knowledge, values, attitudes and willingness to act, as well as actual behavior (Zsóka, 2008). Furthermore, Sakr et al. (2010) investigated environmental awareness from five dimensions, including the dissemination of information, knowledge and the contractors’ environmental responsibilities (Liu and Bai, 2014; Sakr et al., 2010).

The concept of Environmental perception has been described in the literature as the relationship human beings have with the environment, which determines the attitudes of the people in favor of or against it (Starovoytova and Namango, 2018). Moreover, an increased environmental knowledge leads to an enhanced environmental awareness. For decades, institutions have tried to deal with environmental issues, arguing that technological innovation would have eliminated practices that degrade the natural environment; however the gap between the state of health of the environment and the technological progress is always increasing to the detriment of the former (Barr, 2017). For this reason, waste management increasingly takes the form of regulations or incentives by setting standards, regulations, objectives but also rewarding waste disposal systems and “pay as you throw” taxation (Vergara and Tchobanoglous, 2012).

Citizens have more and more responsibility in the planning and decision-making process within the waste management system, both as a decisive part with their active participation (Rowe and Frewer, 2000) and because of their role as consumers and users of waste management services. Therefore, the study of citizens’ perception on this topics is essential for policy-makers in the decision-making process for achieving environmental objectives (Dahlén and Lagerkvist, 2010; Folz and Giles, 2002; Reichenbach, 2008; Starovoytova and Namango, 2018; Wiedemann and Femers, 1993).

3. Methodology

The preliminary goal of this research was to explore, through the analysis of reliable archival data, the percentage amounts of separated waste collection and the number of urban waste collection centers available per inhabitant in some of the most representative Italian cities. Thus, the first phase of the work is based on the elaboration of the data from two highly reliable sources: the Italian waste cadaster managed by the ISPRA, and the report on “Separate waste collection: behavior and satisfaction of citizens and policies for the cities” published by Istat (Istat, 2018). The purpose is to answer the RQ1 and shed light on the current situation of separate collection initiatives and waste collection centers in Italy.

Based on these preliminary evidences presented in section 4.1, it was possible to build up the second phase of this study, based on the analysis of data collected through an online survey on the perceptions of citizens-users, aiming at achieving the following objectives:

- evaluate the presence of eventual differences among northern, central and southern Italian municipalities according to the different variables of *information* (perception of the quality and completeness of information received about separate waste collection and waste collection centers); *quality of services* (perception of the overall quality of separate waste collection and waste collection centers) and self-reported behavior with respect to the separate waste collection process (accuracy and frequency).
- to assess how the variables of the perceived *information* and *quality of services* are predictive of the accuracy and frequency characterizing the waste sorting implemented by the users.

3.1 Conceptual model and variables

The prediction model of this study includes multivariate analysis of variance and multiple regression analyses. In this work we were interested in investigating the predictors of perceived accuracy of the waste sorting (represented by the construct ACC_WS), which represents the dependents variable of the first model; and the predictors of the frequency of use of waste collection centers by citizens-users (represented by the construct TIPFR), which represents the dependent variable of the second model. The predictors, included in both models, were specified as follows:

A. Information variables:

- INFOCIT: represents the perception about the information obtained by the user-citizen concerning the proper waste separate collection process;
- INITER: represents the awareness of the local initiative present in the territory perceived by the user-citizen (e.g. door-to-door collection; recycling facilities etc.).

B. Quality evaluation variables:

- EVAL: describes the overall evaluation on the usefulness and quality of service concerning specifically the waste collection centers;
- PEQU: describes the perceived quality (in terms of innovation, efficacy and effectiveness) of the separate collection of waste services offered by local administrations.

3.2. Questionnaire

An ad hoc multiple-choice questionnaire was developed as the data collection tool in order to operationalize the model variables into items to be answered by citizens through an online survey. For the development of the research instrument the authors referred to both items and positive evidences retrieved from relevant and recent studies in this field (ISPRA, 2018; Istat, 2018; Starovoytova and Namango, 2018; Warunasinghe and Yapa, 2016). Likert-type scales represent the most frequently type used in survey instruments to ensure reliability and validity of measurements (Edwards and Smith, 2016; Hinkin, 1998). In our questionnaire 5-point Likert scales and specific labeling of points were adopted to indicate the degree of agreement, frequency and relevance. For instance, the multiple-items that measured the D.V. “TIPFR” was as follows: “How often do you confer these types of waste in municipal collection centers?”, presenting a 5-point Likert scale ranging from “never” to “very often”.

The outcome variable ACC_WS, on the contrary, was measured on a 10 points continuous scale ranging from “Insufficient” to “excellent”, according to the following item: “Use the following scale to self-assess your degree of accuracy and care in making separate waste collection”. Dicotomic items were also included in the final questionnaire.

3.3. Sample

Invitations with a link to the online questionnaire were transmitted through the several channels (e-mail, social media, etc.) The sampling strategy was conducted beginning with convenience sampling (inviting colleagues and relatives living in the municipalities chosen), snowball sampling (for those initially invited to distribute the link to other acquaintances) and purposive sampling (directly contacting members of specialized groups of interest on specific waste issues).

The final sample accounted a total of 164 respondents from all the 6 municipalities taken into account. The distribution of the sample in macro-areas is shown in Fig. 1. The municipalities were codified as follows: North (respondents from Turin, Milan and Bologna); Center (respondents from Rome); South (respondents from Naples and Palermo). The sample resulted adequate for further statistical analyses (see next sections).

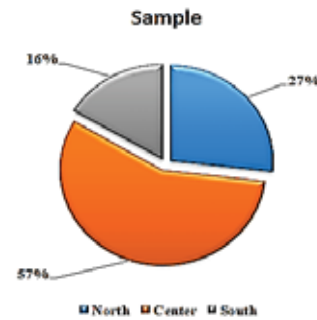


Fig. 1. Survey sample divided into macro-areas

3.4. Data analysis

In order to analyze data as a function of the different aims of the present study we applied the following analyses:

- I. *Multivariate Analysis of Variance in order to test mean differences among the variables included in the models;*
- II. Multiple regression analysis in order to test the predictive power of information and quality variables on accuracy and frequency of waste collection.

4. Results and discussion

4.1. Separate collection and waste collection centers in Italy: preliminary results

In industrialized nations waste tends to be managed formally at a municipal or regional scale (Vergara and Tchobanoglous, 2012). The management of MSW in Italy is obtained through an integrated system, divided into Optimal Territorial Areas. These areas are based on the cooperation among local authorities, with legal, regulatory autonomy, within the organizational and budgetary resources allocated to it by the municipalities, the Province, and the Region (Buratti et al., 2015). Our analysis examines data from a sample of 6 Italian municipalities with a population of over 200,000 and geographically representative of the Italian territory because of their distribution. The time series includes available data between the years 2012 and 2016.

The percentage of separate waste collection in the time period considered (Fig. 2) shows an overall increasing trend - starting from the northern cities with percentages that vary between 42.1% in Turin (2016) and 57.6% in Milan (2016) – passing through the center – 42% in Rome (2016) - up to the south - with significant relevance for the city of Naples 31.3% (2016). The city of Palermo 7.2% (2016) is the only one showing a slightly negative trend among the considered municipalities and time period. By analyzing data concerning the collection centers, in 2017 these areas were used for the provision of urban waste by 45.5% of households throughout Italy at least once. At the regional level, 65.2% of families in the North-East, 57.1% of those residing in the North-West

and 41.3% of the families of the Central Italy. In the South and the Islands, respectively 25.1% and 27.4% of households used those spaces for waste disposal (Istat, 2018).

The percentage variation of separate collection at the considered time t_1 compared to the previous year t_0 , is a useful indicator to measure and evaluate the effectiveness of the introduction of new administrative policies of waste management. Fig. 3 shows a progressive positive variation for the cities of Naples (2013: -1.46%; 2014: 8.37%; 2015: 10.00%; 2016: 29.34%) and at the same time a decreasing trend for the city of Rome (2013: 20.73%; 2014: 19.53%; 2015: 9.30%; 2016: 8.25%). The remaining cities of Turin, Milan, Bologna show ups and downs of growth and degrowth. Furthermore, it is worth noting that the city of Palermo shows only negative values.

About the municipal collection centers, which represent one of the new tools included in the European directives increase the separate collection of MSW and implement policies that encourage the transition to a circular economy, Fig. 4 shows the population with the number of stable collection centers present in the territory. This analysis allows to

compare the average availability of collection centers computed per number of citizens. The evidence indicates a substantial differentiation, from a ratio of 97,000: 1 in Bologna and Naples, to 205,000: 1 in Rome, up to 270,000: 1 in Milan. Currently, there are no fixed collection centers in Palermo. In this case lower is the ratio between the number of inhabitants per urban collection center, greater is the availability and therefore the effectiveness of this waste collection system (used mainly for bulky waste and WEEE).

Therefore, concerning this first objective of the present study, the results show an overall increasing trend of separate collection throughout the Italian peninsula. However, this growth is fluctuating and not yet completely incisive. The objectives set by the European circular economy package are therefore still very distant and complex to reach. The North-South Italy differential analysis highlights also at this stage the structural gap characterizing the southern cities, especially due to a lack of investments (for example reduced number of plants for anaerobic/aerobic integrated treatment of the organic fraction from differentiated collection and incineration plants) (ISPRA, 2018).

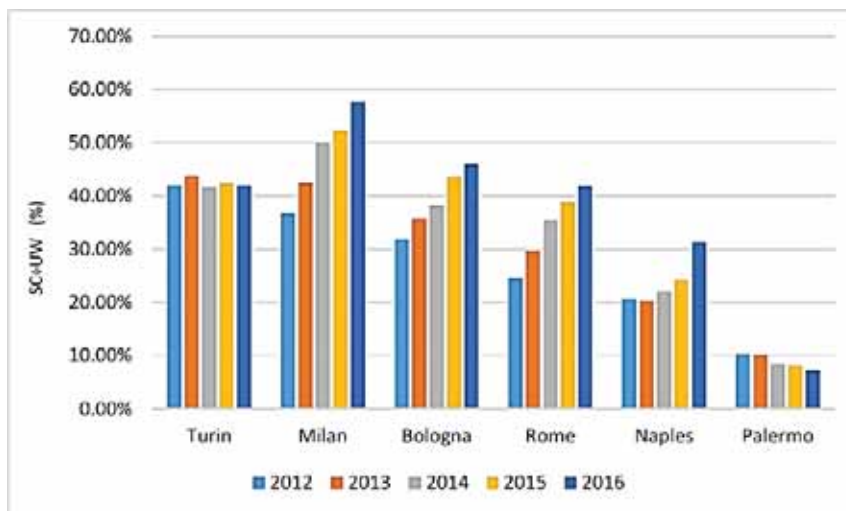


Fig. 2. Trend of the separate collection of municipal waste

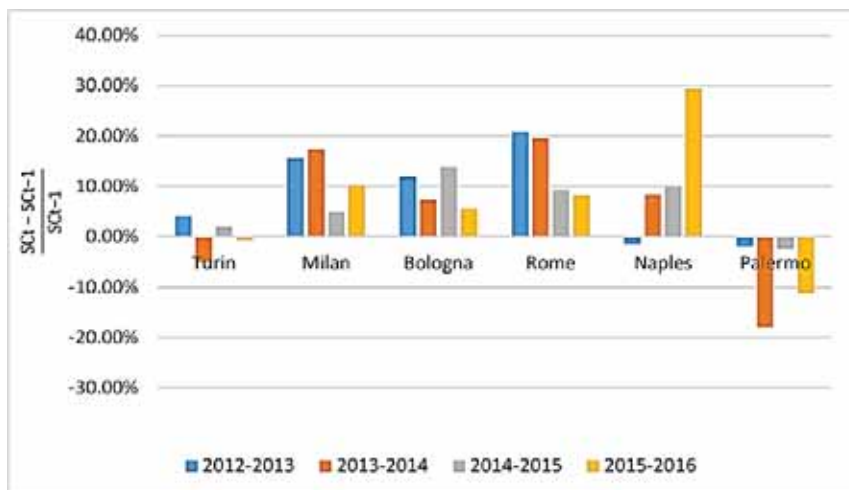


Fig. 3. Percentage variation in the separate collection of municipal waste

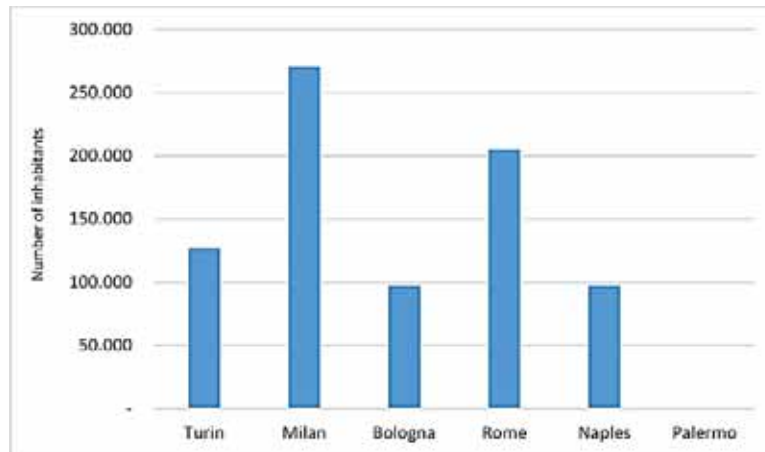


Fig. 4. Number of inhabitants per municipal Collection Center

In the following sections, the results obtained by analyzing the data collected through the online survey will be presented and discussed with reference to our research objectives and questions.

4.2. Descriptive statistics and correlations

Table 1 shows the descriptive statistics (means, standard deviations, asymmetry and kurtosis and internal consistency) of the variables relating to the information/awareness area about the separated waste collection service (INFOCIT, INITER), the quality evaluation area (EVAL, PEQU) and finally to the area of accuracy and frequency of behaviors (ACC_WS, TIPFR). The resulting values of asymmetry and kurtosis are substantially between -1 and 1, indicating that all the variables examined are approximated to the normal distribution. Cronbach’s alpha of the factors computed by multiple items is also reported indicating an adequate level internal consistency for them. Table 2 shows the correlations among the accuracy and frequency variables and the information and quality evaluation area variables. It is worth noting that ACC_WS correlates significantly only with the quality of the information related to the services (INFOCIT) and with the evaluation of the services concerning the waste collection centers (EVAL), with a moderate effect size in the first case and small in the

second one. Moreover, the frequency of use of waste collection centers by citizens-users (TIPFR) correlates significantly with the two indicators of quality evaluation of services with moderate effect size for EVAL and small for PEQU , while it does not correlate with the indicators related to service information (INFOCIT and INITER) .

4.2. Average differences among the macro-areas of North, Center and South

To assess whether the subjects from northern, central and southern Italy show different mean values according to the variables taken into consideration in the present study (i.e. variables in the areas of information, quality assessment of separate waste collection services and the accuracy and frequency with which separate collection is carried out), an analysis of multivariate variance (MANOVA) was conducted, including the residence of respondents (north, center and south) as an independent variable and INFOCIT, INITER, EVAL, PEQU, ACC_WS and TIPFR as dependent variables.

The results of the analysis show a significant multivariate effect of residence variable on the dependent variables considered [F (12, 304) = 11.42, p = .00] with a considerable effect size (Eta square = .31).

Table 1. Descriptive statistics

Variables	Mean	SD	Skewness	Kurtosis	Internal consistency
INFOCIT	2.54	.94	.55	-.36	-
INITER	.77	.27	-1.06	.29	.75
EVAL	3.30	1.14	-.21	-.79	-
PEQU	2.26	.82	.55	-.40	.94
ACC_WS	7.61	1.64	-1.09	1.18	-
TIPFR	1.94	.59	.63	.37	.79

Table 2. Correlations among all variables

	INFOCIT	INITER	EVAL	PEQU
ACC_WS	.31**	.11	.19**	.13
TIPFR	.04	.0	.32**	.22**

** Correlation is significant at the .01 level (2-tailed); * Correlation is significant at the .05 level (2-tailed)

Particularly, as the results presented in Table 3 indicate, the effects of the place of residence are significant on the INFOCIT, EVAL, PEQU and ACC_WS variables, while they are not significant on the remaining two dependent variables (INITER and TIPFR). By taking into account the post hoc comparisons, conducted through the Sidak test, it emerges that the subjects from northern municipalities show higher average scores in the variables INFOCIT [Mean Difference (North vs. Center) = .91, p = .00; Mean Difference (North vs. South) = .62, p = .01], EVAL [Mean Difference (North vs. Center) = .68, p = .00; Mean Difference (North vs. South) = .99, p = .00], PEQU [Mean Difference (North vs. Center) = 1.27, p = .00; Mean Difference (North vs. South) = .68, p = .00] and ACC_WS [Mean Difference (North vs. Center) = .65, p = .09; Mean Difference (North vs. South) = .98, p = .04], if compared to those from the center and south of Italy, with significant differences for all comparisons except for ACC_WS between north and center. On the contrary, the mean scores of the subject's form center and south areas differ significantly only concerning the PEQU variable [Mean Difference (Sud vs. Center) = .59, p = .00]. Overall, these results suggest that the degree of information on separate collection services, the perceived quality of these services and the (self-assessed) accuracy with which this process is conducted by users is higher in the northern cities than in the central and southern ones. These evidences confirm the data presented in section 4.1 related to the inhomogeneous geographic distribution of urban waste collection centers in the different Italian areas. Indeed, from other studies it resulted that more

information encourage families to differentiate waste more and more efficiently. With reference to the year 2017, ISTAT provided an overview of the opinions of Italian families on the actions and policies that would increase the rate of participation in separate waste collection. To improve, both in quantitative and qualitative terms, the participation in separate waste collection, 93.4% of families would like more information on how to separate waste; the 93.3% more numerous and efficient recycling and composting centers; the 83.3% deductions and/or tax or tariff reductions, already existing in some areas of the country (Istat, 2018).

4.3. Multiple regression: predictors of accuracy and frequency of the separate collection

To evaluate the predictive capacity of the variables concerning the service-related information area (INFOCIT and INITER) and those concerning evaluation of the quality of the service (EVAL and PEQU), two regressions were conducted (the first on the accuracy criterion and the second on the frequency criterion). The resulting predictive models are shown in the Figs. (5-6). The first multiple regression analysis accounted for significant portion of accuracy of waste selection variance ($R^2 = .14$), indicated an adequate fit of the model. As shown in Table 4, the variables concerning the information (INFOCIT and INITER) significantly predict the accuracy employed by the subjects in the separate collection activities, while the variables concerning the perceived quality of the service do not offer significant contributions to its prediction.

Table 3. MANOVA as a function of city of residence

	City of residence	Mean	SD	F (2, 157)	P	Eta square
INFOCIT	North	3.14	1.01	16.75	.00	.18
	Center	2.23	.72			
	South	2.52	.94			
INITER	North	.70	.26	2.19	.12	.03
	Center	.81	.25			
	South	.76	.32			
EVAL	North	3.88	.98	8.54	.00	.10
	Center	3.20	1.12			
	South	2.89	1.09			
PEQU	North	3.10	.76	63.24	.00	.45
	Center	1.83	.51			
	South	2.42	.67			
ACC_WS	North	8.16	1.29	3.58	.03	.04
	Center	7.51	1.67			
	South	7.19	1.90			
TIPFR	North	1.93	.59	1.46	.24	.02
	Center	1.91	.55			
	South	2.13	.70			

Table 4. Regression 1: includes the variables of information and quality evaluation as predictors and the self-assessed accuracy (ACC_WS) as the criterion

Predictors	Beta	t	p (t)	R ²	F	df	p (F)
INFOCIT	.34	3.92	.00	.14	6.24	4, 155	<.001
INITER	.15	1.92	.05				
EVAL	.14	1.69	.09				
PEQU	-.08	-.87	.39				

In particular, a better perception of the information related to the service (INFOCIT) goes along with a higher accuracy in the separate collection (ACC_WS). Moreover, the more the subjects are informed about the services available in their territory (INITER), the more the accuracy of their separate collection increases, even if the result presents a tendential significance only. On the contrary, neither the overall evaluation on the usefulness and quality of service concerning the waste collection centers nor the perceived quality of the separate collection significantly predict the self-assessed accuracy of the subjects involved in the research. Fig. 5 represents these results through the first predictive model.

The second multiple regression analysis accounted for significant portion of frequency of use of the collection centers variance ($R^2 = .13$), indicated an adequate fit of the model. Moreover, as shown in Table 5, the information variables do not offer unique contributions to the frequency of use of the collection centers (TIPFR), while the variables concerning the perceived quality of the service offer significant contributions to its prediction.

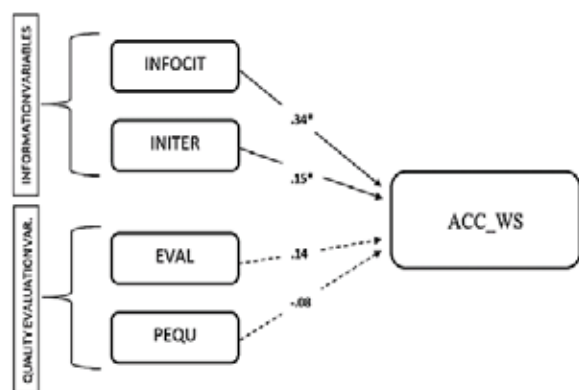


Fig. 5. Predictive Model 1

In particular, neither the perception of the information degree related to the service (INFOCIT) nor the awareness of the local initiative present in the territory (INITER) predict the frequency of use of waste collection centers by citizens (TIPFR). On the contrary, both the overall perception quality of the separate collection services (PEQU), and the overall evaluation on the usefulness and quality of waste collection centers (EVAL) significantly predict the frequency of use of the subjects involved in the research (TIPFR). Fig. 6 represents these results through the second predictive model.

The results from the multiple regressions conducted suggest that the self-reported accuracy in

the separate collection (ACC_WS) is linked to the evaluation of the information received regarding the waste management services (INFOCIT) as well as to the awareness of the local initiative present in the territory perceived by the user-citizen, while it does not seem to be associated with the overall evaluation on the usefulness and quality of the service (EVAL). At the same time, with reference to the second predictive model, the frequency of use of waste collection centers by citizens-users for the separate collection (TIPFR) results linked to the perception of the overall quality and usefulness of the services, while it seems to be independent from the quality and degree of the information received about them.

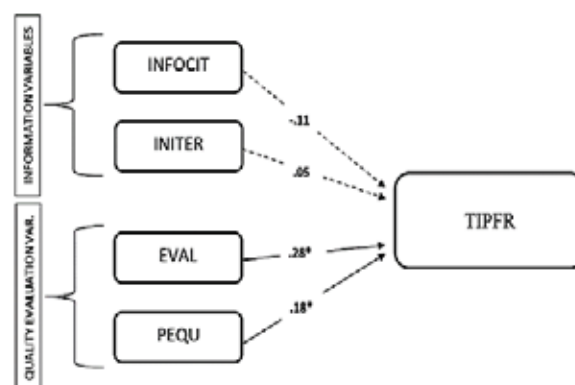


Fig. 6. Predictive Model 2

Overall, results of regression analyses suggest that the predictors of the information area offer a unique contribution in predicting ACC_WS, while they do not add anything in predicting TIPFR. Conversely, the predictors of the perceived quality of services have a unique impact on TIPFR, but do not add anything in predicting ACC_WS. In line with the literature analyzed in section 2, these results indicate that in order to improve accuracy in the behavior of separate collection by users it is necessary to adopt regulatory or incentive actions (Vergara and Tchobanoglous, 2012), as well as to provide adequate information in an appropriate way. In support of this conclusion, it can be added that the implementation of the separate collection according to the territory under examination can be very complex, requiring both adequate information tools and infrastructures for the users to carry out the separate collection carefully and to use frequently the urban collection centers. To improve this frequency, evidences indicate that users should perceive a good quality of services. This perception resulted on average higher in northern cities than in central and southern ones

Table 5. Regression 2: includes the variables of information and quality evaluation as predictors and the frequency of use of the collection centers (TIPFR) as the criterion

Predictors	Beta	t	p (t)	R ²	F	df	p (F)
INFOCIT	-.11	-1.30	.20	.13	5.58	4, 157	<.001
INITER	.05	.67	.51				
EVAL	.28	3.46	.01				
PEQU	.18	1.97	.05				

4. Conclusions

In conclusion, this study presents some significant results that offer an overview of the way separate collection services, and specifically the use of urban waste collection centers, are perceived in some important Italian cities, as well as of the perceived accuracy and frequency characterizing these settings. Particularly, it is important to highlight that the predictive models tested indicate that the accuracy seems to be influenced by the information available on waste collection services, while the frequency seems to be more closely linked to the perception of the quality of these services.

In this context, the evolution of specific regulations (i.e. EU Action Plan) is expected to lead to a strong increase in the number of collection centers, in the percentage of separate collection as well as in recycling, reuse and energy generation activities. This will also depend on the investments made by public institutions in infrastructures and information. Consequently, citizens' awareness of these issues will increase, as will their degree of knowledge and frequency of use of these services.

The present research faces also some limitations. Among those it is possible to highlight that participants are recruited from only six municipalities, although selected as the most populated and located three main macro-areas of the country (north, center and south Italy), limiting the generalizability of results. Moreover, the limited number of participants may have reduced the statistical power of the study. Finally, the lack of objective evaluations to measure the variables included in the model may have biased our results.

Future studies are called to make a comparison between the perception of citizens-users and objective measures relating to the abovementioned three dimensions. Furthermore, the enlargement of the sample, together with the investigation of additional municipalities sited in different areas, would improve the power of statistical tests and the external validity of the results.

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SUSTAINABILITY ASSESSMENT OF TWO DIGESTATE TREATMENTS: A COMPARATIVE LIFE CYCLE ASSESSMENT

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Abstract

Digestate derived from the anaerobic digestion of biowaste is a nutrient-rich substance whose direct use on land is not permitted by the Italian Legislation. The possibility of recovering its nutrients can be given by the processes of stabilisation and sanitation required by the Italian Legislation. Among these processes, composting and calcium hydrolysis with neutralization (CHN) permit to obtain useful soil improvers like compost and defecation gypsum (DG). In this paper a gate-to-gate Life Cycle Assessment (LCA) of these two processes is performed to evaluate their relative environmental sustainability, by using the ReCiPe H midpoint and endpoint impact assessment methods. The functional units (FUs) used in this analysis are one tonne of digestate treated by each process, and the amount of compost and DG necessary to amend one hectare of maize cultivation. Data used in the assessment were collected from plants located in Northern Italy and were referred to one year of operation. The processes of transport and spreading on land of the final products were not considered. The results of both the analyses show that CHN is the process with the largest environmental impacts, mainly due to the use of chemicals (i.e., sulfuric acid and calcium oxide). For both processes and FUs, the most impacted midpoint categories are Natural land transformation, Marine ecotoxicity and Freshwater ecotoxicity. Among the endpoint categories Resources is the most impacted one (followed by Human Health and Ecosystems), for both FUs, although showing larger differences for the agronomic use.

Key words: anaerobic digestion, circular economy, digestate treatment, LCA, sustainability assessment

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1. Introduction

In Italy the number of mechanical-biological processes for the urban waste treatment, before their definitive disposal or recovery, is increased in last years. In 2017, 340 out of 644 urban waste treatment plants were dedicated to the treatment of Organic Fraction in Municipal Solid Waste (OFMSW), 130 to mechanical or mechanical-biological treatment and 123 to landfills, while the others consisted in incinerators and industrial co-incinerators (ISPRA, 2018). During the last ten years, anaerobic digestion (AD) was implemented in several wastewater treatment plants as co-digestion of the OFMSW and

waste activated sludge (WAS) (Tyagi et al., 2018). This approach allows both to increase wastewater treatment efficiency and to optimize the energy recovery. The AD liquid effluent, obtained after solid-liquid separation, is usually recycled back into the wastewater treatment plant, while the solid part is sent mainly to composting plants.

Considering the constant increase of urban waste production and consequently the increase in the amount of organic fraction sent to biological treatments, finding appropriate and sustainable processes to manage the digestate generated by AD of biowaste is urgently needed. In Italy, 55% of organic waste is treated by composting, 5% by AD and 40%

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by combined aerobic/anaerobic treatment. Among the waste sent to composting, 33% is composed of green organic materials, 50% of OFMSW, 12% of sewage sludge (SS) and the remaining 8% of other waste including digestate from AD (ISPRA, 2018).

To address the goals of the European Union “Circular Economy Action Plan” (EC Directive, 2015a), and the proposal of the European Commission on waste management (EC Directive, 2015b), the use of digestate as fertilizer has become an interesting practice to both reduce the amount of waste to landfill and close the nutrient cycles.

In Italy, the practice of spreading sewage sludge on agricultural land is widely adopted (Collivignarelli et al., 2015), as well as of spreading digestate deriving from the AD of biomass (agricultural, zootechnical, etc.) that can be directly applied on field or used as basis for fertilizers’ production (DM, 2016). In the case of digestate derived from AD of biowaste this direct use is not permitted by the Italian Legislation as the digestate needs further processes of stabilization and sanitation (L.D., 2010) to inactivate pathogens for its safe release.

Currently, in the European Union a common regulatory framework that defines the general rules and guidelines for the sustainable management of digestate produced by anaerobic treatment of organic waste does not exist. In order to provide the scientific background in support of legislative decisions, it is therefore crucial to assess the potential environmental impacts of the technological processes related to different management scenarios of this by-product.

Currently, in Italy, digestate from biowaste can be treated by several processes; the main alternatives are: treatments for organic soil improvers’ production, like composting and calcium hydrolysis; heat treatments, like incineration and pyrolysis for energy and biochar recovery; landfilling (Oldfield et al., 2018, Vázquez-Rowe et al., 2015); and processing in wastewater treatment plants (Di Maria and Sisani, 2019). Among these technologies, calcium hydrolysis with neutralization (CHN) is adopted to produce the so called “defecation gypsum” (DG), usable as soil improver and alkaline-controller, while composting leads to the production of compost.

Several LCA studies analysed the compost production (as result of biowaste treatment) and its land application, also as AD post-composting step (Bernstad and la Cour Jansen, 2011; Blengini, 2008; Cremiato et al., 2018; De Feo et al., 2016; Di Maria et al., 2016; Di Maria and Micale, 2015; Jensen et al., 2017, Neri et al., 2018), while no studies, to the best of our knowledge, can be found in the literature about DG production.

This paper contributes to the environmental impact assessment of digestate treatments, filling the gap in the analysis of the DG production and comparing its environmental performances with the more known and widely adopted composting process.

2. Material and methods

2.1. Goal and scope definition

In this paper a preliminary analysis of the potential environmental impacts at a global scale of composting and CHN of dehydrated digestate from biowaste is performed, by using the LCA (Life Cycle Assessment) method according to the ISO 14040 (2006) and ISO 14044 (2006) standards. This analysis was performed to evaluate the environmental performance of these two processes, and consequently understand which is the best alternative for the digestate treatment and which are the phases, for each process, most impacting human health and the environment.

The processes considered are: composting performed by the Energia Territorio Risorse Ambientali (ETRA) S.p.a. plant located in Veneto Region (Italy), where dehydrated SS coming from the civil wastewater treatment plant is processed together with green organic materials; and calcium hydrolysis performed by a plant located in Lombardy Region, treating SS and dehydrated digestate. In the LCA analysis, dehydrated SS and dehydrated digestate were assumed to be comparable, as supported by the physico-chemical characterisation results presented in Section 3. The LCA performed adopts a “gate-to-gate” approach. In fact, since the main input materials for both plants are coming from different locations (e.g. green organic materials collected in different villages) or treatment plants, their production was not included in the analysis, as well as the transportation and spreading phases, which are assumed to be similar for both compost and DG and not providing a relevant contribution. In fact, previous studies (Bernstad and la Cour Jansen, 2011; Blengini, 2008) showed that transport operations play a minor role in the overall environmental impact (e.g., providing the lowest contribution, except for Ozone layer depletion, in Blengini (2008), due to the fact that the involved distances are usually reasonably short (an average of 25 km). As far as the construction of the plant is concerned, it was not included in the analysis because both the plant buildings have a long lifetime and consequently its contribution to the environmental impact assessment of one year of plants operation would not be so relevant. In addition, data about maintenance operation of the plants were not available and therefore they were not included in the analysis. Data on soil occupation for the composting plant were also not available since the composting process is integrated into a much larger wastewater treatment plant area and therefore they were not included in the analysis (although the information was available for the CHN plant). Finally, in the composting process the production of fine compost was not considered as it is optional.

The Functional Unit (FU) of this analysis is one tonne of digestate treated by each plant during one

year of plant operation. In addition, to assess the environmental performance of the soil amendments produced, the amount of product necessary to improve one hectare of maize cultivation was considered as second FU. Primary data of material, fuels and energy consumptions were collected directly from the plants or calculated from assumptions made by the authors, while for secondary data the database Ecoinvent v 3.3 (Wernet et al., 2016) was adopted. Data were elaborated by means of SimaPro 8.3 software and the selected treatment options were compared following the ReCiPe v.1.13 assessment method (Goedkoop et al., 2013), that quantifies the environmental impacts on 18 categories at the midpoint level, and 3 at the endpoint level. This method represents the highest level of convergence with the ILCD recommendations (ECJRC-IES, 2011). The midpoint approach was adopted in this study to evaluate the 18 impact categories individually under a Hierarchist perspective, which consider average conditions in terms of time horizon, pessimistic or optimistic point of view, type of effects considered etc.

Although normalisation is an optional step under ISO 14044:2006, it was applied to the midpoint results in order to support the interpretation of the two processes' impact profiles. This additional step is frequently adopted in other LCA studies on composting processes (e.g., in Di Maria et al. (2015) and (2016)). Normalization reports the characterized results of the impact categories at the same scale and consequently it allows evaluating the relative magnitude of potential impacts. The normalization set selected in this study is Europe ReCiPe H, 2000, which refers to the environmental impacts of Europe in 2000 (Sleeswijk et al., 2008).

3. Case studies

The physico-chemical characteristics of the dehydrated digestate treated by the DG production plant are reported in Table 1 together with the characteristics of the dehydrated SS treated by the composting plant. Since the two materials show similar characteristics, we could assume in the LCA analysis that the same composting process used to treat SS can treat digestate without significantly affecting input and output parameters.

3.1. The composting plant

The composting plant analysed in this study is part of the ETRA S.p.a. biotreatment centre located near Padova, in Northern Italy, where urban wastewater is treated and the derived SS is composted together with branches and green organic material, reaching a total amount of treated material of approximately 20 000 tonnes per year. For the purpose of this study, that is to analyse the treatment processes of digestate, SS was replaced by digestate (as justified by Table 1). Therefore, we considered branches, green organic materials and digestate as input materials for this plant.

The composting process (Fig. 1) starts with the storages of the digestate arriving to the plant, which occurs in a depressed shed, where the exhausted air is extracted by four fans. This building has a maximum capacity of 1 000 m³ and is also used for the storage of mature compost and the shredded green organic material before its processing. Digestate is combined with green organic materials to reach the C:N ratio (25:1-30:1) and moisture content (~60%) optimal to the composting process.

In the first phase, called "accelerated bio-oxidation", the degradation of organic matter takes place thanks to the accelerated metabolism of decomposing microorganisms, with the consequent production of odorous emissions and heat. In the centre of the bio-oxidation shed the input material is loaded with mechanical shovels, alternating layers of digestate and lignocellulosic material in appropriate ratio, according to the chemical composition of digestate. During this phase air is supplied in the mixture through nine fans and overturning is performed with an automatic turning machine. The high temperature reached by the decomposing materials (~70°C) allows to sanitize the mixture by removing weed seeds and pathogenic microorganisms. The mixture is also ventilated to remove the excess of moisture, by means of some grids placed on the floor of each lane, under the piles.

The air removed from the low-pressure area and the bio-oxidation locals is treated by biofilters before being emitted in air. These filters are made of wood chips of 20-40 mm and of wooden root of about 40 cm. These materials are also enriched in granular lime. When the bio-oxidation process ends, after thirty days from its beginning, the fresh compost is transferred to an external lot through a mechanical blade machine and the screening phase takes place. This phase consists in the mechanical separation of organic particles with diameter lower than 20 mm (i.e., undersize materials), from the remains (i.e., oversize materials), consisting on pieces of wood and small amount of plastic that is typically used for landfill coverage.

The undersize material continues its processing in the maturation phase, which occurs outdoors for eight weeks. The fresh compost is accumulated to favour the aeration and the outflow of rain. Periodically, the compost in maturation is analysed in its content of humidity, pH and temperature until its complete maturation. Another additional (and optional) step is the refining process, where a vibrating screen with 10 mm meshes separates the fine compost, that can be sold as improver for plant nurseries, farms or individuals, from the coarse one.

3.2. The CHN plant

The CHN plant analysed in this study is located in the Lombardy Region, in Northern Italy, and it carries out the recovery of a total amount of about 50 000 tonnes per year of SS and digestate, converting them into DG.

Table 1. Physico-chemical characteristics of the dehydrated digestate treated by the calcium hydrolysis plant and the dehydrated SS treated by the composting plant. (TKN=total Kijendhal Nitrogen, TP= total phosphorus COD = Chemical Oxygen Demand, TOC = Total Organic Carbon, DM = Dry Matter)

Parameter	Unit	Dehydrated digestate	Dehydrated SS
pH		8.17	7.22
DM (%)	%	23.31	21.58
TKN	%DM	5.33	5.79
NH ₄ ⁺	%DM	0.86	0.13
TP	%DM	2.40	2.02
COD	%DM	80.39	83.31
TOC	%DM	42.09	37.41

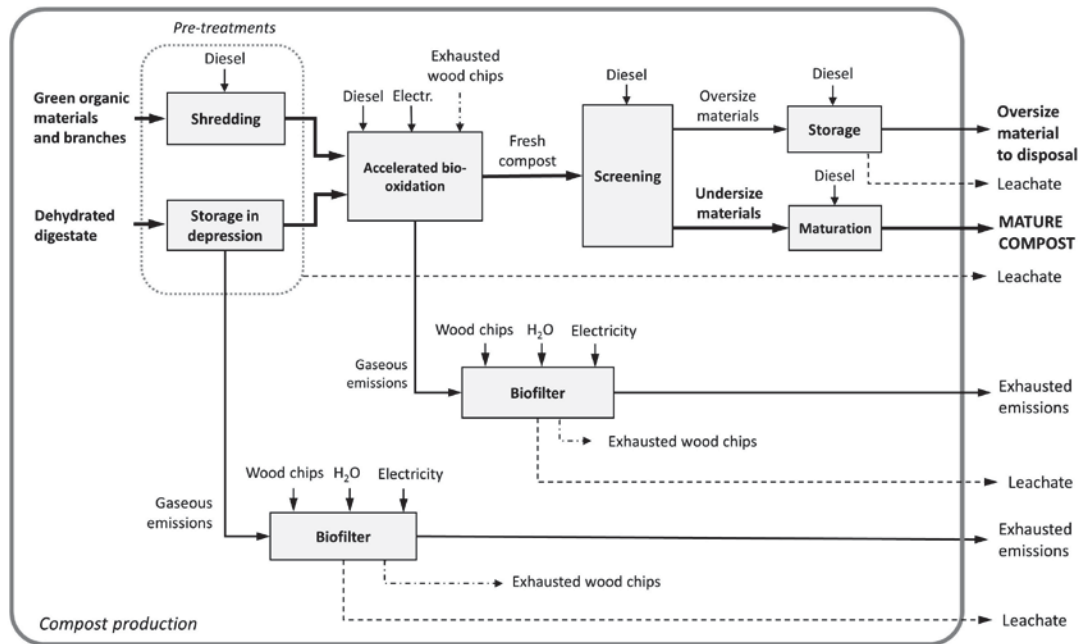


Fig. 1. Flow diagram of composting process including the emission treatment

This product is composed by CaO and SO₃, for 20% and 15% of the total dry weight respectively, and its use is recommended before the soil's ploughing operations as soil improver, calcium and sulphur provider and alkalinity corrector. The SS (around 95% of the total input) and the dehydrated digestate (about 5% of the total) are the main waste treated in this plant and are always checked for physico-chemical characteristics before starting the treatment and during the process.

In Fig. 2 the flow diagram of the DG production phases is reported. The matrices accepted by the plant are temporary stocked in closed tanks equipped with suction systems and mixed by an excavator. If necessary, a small percentage of liquid sludge and water is added to fluidify the mixture. Afterwards, the mixture is transferred by a telescopic blade in another tank where calcium sulfide (CaSO₄) starts the alkaline hydrolysis process.

The alkaline chemical products are then neutralized by the hydrochloric acid (HCl). At the end of this process the DG is ready and is transferred by gravity in another storage tank where it loses moisture

by evaporation. If the lot is not compliant to the parameters fixed by the company, it will be processed again. In order to contain the emissions from the chemical processes that occur during the DG production, all phases take place in closed, low pressure areas. The intake air is conveyed in a dedicated treatment emission plant that consists in a wet scrubber, an expansion chamber and a biofilter. The main pollutants intercepted are ammonia, odorous emissions, volatile organic compounds and HCl.

3.3. Inventory

Data input and output of the systems depicted in Fig. 1 and Fig. 2 include energy and matter flows for each process unit (Tables 2, 3), that were collected directly from the plants or estimated by the authors. Water consumption by composting biofilters have been estimated from the consumption of water by DG production's biofilters. All phases of both processes take place in closed buildings and therefore the only emissions included were those coming out from biofilters, which were considered as addressing the emissions legal limits (L.D., 2006).

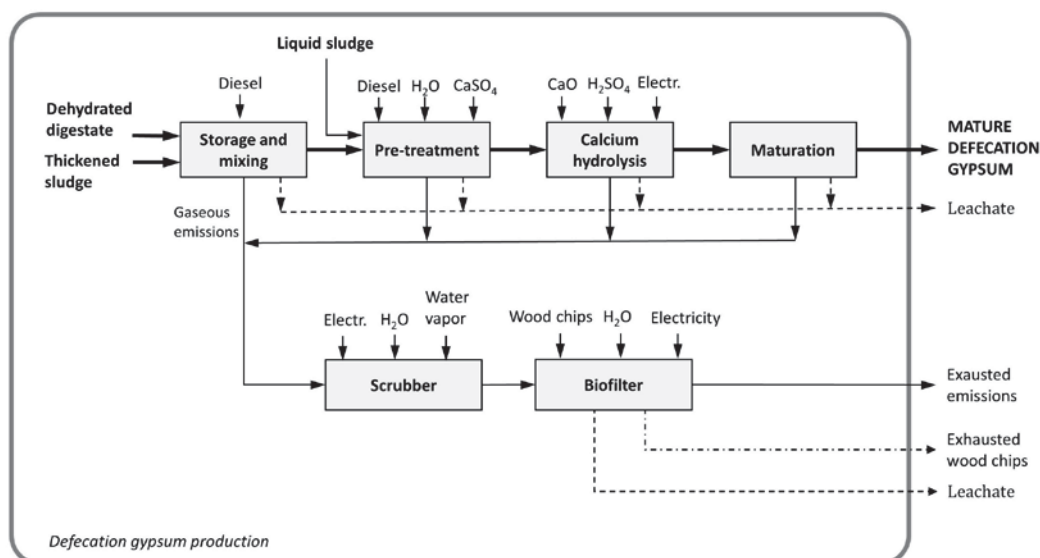


Fig. 2. Flow diagram of DG production including the emission treatment

The product quantities considered for the impact assessment according to the second FU were those necessary to amend one hectare of maize cultivation, by considering their production only (not including their transport and spreading). The amount of compost considered was obtained by agronomical tests on Italian sites, that are consistent with average values present in the literature (www.venetoagricoltura.org). Regarding the DG, the value considered came from a specific test made by the DG producer (Agrosistemi S.r.l., private communication) as in literature, to the best of our knowledge, there are not studies on this product and further analysis would be needed to better understand its amendment power and security. From the collected data (reported in Table 4), it emerged that about the same amount of compost and DG is necessary to amend one hectare of soil for maize cultivation, although DG does not provide any contribution to phosphorous and potassium content, thus requiring additional input of these mineral fertilizers.

However, it must be noted that sometimes also amendment with compost requires an addition of these minerals since the compost nutrients content strongly depend on the characteristics of the materials used as input to the composting process.

4. Results and discussion

Fig. 3 reports the LCA comparison of compost and DG production, calculated by using the ReCiPe Hierarchist midpoint method with normalization of results. The comparison shows that the highest environmental burden is associated to DG production, where Natural land transformation, Marine ecotoxicity and Freshwater ecotoxicity are the most impacted categories. This result can be explained by the use of some chemicals (i.e., sulfuric acid and calcium oxide) in the CHN process, which cause the most relevant contribution to the three impact categories (Figs. 4-6).

Table 2. Total annual input and output of composting process including the emission treatment

<i>Input</i>	<i>Amount</i>	<i>Unit</i>
Dehydrated digestate	3 460	tonnes year ⁻¹
Green organic materials and branches	15 720	tonnes year ⁻¹
Diesel	40 080	L year ⁻¹
Electricity	1 070 195	kWh year ⁻¹
Wood chips	1535	m ³ year ⁻¹
Water	55.5	m ³ year ⁻¹
Output	Amount	Unit
Compost	5 560	tonnes year ⁻¹
Dust	0.27	mg Nm ⁻³
Ammonia	0.57	mg Nm ⁻³
Sulfuric acid	0.57	mg Nm ⁻³
VOC	0.11	mg Nm ⁻³

Table 3. Total annual inputs and outputs of defecation gypsum production including the emission treatment

<i>Input</i>	<i>Amount</i>	<i>Unit</i>
Thickened sludge	47 500	tonnes year ⁻¹
<i>Dehydrated digestate</i>	2 500	tonnes year ⁻¹
Liquid sludge	1 280	tonnes year ⁻¹
Diesel	18 500	L year ⁻¹
Electricity	317 100	kWh year ⁻¹
Calcium sulphate	2 500	tonnes year ⁻¹
Sulfuric acid	7 500	tonnes year ⁻¹
Calcium oxide	7 500	tonnes year ⁻¹
Wood chips	160	m ³ year ⁻¹
Water	1 300	m ³ year ⁻¹
Output	Amount	Unit
Mature defecation gypsum	31 135	tonnes year ⁻¹
Water vapor	14 650	tonnes year ⁻¹
Ammonia	0.6	mg Nm ⁻³
VOC	1.5	mg Nm ⁻³
Mercaptans	0.5	mg Nm ⁻³
Hydrogen sulphide	0.1	mg Nm ⁻³
Exhausted wood chips	100	m ³ year ⁻¹

Table 4. Quantity of compost and DG necessary to amend one hectare of soil for maize cultivation and respective nutrients content. Values in brackets are from additional mineral fertilizers

<i>Soil improver</i>	<i>Amount (kg/ha)</i>	<i>N (kg/ha)</i>	<i>P₂O₅ (kg/ha)</i>	<i>K₂O (kg/ha)</i>	<i>Yield (kg/ha)</i>
Compost	33 000	116	243	232	10000
Defecation gypsum	32 400	227	(0.79)	(0.4)	10000

On the other hand, for the composting process the impacts to these categories, despite being much lower than those of DG, are linked to the electricity consumptions of fans, windrows and grinder. Comparing the composting process and the anaerobic digestion of biowaste, Blengini (2008) shows high environmental impacts for the categories Acidification and Nutrient enrichment caused by the biogenic emissions from the aerobic degradation process and remarkable Gross Energy Requirement of compost process. However, as highlighted in the review by Bernstad and la Cour Jansen (2012) about LCA of different treatment systems, estimated impacts vary largely among different studies, due to the different setting of the system boundary methodology (e.g., including or not transport), the selected methodology (e.g., different impact assessment methods, including or not avoided impacts), and the variation on input data (e.g., the characterization of the treated biowaste). These considerations lead to some difficulties in comparing our study with others in the field and suggest the need to establish more detailed guidelines to improve such situation.

To assess the environmental performance of the two processes in relation to the amendment power of compost and DG, the amount needed to improve one hectare of maize cultivation was considered. The impact profile (Fig. 7) shows again that for the category Natural land transformation the higher impact is clearly associated to DG, while for other

categories like Freshwater eutrophication, Freshwater ecotoxicity and Marine ecotoxicity the production of compost is providing a slightly higher contribution compared to DG. Finally, looking at the endpoint impacts on Human health, Ecosystems and Resources (Fig. 8), DG production shows the higher impacts according to both our FUs.

However, it must be noted that these are the results of a preliminary “gate-to-gate” LCA, where processes that occur before and after the production of compost and DG (like construction and maintenance of the plant and transportation of waste material) were not considered. By expanding the systems’ boundaries, maintenance of the plant could play a relevant role in the generation of environmental impacts while the contribution of transportation of waste material to treatment plants should be negligible if the digestate treatment takes place in the proximity of its production (which is usually the case as reported by Bernstad and la Cour Jansen (2011) and Blengini (2008)).

Moreover, results could be affected by including additional information which were not available to the authors. The most relevant case would be the possibility to consider the portion of land covered by each phase of the two treatment processes. In fact, it is known that for the composting process large areas are usually needed and this could impact the Natural land transformation category in favour of CHN process.

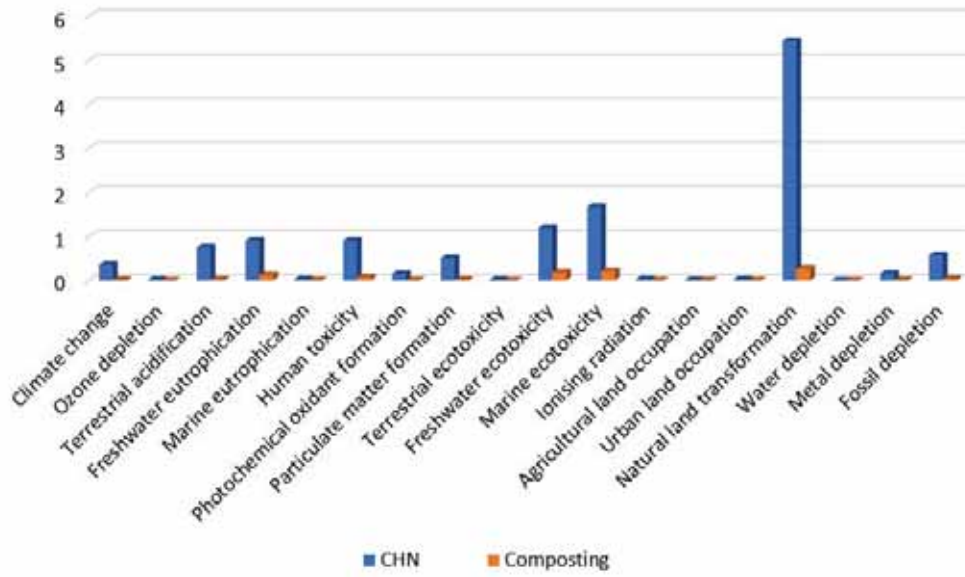


Fig. 3. Comparison of normalized results for the treatment of 1 tonnes of digestate with CHN and composting processes

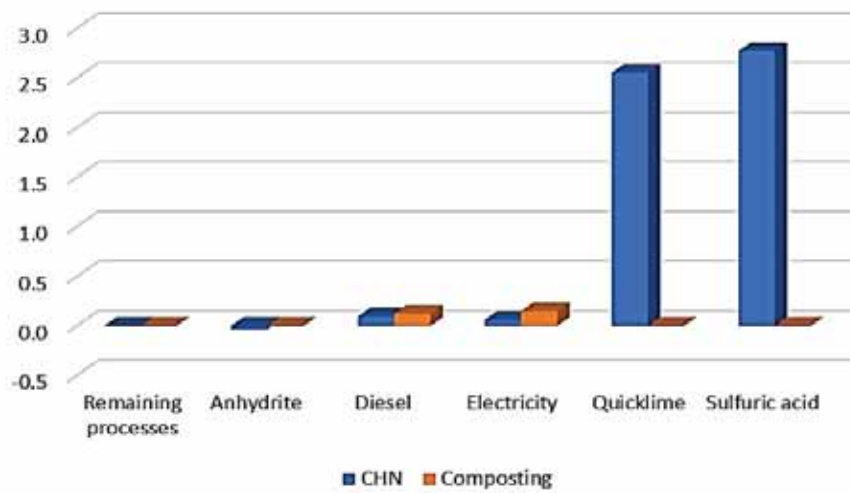


Fig. 4. Most impacting processes on Natural land transformation category

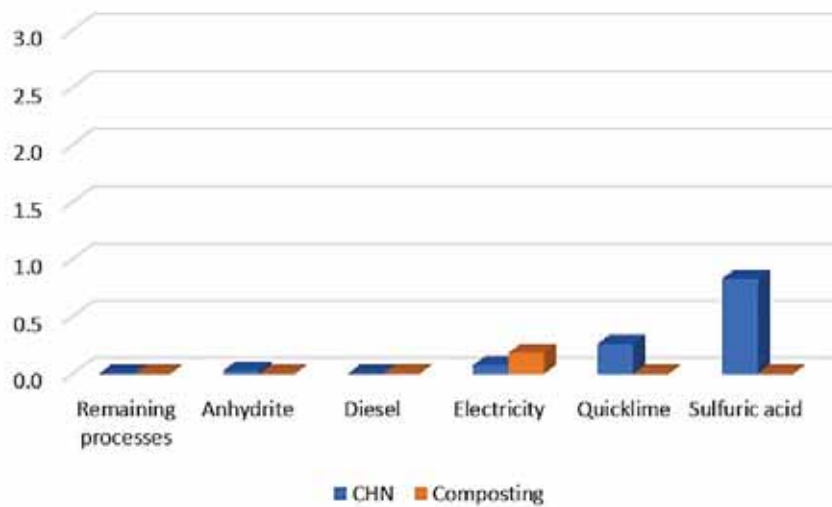


Fig. 5. Most impacting processes on Freshwater ecotoxicity category

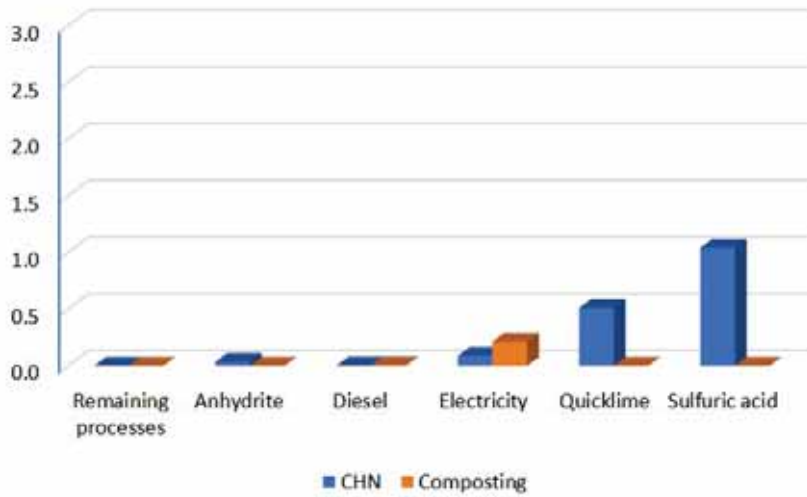


Fig. 6. Most impacting processes on Marine ecotoxicity category

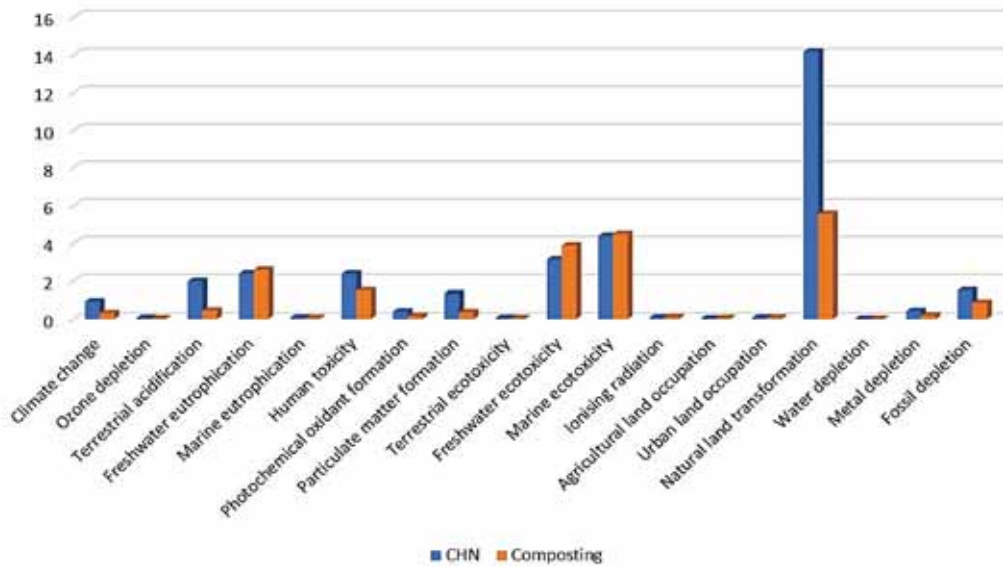


Fig. 7. Comparison at midpoint level between CHN and composting, according to the agronomic utilization

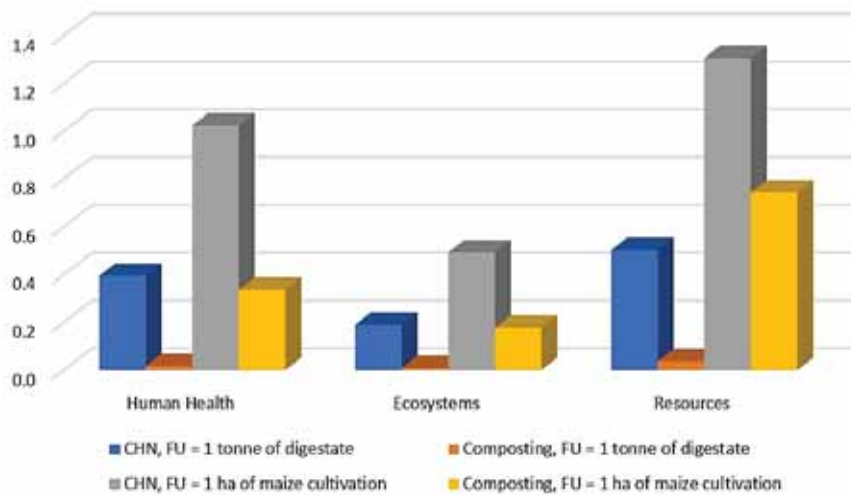


Fig. 8. Comparison at endpoint level between CHN and composting, according to the two FUs used in the study

5. Conclusions

In this paper the environmental impacts of the treatment of waste-digestate by composting and CHN processes are reported. These processes are suitable alternatives to landfill or incineration, and lead to the production of useful soil improvers (i.e., compost and DG).

Although the gate-to-gate LCA results indicate that CHN is responsible for higher environmental impacts compared to composting at both midpoint and endpoint levels and according to both our FUs, the following additional observations can be made. The composting process considered in this study lasts about three months, while the CHN process takes place in about thirty days.

This means that, with the same processing capacity, CHN can stabilize a higher amount of organic waste in the same time frame, thus speeding up the overall waste treatment process and therefore leading to an economic benefit.

On the other hand, improvements could be envisaged also for the composting process. Indeed, digestate is the effluent of an anaerobic stabilization process intended to remove the organic substance by converting it to carbon dioxide and methane; therefore, its characteristics can vary significantly as the input matrices and the operating parameters set (e.g. retention time and organic load) change. This means that knowing the characteristics of the specific digestate would allow to optimize the composting treatment phases in terms of duration as well as materials and energy use.

As an example, the duration of the oxidation phase could be reduced according to the quantity of putrescible matrix in the digestate. Therefore, an enhanced monitoring of the physico-chemical and biological parameters of the digestate along with the possibility to set up flexible composting processes, tailored to the input needs, would allow to end up with more environmentally sustainable solutions.

Acknowledgements

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TECHNOLOGIES FOR THE CONTROL OF EMERGING CONTAMINANTS IN DRINKING WATER TREATMENT PLANTS

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Abstract

In recent years, so-called emerging contaminants (ECs) have attracted growing interest as they have been detected in reservoirs and even in drinking water. The new proposal *Drinking Water Directive* (2018) provides for the introduction of new parameters relevant to the ECs category. As these contaminants today tend to be present in a growing number of water sources, provide treatment systems that ensure compliance with regulatory limits and the protection of public health has become essential. The aim of this paper is to provide essential information on five ECs (more precisely: haloacetic acids, microcystine-LR, Perfluoro Alkylated Substances, Bisphenol-A and Nonylphenol) and to explain useful processes for their removal in a DWTPs. For each contaminant, current and future legislation, health aspects and in particular a focus of the chemical and physical removal technologies already existing and under study are reported. The effectiveness of both conventional (e.g. chemical oxidation, coagulation/flocculation, adsorption on Granular Active Carbon (GAC), ion exchange) and advanced treatments (e.g. membrane filtration, AOPs) is presented and discussed.

Keywords: bisphenol, haloacetic acids, microcystins, nonylphenol, PFAS

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1. Introduction

In recent years, so-called emerging contaminants (ECs) have attracted growing interest. Teodosiu et al. (2018) reported the definition of ECs as substances detected in the environment but currently not included in routine environmental monitoring programmes and which may be candidate for future legislation due to their adverse effects and/or persistency. In addition, most ECs have been discharged into the environment for years, but their presence has only recently begun to be investigated (Dulio et al., 2018) or are only recently recognized as potential causes of adverse effects on ecosystems or humans (Houtman, 2010). Most of this ECs are not yet included in the current drinking water legislation (e.g. *Directive 98/83/EC of the European Council* (EC,

1998)) (Riva et al., 2018). ECs include more than 1000 substances (Teodosiu et al., 2018) such as pharmaceuticals, endocrine disruptors, perfluorinated compounds (PFAS), cyanotoxins, haloacetic acids (HAAs), surfactants, plastic products, chromium VI, emerging DBPs, radioactivity, 1,4-Dioxane and new pesticides (Adamson et al., 2017; Petrovic, 2003; Sharma et al., 2019; Sillanpää et al., 2018b; Ternes et al., 2015). ECs have been detected in reservoirs and even in drinking water and are today increasingly object of research thanks to better existing analytical techniques and new toxicological evidence (Khatibikamal et al., 2019; McCleaf et al., 2017; Sillanpää et al., 2018b; Westrick et al., 2010).

Based on the indications provided by the most recent World Health Organization guidelines (WHO, 2017a), the new proposal for a *Drinking Water*

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Directive (EC, 2018), hereinafter called DWD 2018, provides parametric values for some of the over 1000 ECs. Firstly, among the ECs inserted in the DWD 2018 proposal, those that were discussed in this paper were selected considering ECs classified as possible carcinogens (class 2B), namely: (i) HAAs-, more precisely Dichloroacetic acid (IARC, 2014); (ii) microcystin-LR (MC-LR) (IARC, 2010); (iii) Perfluoro Alkylated Substances (PFAS), more precisely Perfluorooctanoic Acid (IARC, 2017). Furthermore, among the endocrine disruptors (EDCs), the other contaminants analysed in this paper were Bisphenol-A (BPA) and Nonylphenol (NP) due to their health effects and their presence in natural and treated water (WHO, 2017a).

Regarding the five selected compounds, the new proposal of DWD 2018: (i) introduces the limit of 80 µg/L for the HAAs; (ii) indicates a limit for MC-LR of 1 µg/L according to the WHO suggestion; (iii) regulates the PFAS and Total PFAS with two new proposed limit of 0.10 µg/L and 0.5 µg/L respectively; (iv) prescribes 0.01µg/L as limit for BPA and finally regulates for the first time the NP with a limit of 0.3 µg/L. In order to comply with the requirements that will be provided by the new proposal for a *Drinking Water Directive* (EC, 2018), the following solutions can be applied: (i) identify new water sources of better quality, (ii) optimize the management of Drinking Water Treatment Plants (DWTPs) (Sorlini et al., 2015b) and (iii) identify new treatment solutions for ECs removal.

As previously reported, these contaminants tend to be present today in a growing number of water sources; therefore, providing treatment systems that ensure compliance with regulatory limits and the protection of public health becomes essential (Ternes et al., 2015). The ECs can represent a problem in DWTPs because their removal cannot always be efficient with conventional treatments (Simazaki et al., 2015). The conventional treatments include adsorption on activated carbon, sand filtration, ion exchange and chemical oxidation. In order to cope with the recalcitrant ECs, advanced treatments are also available. Among these processes, there are the

advanced oxidation processes (AOPs), membrane filtration and biological processes. In Fig. 1, a list of the treatments presented in the paper is reported.

In this paper, specific insights regarding HAAs, MC-LR, PFAS, BPA and NP are presented. For each parameter, the origin, the adverse effects on human health and some considerations about the presence in the environment are presented. Moreover, the removal technologies already existing and under study are also reported.

2. Emerging contaminants (ECs)

2.1. Haloacetic acids

The HAAs are a group of compounds of organic nature that are formed as disinfection by-products (DBPs) following the presence of Natural Organic Matter (NOM) in waters subjected to chlorination process (Collivignarelli et al., 2017; Wang et al., 2017). They are one of the largest groups of water DBPs, based on weight, and represent (with THMs) over 50% of total halogenated DBPs (EPA, 2018; WHO, 2017a; 2017b). The main factors influencing the formation of HAAs are: (i) the temperature, (ii) the pH, (iii) the contact time, (iv) the presence and concentration of halogen ions (e.g. Cl⁻, Br⁻ and I⁻) and (v) the presence of NOM (EPA, 2018; He et al., 2018; Postigo and Zonja, 2018; Xue et al., 2017).

HAAs are considered cytotoxic and genotoxic (Dad et al., 2018; Postigo and Zonja, 2018). The route of exposure to these contaminants is commonly the ingestion of drinking water containing HAAs. According to the Environmental Protection Agency (EPA, 2018), inhalation and dermal exposure can also occur but, given the chemical properties of these compounds (i.e. low volatility and high polarity), these exposure paths have a limited impact on the population. In the United States, according to EPA (2018), most of the population is exposed to HAAs. Referring to WHO (2017b), evidence of carcinogenicity to health is not completely clear for all HAAs compounds.

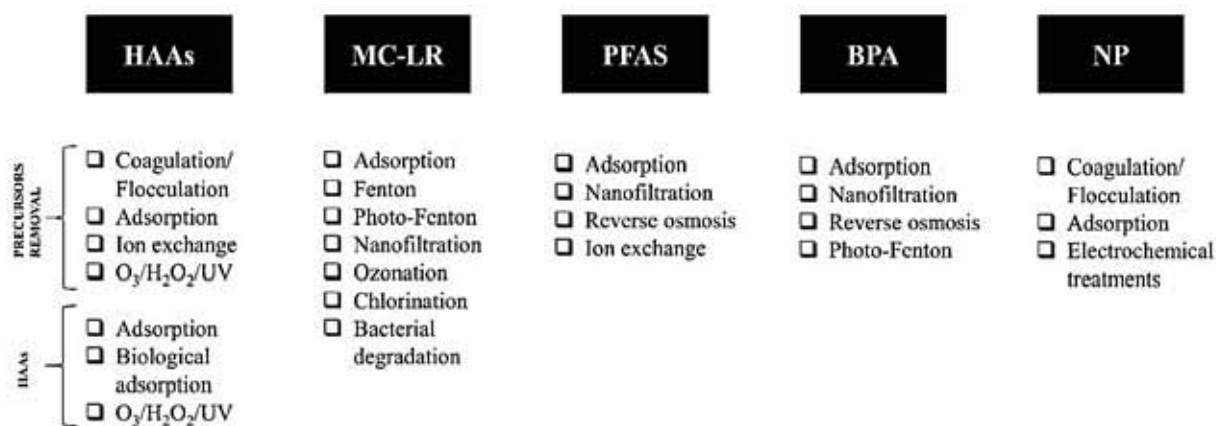


Fig. 1. List of the treatments suitable for ECs removal in DWTPs, presented in this paper

For some compounds, the possible carcinogenicity is demonstrated; for example, dichloroacetic acid is classified 2B by IARC. For others, e.g. monochloroacetic acid and trichloroacetic acid, there is still no direct evidence of carcinogenic properties on humans. For the bromoacetic acids, insufficient data are available for health effect classification.

Given the effects on public health, the World Health Organization (WHO, 2017a) suggested in its guidelines a limit of 80 µg/L for the sum of nine different compounds considered representative (mono-, di- and trichloroacetic acid, mono- and dibromoacetic acid, bromochloroacetic acid, bromodichloroacetic acid, dibromochloroacetic acid and tribromoacetic acid). The new DWD 2018 (EC, 2018) fully incorporates the suggestions provided by the WHO (WHO, 2017a) and introduces this new parameter in Part B concerning chemical parameters.

In order to reduce the concentration of HAAs in drinking water, the types of intervention are essentially: (i) removal of precursors before disinfection, (ii) modification of disinfection operating practices and (iii) removal of HAAs after their formation (EPA, 2018). With regard to the removal of precursors, many techniques are available. For example, coagulation, by means of iron and aluminium based reagents, allows to eliminate on average between 20% and 75% of NOM and Dissolved Organic Carbon (DOC), precursors in the formation of HAAs (Sillanpää et al., 2018b; Zheng et al., 2015). Furthermore, the ion exchange technique shows excellent results in the field of precursor removal, with removal yields between 50% and 70% (EPA, 2018; Finkbeiner et al., 2018); higher values are obtained if this technique is combined with coagulation. In this case the coagulant consumption can be reduced up to 50% (Metcalf et al., 2015) and removal of up to 80% of the precursors can be achieved (EPA, 2018). Moreover, HAAs precursors can be removed also by AOPs, in particular O₃/H₂O₂/UV (Sillanpää et al., 2018a). The main objective of this treatment is oxidizing the organic matter by high reactive and non-selective species (OH•) produced in the process (Giannakis et al.,

2016). In this case the HAAs formation potential can be reduced up to 70% (Sillanpää et al., 2018a).

Regarding the operational changes, pre-oxidation with chlorine could be replaced by potassium permanganate, hydrogen peroxide or ozone (EPA, 2018). Another alternative may be to replace chlorination with chloramination that reduces the formation of HAAs but the production of N-DBPs (nitrogenous disinfection by-products) is increased (Bond et al., 2011). Also, the chlorine dioxide (ClO₂) if used in the presence of NOM does not lead to the formation of HAAs but can however form other DBPs such as chlorites and chlorates (Sorlini et al., 2015a, 2016). UV rays are also excellent candidates for complete or partial replacement of chlorine (Wang et al., 2015). However, they have no persistence and are therefore unable to provide coverage in the water distribution network (EPA, 2018).

Finally, as suggested by EPA (2018), another alternative is to continue to use chlorine as an oxidizing agent and to remove HAAs after their formation by, for example, filtration using biologically active (EPA, 2018) or not biologically active (Jiang et al., 2017) granular activated carbon (GAC). Jiang et al. (2017) studied the adsorption on AC of chlorinated water according to a different approach from the traditional one in which the precursors of the formation of DBPs are removed. DBPs, including the HAAs, are removed only once they have been formed. Their results indicate that the new approach is substantially more effective in controlling halogenated DBPs than the traditional approach. Also, in this case, as reported by Matilainen and Sillanpää (2010), the application of O₃/H₂O₂/UV can be effective (50-90%) in HAAs reduction if applied in DWTPs. They also explained that the presence into drinking water of humic acids can interfere with the decomposition of HAAs by the process reducing the yields by 20-40% (Matilainen and Sillanpää, 2010). In fact, humic acids could cause the H₂O₂ accumulation and the decrease in rate constants of HAAs decomposition (Wang et al., 2009). In Table 1, examples of suitable treatments for HAAs and precursors removal in DWTPs are reported.

Table 1. Examples of suitable treatments for HAAs and precursors removal in DWTPs. ^a: ADS= adsorption, CF= coagulation/flocculation, IE= ion exchange. ^b: L= laboratory scale, F= full-plant scale. ^c: DW= drinking water; R= real drinking water, S= synthetic drinking water. ^d: PAC= powdered activated carbon; GAC= granular activated carbon; HL= Hydraulic load. n.a.= not available

Target compounds	Treatment ^a	Scale ^b	Source	Type of DW ^c	Parameters ^d	Summary of results	References
HAAs precursors	ADS	L	Surface water	R	type of PAC= TiO ₂ NB 550 or TiO ₂ NB 700; TiO ₂ = 1.5 g/L	TiO ₂ NB 550: reduction of HAAs formation potential= 50%; TiO ₂ NB 770: reduction of HAAs formation potential= 25-30%	Gora and Andrews (2019)
HAAs precursors	ADS	L	Surface water	R	type of GAC= CJ15; HL= 4 m/h; period = 7-10 d; GAC= Φ1.5 mm × 5 mm; H = 1500 mm	HAAs precursors removal= 9 ± 20%	Chen et al. (2007)

HAA precursors	ADS	L	Surface water	R	adsorption on GAC; HL= 2 m/h; EBCT= 30 min	HAA precursors removal= 5-15% (rapid exhaustion)	Amini et al. (2018)
TCAA	ADS	L	n.a.	S	spherical cellulose adsorbent; solid-to-solution ratio of 1.0 g/L; initial TCAA= 50 mg/L; reaction time= 5 h; pH > 5	TCAA removal= 80-100%.	Lin et al. (2016)
HAA precursors	CF	L	Surface water	R	coagulant= FeCl ₃ ; FeCl ₃ = 17-25 mg/l	HAA precursors removal= 21 ± 10%	Chen et al. (2007)
HAA precursors	IE	L	Surface water	R	HL= 2 m/h; EBCT= 30 min	HAA precursors removal (DOC)= 80%	Amini et al. (2018)
HAA precursors	IE+CF	L	Surface water	R	coagulant= Al ₂ (SO ₄) ₃ ·(14-16)H ₂ O; IE resin in slurry form= 1-6 mL/L	Reduction of HAA formation potential > 60%	Levchuk et al. (2018); Singer et al. (2002)
TCAA+DCAA	O ₃ /H ₂ O ₂ /UV	L	n.a.	S	O ₃ = 0.3 mg/min; H ₂ O ₂ = 2.5 mg/L; UV at 254 nm; t= 3 min	TCAA removal > 50%. DCAA removal > 90%	Matilainen and Sillanpää (2010)
HAA precursors	O ₃ /H ₂ O ₂ /UV	L	Surface water	R	O ₃ = 1-2 mg/mgDOC; H ₂ O ₂ = 10 mg/L; UV at 254 nm	Reduction of HAA formation potential= 45%	Sillanpää et al. (2018a)
HAA precursors	O ₃ /H ₂ O ₂ /UV	L	Groundwater	R	O ₃ = 0.5 mg/L; H ₂ O ₂ = 10 mg/L; UV at 254 nm; UV fluence= 0.6 and 3 J/cm ²	Reduction of HAA formation potential= 68%	Sillanpää et al. (2018a)

2.2. Microcystin-LR

Cyanobacteria are able to produce a large variety of bioactive substances. Some of these, cyanotoxins, are toxic to humans (WHO, 2015). To date, over 80 different types of cyanotoxins are known including microcystins that are produced by several species of common planktonic cyanobacteria. The most common, as well as the most toxic, in surface waters is the microcystin-LR (MC-LR where L is leucine and R is arginine) (Antoniou et al., 2018; Takumi et al., 2017), a cyclic heptapeptide toxin (Takumi et al., 2017). The most common types of cyanobacteria that can produce microcystins are *Microcystis*, *Nodularia*, *Oscillatoria*, *Nostoc*, *Planktothrix* e *Anabaena* (Takumi et al., 2017; Westrick et al., 2010; WHO, 2017b). Acute symptoms due to ingestion of contaminated water are episodes of gastroenteritis, fever and skin, eyes, throat and respiratory tract irritation and neurotoxicity (ISS, 2011; WHO, 2015). However, the liver is the main target of the toxicity of the microcystin (ISS, 2011; WHO, 2017b; Woolbright et al., 2017). Long-term chronic effects, on the other hand, also include genotoxicity and carcinogenicity (IARC, 2010).

The current European Directive 98/83/CE (EC, 1998) does not include a limit neither on microcystins nor on MC-LR. Because of the dangers on human health due to the toxicity, evidenced since 2003 (Antoniou et al., 2018; Takumi et al., 2017), some member states provided autonomous provisional limit values over the years. For example, Italy (IMH, 2012) introduced a limit of 1 µg/L as an equivalent MC-LR referring to the sum of the concentrations of the different microcystins congeners present in the sample. In 2017 the WHO (2017b) recommended a

temporary limit on the MC-LR of 1 µg/L. This value was confirmed in the support document to the revision of the Annex I Council Directive 98/83/EC (WHO, 2017a) and fully incorporated in the revision of the DWD 2018 with a proposed value of parameter of 1 µg/L. The main approaches related to the removal of MC-LR, such as cyanotoxins in general, can be: (i) removal of cyanobacteria responsible for the production of MC-LR or (ii) removal of toxins present in the water (Vlad et al., 2014; Westrick et al., 2010). For the first and easiest approach, the WHO (2015, 2017b) suggests coagulation/flocculation, sand filtration and micro and ultrafiltration (MF and UF) as effective treatments. The study by Westrick et al. (2010) showed percentages of intact cyanobacteria cell removal, with coagulation/flocculation, sand filtration and (MF/UF) of 99.5%, 99.5% and 98%, respectively, thus demonstrating the efficiency of these treatments for eliminating directly the cyanobacteria cells.

Regarding the removal of extracellular MC-LR, more critical approach but also more effective because it acts on the free toxin present in the drinking water, WHO (2015, 2017b) suggests membrane filtration and adsorption on AC as optimal treatments. Not all types of membrane filtration are useful for MC-LR removal. As reported by Eke et al. (2018) the molecular weight of MC-LR is approximately 1kDa, while the literature molecular weight cut-off for UF and nanofiltration (NF) ranges from 10kDa-100kDa and 1kDa-10kDa, respectively. Therefore, they found that the concentration of MC-LR is not influenced by UF while NF can partially remove the toxin. The study by Cermakova et al. (2017) confirms the possibility of adequately removing organic nitrogen compounds, with a very low molecular weight (e.g. cyanobacterial

toxins and therefore also MC-LR), through adsorption on AC. These results are also confirmed by the study of Guerra et al. (2015) that reaffirm the use of AC as a very effective solution in the removal of MC-LR.

The study of Sorlini and Collivignarelli, (2011) and Sorlini et al., (2018) confirms removals between 80% and 90% of cyanotoxins, in particular MC-LR, by means of adsorption on AC. For MC-LR, the literature also noted that an adjustment of the solution pH conditions, to low pH, results in an enhanced adsorption of free toxins (Sorlini et al., 2018). Instead, Lopes et al. (2017) evaluated the removal efficiency of MC-LR from drinking water using a combined treatment that included Fenton oxidation, an AOPs, sand filtration and GAC adsorption. They concluded that the Fenton process in combination with a GAC filter is a viable and effective option for purifying water containing even high concentrations of MC-LR. Oxidation with ozone and chlorine is reported as a possible treatment for the removal of MC-LR (Westrick et al., 2010); but this solution can also be a possible cause of the release of new cyanotoxins in the treated water (WHO, 2015).

Furthermore, in recent years the photo-Fenton process, another AOPs, has been applied to remove MC-LR in DWTPs. For instance, Karci et al. (2018)

studied the influence of H₂O₂ and Fe²⁺ concentration on removal efficiencies and noted that higher yields can be obtained increasing concentrations of H₂O₂ and Fe²⁺. The main disadvantage due to the photo-fenton process is the production of chemical sludge that must be properly treated and disposed of with an increase in the costs of managing the DWTPs (Collivignarelli et al., 2019b; Sillanpää et al., 2018a).

Recently biological degradation became a promising technology (Kumar et al., 2019; Thees et al., 2019; Yang et al., 2014). The *Sphingomonas sp.* was the first strain reported to degrade MC-LR (Jin et al., 2018); moreover, some others naturally occurring bacterial species, such as *Arthrobacter sp.* and *Rhodococcus sp.*, showed a fast removal rate of MC-LR (up to 100%) (Kumar et al., 2019, Thees et al., 2019).

This treatment method is currently under study, but it potentially can be applied in the DWTPs as biological activated carbon filter or biosand filter. In fact, biological processes present some advantages over physicochemical treatment. They are: (i) more economical, (ii) more effective and (iii) they produce fewer toxic by-products (Kumar et al., 2019). In Table 2, examples of suitable treatments for MC-LR removal in DWTPs are reported.

Table 2. Examples of suitable treatments for MC-LR removal in DWTPs. ^a: ADS= adsorption, UF= ultrafiltration, NF= nanofiltration, PF= photo-fenton, BD= bacterial degradation. ^b: L= laboratory scale, F= full-plant scale. ^c: DW= drinking water; R= real drinking water, S= synthetic drinking water; ^d: PAC= powder activated carbon. n.a.= not available

Target compounds	Treatment ^a	Scale ^b	Source	Type of DW ^c	Parameters ^d	Summary of results	References
MC-LR	ADS	L	Surface water	R	mesoporous PAC; PAC= 20 mg/L; BET surface area= 257 m ² /g; initial MC-LR= 5 µg/L; pH= 3.2-8.0; t= 10 min	MC-LR removal= 80-90%	Park et al. (2017)
MC-LR	ADS	L	n.a.	S	adsorption on PAC-Fe(III); PAC= 500 mg/L; initial MC-LR= 10 mg/L; pH= 4-10	MC-LR removal= 70%	Dai et al. (2018)
MC-LR	ADS	n.a.	n.a.	n.a.	<i>Moringa oleifera</i> Lam. seeds powder (PAC); PAC= 0.25–1.0 g/L; MC-LR= 15–120 mg/L; t= 15-360 min; pH= 2-7	MC-LR removal= 98%	Yasmin et al. (2019)
MC-LR	UF or NF	L	n.a.	S	membrane in cellulose acetate; P _{UF} = 2.76 bar; P _{NF} = 4.83 bar; initial MC-LR= 10 mg/L; pH > 7	UF: MC-LR removal= 10%; NF: MC-LR removal= 40%	Eke et al. (2018)
MC-LR	O ₃	n.a.	n.a.	n.a.	initial MC-LR= 21 µg/L; O ₃ = 1.2 mg/L; t= 5 min	MC-LR removal= 73%; (O ₃ residual= 0.13 mg/L)	Hitzfeld et al. (2000)
MC-LR	O ₃	L	n.a.	S	Initial MC-LR= 12 mg/L; O ₃ = 0.4 mg/L; t= 4 min; pH > 7,	MC-LR removal= 100%	Eke et al. (2018)
MC-LR	O ₃	n.a.	n.a.	n.a.	initial MC-LR= 9 µg/L; O ₃ = 1 mg/L; t= 5 min	MC-LR removal= 50%	Hitzfeld et al. (2000)
MC-LR	PF	L	n.a.	S	initial MC-LR= 1 mg/L; Fe ²⁺ = 7.2 µM; H ₂ O ₂ = 300 µM; UV= 70 mW/cm ² ; pH=5.7	MC-LR removal= 80-100%	Karci et al. (2018)
MC-LR	BD	L	Surface water	R	<i>Arthrobacter spp.</i> ; initial MC-LR= 5 µg/L; t= 2d	MC-LR removal= 84%	Kumar et al. (2019)
MC-LR	BD	L	n.a.	S	<i>Stenotrophomonas acidaminiphila</i> strain MC-LTH2; initial MC-LR= 21.2 mg/L; T= 30°C; pH= 6-8; t= 7d	MC-LR removal= 100%	Yang et al. (2014)
MC-LR	BD	L	n.a.	S	<i>Bacillus sp.</i> ; initial MC-LR= 0.22 mg/L; bacterial= 8.3 × 10 ⁶ CFU/mL; T= 12d;	MC-LR removal= 74%	Kansole and Lin (2016)

2.3. PFAS

The acronym PFAS (Perfluoro alkylated Substances) refers to a family of persistent organic pollutants (POPs) consisting of chains of carbon atoms, linear or branched, of variable length and linked to fluorine atoms and other functional groups (Buck et al., 2011; Xiao et al., 2013). The most studied and known PFAS molecules are those composed of 8 carbon atoms, namely Perfluorooctanoic Acid (PFOA) and Perfluorooctylsulfonic Acid (PFOS) (Appleman et al., 2014; Banzhaf et al., 2017; Flores et al., 2013; Sun et al., 2016). Given the particular chemical properties, (the C-F bond is very stable), these molecules are decisively resistant to the environment.

For this reason, over the years PFAS molecules have seen an ever-increasing use in the industrial sector especially as chemicals in industrial processing or as additives in consumer products (Buck et al., 2011; Sharma et al., 2016; Sun et al., 2016; Xiao et al., 2013). However, PFAS are highly persistent but unlike many other pollutants (e.g. dioxin) are soluble in water (Appleman et al., 2014). Therefore, the major concern given by the PFAS is aroused by the long time it takes for a person to dispose of them from his body. They can cause liver damage and are considered potential toxic agents for human reproduction (IARC, 2017; WHO, 2017a). In fact, for instance in 2017 the IARC inserted the PFOA in the list of possible carcinogenic compounds - class 2B (IARC, 2017).

Currently there is no European legislation on this type of pollutants. The DWD currently in force does not provide for any limit value. The suggestion of WHO (2017a) is to introduce a limit values of 0.4 µg/L for PFOS and 4 µg/L for PFOA. The new parameter values proposed in DWD 2018 are 0.1 µg/L for the individual PFAS and 0.5 µg/L for PFAS-Total (this is the sum of per- and polyfluoro alkyl substances). This is because the European Commission defines as a priority the application of the precautionary principle already adopted for example to set parameter values of pesticides (EC, 2018).

Regarding the possible treatments for removing the PFAS from contaminated water, in recent years, research has focused both on the application of conventional treatments and on the study of advanced treatments. It is necessary to highlight that conventional treatments, except ion exchange resins, give contrasting results depending on the type of compound considered. The coagulation/flocculation allows a very low removal of PFOA and PFOS (10-30%) when the contaminant concentrations is in the order of µg/L (Appleman et al., 2014; Xiao et al., 2013). Instead, the impact of filtration on GAC has been studied by Eschauzier et al. (2012). While for the PFOA and PFOS a reduction of about 50% and over 90% respectively can be noted, for the PFXa, PFXs and PFNA the removal is negligible. Indeed, the PFBA and the PFBS are easily

released by the filter, making this type of treatment inapplicable for their removal.

The most recent study of McCleaf et al. (2017) has confirmed the most efficient removal, by means of adsorption on AC, of compounds such as PFOS and PFOA in spite of compounds such as PFBA which instead can give rise to the desorption phenomenon. Also, the study of Sun et al. (2016) confirms the greater simplicity in removing long chain PFAS (e.g. PFOS, PFOA) with powder activated carbon (PAC) in spite of those with short chain (e.g. PFBA). Appleman et al. (2014) demonstrated that conventional chemical oxidation treatments and AOPs do not have significant oxidative power against PFAS. PFOS and PFOA are slightly removed through processes such as UV/H₂O₂ and even treatments such as ozonation are counterproductive.

It is worth to note that UV-alone process gives encouraging results (about 35%) on PFOS removal (Appleman et al., 2014). With regard to membrane processes, it was found that MF and UF are not effective in removing PFAS (Appleman et al., 2014; Flores et al., 2013). Instead, NF and reverse osmosis systems are able to ensure a high effectiveness of removal of both short chain PFAS (e.g. PFBA) and long chain PFAS (e.g. PFOA and PFOS) (Hopkins et al., 2018). In fact, Flores et al. (2013) obtained a reduction of over 99% of long chain PFAS by reverse osmosis.

In recent years, several studies showed a significant effectiveness of ion-exchange resins in the removal of PFAS (both long and short chain) (Appleman et al., 2014; Hopkins et al., 2018; McCleaf et al., 2017; Woodard et al., 2017) overcoming some limits related to the use of AC (Eschauzier et al., 2012). Pilot-scale experiments, made by Conte et al. (2015), demonstrated that all tested materials (resins A600E, PAD500, PAD428 and MN102) removed the long chain PFAS (PFOA and PFOS) with almost 100% efficiency for long time, although they evidenced significant differences in the short chain PFAS (e.g. PFBA) removal.

Despite the excellent yields, to date the disadvantages of ion-exchange resins are essentially two: (i) exhausted resin must be managed and (ii) performances strongly depend on resin properties (Hopkins et al., 2018). In Table 3, examples of suitable treatments for PFAS removal in DWTPs are reported.

2.4. Bisphenol A and Nonylphenol

Bisphenol A (BPA) and Nonylphenol (NP) are two chemicals used in the production of polycarbonate plastics, epoxy resins and other polymeric materials (Chen et al., 2016; Lee and Choi, 2006). Polycarbonate plastic materials are used, for example, for the production of food and beverage containers, water containers and information technology equipment (Chen et al., 2016; Muhamad et al., 2016; Seachrist et al., 2016).

Table 3. Examples of suitable treatments for PFAS removal in DWTPs. ^a:ADS=adsorption, CF=coagulation/flocculation, SF= sand filtration, IE=ion exchange, NF=nanofiltration, RO=reverse osmosis. ^b:L= laboratory scale, F=full-plant scale. ^c:DW= drinking water; R=real drinking water, S= synthetic drinking water. ^d:GAC= granular activated carbon; HL=Hydraulic load. n.a.=not available

Target compounds	Treatments ^a	Type of plant ^b	Source	Type of DW ^c	Parameters ^d	Summary of results	References
PFOS	ADS+Cl ₂	F	Groundwater	R	adsorption on GAC; PFOS= 29-59 ng/L	PFOS removal= 7%	Rahman et al. (2014)
PFOS	CF+SF+O ₃ +A DS+SF	F	Surface water	R	adsorption on GAC; PFOS= 8.2 ng/L	PFOS removal= 97%	Rahman et al. (2014)
PFOA	SF+ADS+Cl ₂	F	Surface water	R	adsorption on GAC; PFOA= 67-92 ng/L	PFOA removal= 90-92%	Rahman et al. (2014)
PFBS	CF+SF+O ₃ +A DS+SF	F	Surface water	R	adsorption on GAC; PFBS= 35 ng/L	PFBS removal= 43%	Rahman et al. (2014)
PFOS, PFOA, PFBS, PFBA	IE	L	Groundwater	R	IE resins Purolite [®] A600E, A520E, A532E; resin= 1 g/L total capacity= 0.85-1.6 eq/L; PFOS= 27 ng/L; PFOA= 430 ng/L; PFBS= 171 ng/L; PFBA= 212 ng/L	PFBS and PFBA removal= 100% (with 20000 bed volumes); PFOS and PFOA removal= 100% (with > 60000 bed volumes)	Zaggia et al. (2016)
PFOS, PFOA, PFBS, PFBA	ADS	F	Groundwater	R	adsorption on GAC; minimum iodine number= 900-1100 mg/g; HL= 10-11 m/h; EBCT= 10-11 min; PFOS= 27 ng/L; PFOA= 430 ng/L; PFBS= 171 ng/L; PFBA= 212 ng/L	PFOS adsorption capacity= 2.4-4.1 µg/L; PFOA adsorption capacity= 17.3-39.6 µg/L; PFBS adsorption capacity= 6.8-8.1 µg/L; PFBA adsorption capacity= 3.8-4.3 µg/L	Zaggia et al. (2016)
PFOS	NF and RO	L	n.a.	S	NF in crossflow at 1.37 L/min; PFOS= 10 mg/L; P= 1379 kPa; pH= 4	NF: PFOS removal= 90-99%; RO: PFOS removal= 99%	Tang et al. (2007)
PFOS PFOA	ADS and IE	L	Groundwater	R	adsorption on GAC; EBCT _{GAC} = 20 min; EBCT _{IE} = 7.5 min; PFOS= 26 µg/L; PFOA= 12 µg/L;	GAC: PFOS and PFOA removal= 99%; IE: PFAS and PFOA removal= 99%; (IE removed over four times as much total PFAS/g as GAC before significant breakthrough was observed)	Woodard et al. (2017)

Epoxy-phenolic resins based on BPA are also used as protective coatings for tanks and drinking water pipes (Chen et al., 2016). Furthermore, NP is a biodegradation product of nonylphenol ethoxylate (NPE) which is the most common non-ionic surfactant used daily (Khatibikamal et al., 2019) BPA and NP, as well as all EDCs, have for years been the subject of a strong scientific controversy regarding their health risk. For example, the European CHemicals Agency (ECHA) in 2017 issued a unanimous verdict on BPA defining it as an EDC with probable serious effects on human health (ECHA, 2017). Also, in the case of the

BPA, Arnold et al. (2013) conducted a comprehensive research study, describing BPA concentrations in drinking water-using the data contained in 65 papers (31 of North America, 17 of Europe and 17 of Asia) to assess the relevance of drinking water as a source of human exposure and risk.

On the contrary, they have been able to assert that the data indicate that ingestion of drinking water represents the source of only 2.8% of the total intake of BPA by monitored human individuals. Although the WHO (2017a), according to this vision, has declared that currently there are no proven health risks

deriving from the presence of BPA and NP in drinking water, the DWD 2018 provides for the introduction for these parameters of the specific limit values of 0.01 µg/L for BPA and 0.3 µg/l for NP (EC, 2018). In Table 4, examples of suitable treatments for BPA and NP removal in DWTPs are reported. Regarding the removal of BPA, the conventional treatment technologies applied in the drinking water treatment plants have a satisfactory yield of 76-99% (Arnold et al., 2013). For example, Stackelberg et al. (2007)

obtained a 76% removal of BPA in a typical potabilization plant composed of clarification with ferric chloride, primary disinfection with sodium hypochlorite, sand filtration, GAC and secondary disinfection. In this study, it is also noted that the adsorption on GAC is responsible for more than 50% of the BPA removed. Membranes are another technology that can significantly remove BPA; however, the removal yields are sensibly dependent on the type of the membrane material.

Table 4. Examples of suitable treatments for BPA and NP removal in DWTPs. ^a: ADS= adsorption, NF= nanofiltration, RO= reverse osmosis, PF= photo-fenton, CF= coagulation/flocculation, SF= sand filtration. ^b: L= laboratory scale, F= full-plant scale. ^c: DW= drinking water; R= real drinking water, S= synthetic drinking water; ^d: GAC= granular activated carbon; PAC=powdered activated carbon. n.a.= not available

Target compounds	Treatment ^a	Type of plant ^b	Source	Type of DW ^c	Parameters ^d	Summary of results	References
BPA	ADS	L	n.a.	S	adsorption on GAC; GAC from Macauba palm; surface area= 907.0 m ² /g; GAC= 10 g/L; initial BPA= 100 mg/L; T= 25-80 °C; pH= 3-9;	BPA removal= 50-100%	Moura et al. (2018)
BPA	ADS	L	n.a.	S	adsorption on biobased surface functionalized cellulose fibers; fiber loading = 1 g/50 mL; initial BPA= 15 mg/L; t= 60 min; pH= 5	BPA removal= 70-80%	Tursi et al. (2018)
BPA	ADS	n.a.	Surface water	n.a.	adsorption on PAC; PAC= 5-15 mg/L; PAC breakthrough= 19597 bed volume; initial BPA= 45 ng/L; EBCT= 1.5-3 min	BPA removal= 68%	Hernández-Leal et al. (2011)
BPA	NF	n.a.	n.a.	S	membrane NF90 and NF270 (Dow Filmtec); initial BPA= 20 mg/L	NF270: BPA removal= 55%; NF90: BPA removal= 94%	Muhamad et al. (2016)
BPA	RO	n.a.	n.a.	S	membrane TW30-1812-100 (Dow Filmtec); initial BPA= 50 mg/L; P= 408.1 kPa; flow rate= 1.172 L/min; pH= 8	BPA removal= 87.3 %	Muhamad et al. (2016)
BPA	PF	n.a.	n.a.	S	initial BPA= 43.8 µmol/L; H ₂ O ₂ = 4x10 ⁻⁴ mol; Fe ²⁺ = 4x10 ⁻⁵ mol; UV= 320-410 nm on 0.5 mW/cm ² ; t= 9 min; pH= 4.0;	BPA removal= 100%	Liang et al. (2015)
NP	CF+SF	F	Surface water	R	initial NP= 0.1-7.3 µg/L; T=12-28 °C	NP removal= 62-95%	Shao et al. (2005)
NP	ADS	LS	n.a.	S	adsorption on GAC; GAC= coal-based Calgon Filtrasorb® 400 and coconut shell-based PICACTIF TE; iodine number= 1000 mg/g and 1237 mg/g; SA BET= 1030 m ² /g and 1156 m ² /g; initial GAC= 13-16 mg/L; initial NP= 500 ng/L	NP removal= 90% (with both type of GAC)	Yu et al. (2008)
NP	ADS	n.a.	Surface water	n.a.	adsorption on coal-based PAC; PAC breakthrough= 44,141 bed volume; initial NP= 15 µg/L; EBCT= 15 min	NP removal= 50-90%	Hernández-Leal et al. (2011); Yang et al. (2017)

Yüksel et al. (2013) tested UF and reverse osmosis with the aim of identifying the optimal material for BPA abatement. The result is a better rejection of BPA with polyamide membranes compared to cellulose acetate membranes. Furthermore, it has been shown that up to 98% removal can be achieved with reverse osmosis (Yüksel et al., 2013). These excellent results have been confirmed by the study of Albergamo et al. (2019) that have obtained more than 75% rejection of BPA. Rodriguez-Narvaez et al. (2017) have instead tested the removal of BPA thanks to a Photo-Fenton process, an AOP. The result is a removal of about 98% in just 20 minutes of treatment.

NP, as well as BPA, is removed quite easily with conventional treatments such as AC adsorption. Yang et al. (2017) showed removal from 50 to 90% using GAC. Studies are also looking for alternative adsorbents optimized for the removal of EDCs, such as NP. For example, Khatibikamal et al. (2019) found 67% removal of NP from drinking water using poly (amidoamine) coated magnetic iron oxide nanoparticles as adsorbent substance. Conventional chemical oxidation processes (e.g. ozonation), remove only around 30% of NP (Barrera-Díaz et al., 2018). On the contrary, electrochemical treatments, which can also be used for the removal of other pollutants from wastewater (e.g. heavy metals) (Collivignarelli et al., 2019a), allow a reduction of NP concentration of 70% from drinking water (Barrera-Díaz et al., 2018).

3. Summary and future outlooks of the research

- **HAA:** the removal of HAAs precursors solves only partially the problem due to the HAAs formation. In fact, yields of HAAs precursors removal are in the range of 50-80%. At the same time, focus the attention only to directly HAAs removal can be low effective; in fact, if high concentration of NOM is present in the water, HAAs will be present in too high concentration to be completely removed. Therefore, both types of interventions should be studied in deep with further research, particularly the ion exchange and activated carbon techniques, which show excellent results in the field of precursor and HAAs removal respectively. Regarding operational changes, chlorine could be replaced by other oxidants, but it must be considered that other DBPs can be formed (for example by dosing ozone or chlorine dioxide) and that other oxidants (e.g. UV rays) may not have persistence in the drinking water distribution network.

- **MC-LR:** currently, the research is focusing in particular on the removal of extracellular MC-LR, more critical approach but also more effective because it acts on the free toxin present in the drinking water. Adsorption and chemical oxidation provide good removal yields while not all types of membrane filtration are useful for MC-LR removal: the concentration of MC-LR is not influenced by UF while NF can only partially remove the toxin. Moreover, in future an interesting point could be

analyse better the combined effect of MF or UF with PAC. In fact, PAC would be able to remove effectively MC-LR toxin and, at the same time, MF or UF would allow to remove cyanobacteria (for preventive purpose) and separate the PAC from drinking water. Recently, also the biological degradation became a promising technology. However, further studies are needed on the application of biological treatments for the removal of free MC-LR toxin, in particular about the factors that can influence the degradation process in such a way as to maximize the process performance.

- **PFAS:** in general, PFAS removal from drinking water still remains a significant problem, especially for short chain PFAS (e.g. PFBA). Research is focusing in particular on the removal by ion exchange resins and membranes (NF and reverse osmosis), that ensure higher removal yields both for long and short chain PFAS, while other processes such as chemical treatments don't have enough oxidative power against PFAS. AC ensure good removal yields on long chain PFAS, but the main gap is related with the ineffective removal of short chain PFAS. The perspective of future research in this field could be also the integration of AC with NF or reverse osmosis in order to ensure the compliance with the new limits introduced by the proposal of DWD 2018. The research on AC application could be implemented studying new adsorbent materials and optimizing the configuration of the reactors.

BPA and NP: BPA and NP are removed easily with conventional treatments, such as AC adsorption or coagulation/flocculation. Currently, studies are focusing on: (i) research alternative adsorbents (e.g. derived from palm or coconut shell) optimized for the removal of BPA and NP in order to increase removal yields, (ii) evaluate the efficiency of AOPs (in particular Photo-Fenton process) and (iii) search and test optimal materials for membranes (e.g. polyamide), considering that this aspect significantly influences the efficiency of removal.

4. Conclusions

This paper presented the results of the application of conventional and advanced treatments for the removal of emerging contaminants (ECs). In particular, the following contaminants were investigated: HAAs, MC-LR, PFAS, BPA and NP. The comparison of more than 100 documents, articles and reviews showed that for some ECs it is possible to operate in different ways. For example, for HAAs the target can be directly the contaminant or the precursor responsible for its generation. At the same way, MC-LR can be directly removed as toxins (critical aspect), after they have been dissolved in water, or it is possible to act on the removal of suspended cyanobacteria that are responsible for their release in water.

Overall, it can be asserted that while for some ECs (e.g. HAAs, MC-LR, BPA, NP) conventional

remediation treatments can be considered highly effective, for others (e.g. PFAS) these may not be enough. In these cases, it is necessary to adopt advanced treatments in order to allow compliance with the limits set by legislation and therefore the protection of public health.

In some cases, also advanced processes have a limited effectiveness and new researches are necessary for identifying effective solutions. However, while for HAAs the number of applicable treatments is higher, as regards the removal of MC-LR, PFAS, BPA and NP further studies are needed to optimize the performance of those already identified and to search for further effective treatments.

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SUSTAINABLE MANAGEMENT OF SEDIMENTARY RESOURCES: A CASE STUDY OF THE EGADI PROJECT

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Abstract

Multiple activities carried out on coastal areas expose marine sediments to contamination and their management has a great socio-economic importance with a high impact on economic development of coastal areas. However, there is an increasing shift towards the use of more sustainable approaches for managing ‘contaminated’ sediments. Using a case study of the Favignana Harbour in Italy, this paper evaluates three approaches for the management of these sediments. The results of simulations carried out by SiteWise™ software show that the use of contaminated sediment as filling material for Confined Disposal Facilities has lower environmental footprint than treatment and reuse of sedimentary resources on shore. The implications for these results for the development of effective policies and practices by all key stakeholders are discussed.

Key words: dredging, footprint, green and sustainable remediation, LCA, LCC, natural resource management, sediment, SiteWise™

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1. Introduction

A wide range of mechanical-biological processes Bortone and Palumbo (2007) define **sediments** as: *suspended or deposited solids, acting as a main component of a matrix, which has been, or is susceptible to being transported by water and they are an important natural resource for the economic development of many Countries.* Fig. 1 shows that effective sediment management plays a crucial role in the environmental, social and economic sectors, including habitat management, recreation and agriculture (Do-Hyung et al., 2013; Manap and Voulvoulis, 2015; Wen-Yen et al., 2016). From the social point of view, sediments form beaches which serve to reduce flooding, as well as to provide recreational spaces. From the economic point of view,

especially in small islands, recreational spaces (e.g. for boating) are reduced as sedimentation can reduce the capability of coastal infrastructure to tie up boats (Ausili et al., 2012; Cappucci et al., 2011; Fernández-Fernández et al., 2019). From an environmental perspective, sediments play a vital role in maintaining the health and viability of aquatic ecosystems (Puentes-Rodríguez et al., 2015).

Pollutants tend to be absorbed by particulate matter in aquatic environments, and to settle on the bottom, forming contaminated sediments (EPA, 2005). The accumulation of these contaminants can be due to natural or anthropogenic sources (Matache, 2018). Natural factors include all phenomena that exclude human impacts (e.g. volcanic eruptions, forest fires, and the natural processes performed by plants and animals).

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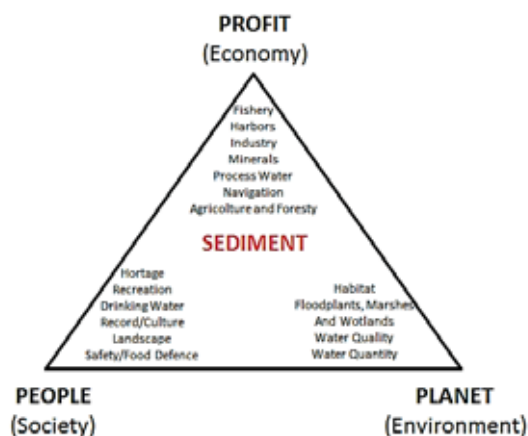


Fig. 1. The three fields of sustainable development and their interaction in relationship with sediment (Adapted upon Munasinghi (1998))

However, natural factors can lead to pollutant concentrations exceeding the threshold of contamination defined by national legislation (Aptiz et al., 2017; Cappucci et al., 2011). Anthropogenic sources, however, are represented by all the activities producing toxic or harmful substances and affecting the ecosystems (IMO, 2000). Anthropogenic contaminants may enter the aquatic environment through point sources, such as industrial or civil discharges, or from diffuse sources, such as runoff, erosion of farmland treated with pesticides and atmospheric deposition (Brettschneider et al., 2019; Moreira et al., 2019; Soliman et al., 2019). Apart from application on shore, few approaches have been outlined for remediation of contaminated sediments offshore (Aptiz, 2008; Sparrevik et al., 2012). For this reasons, a conceptual model of contamination is the key part of seabed remediation process. Classification of sediments and management options are related to their level of toxicity and contamination. Usually three different colours, namely “green”, “yellow” and “red”, can be adopted to show whether dredged sediments can be effectively used for beach formation (Conti et al., 2009), disposed within Confined Disposal Facilities (CDFs) or landfilled (Cappucci et al., 2011).

While the legislation tends to mitigate the environmental impact derived from the movement of sediments in the coastal marine environment, there is much debate about prevention of the disposal of dredged material into marinas (Aptiz et al., 2017). Hot spots of contamination are often the navigation channels, or internal parts of the harbors (Ausili et al., 2012).

Thus national and international legislations are rapidly evolving to take account of the rise of anthropogenic ‘contaminated’ materials in the sea, balanced against the concept of sediment as a “resource” and not as a “waste” (Junakova and Junak, 2017). The application of sustainability to reclamation known as sustainable remediation, together with the “green remediation”, has been the object of recent studies (Anvar et al., 2018; Aptiz et al., 2017; Zijp et al., 2016). The SuRF groups, the Network for

Industrially Contaminated Land in Europe (NICOLE), national bodies and agencies (USEPA and OSWER) were the main promoters of the development of these new concepts. The sustainable remediation is also the topic of ISO Standard 18504:2017 (ISO, 2017). However, environmental remediation within the contaminated sites generally focuses on restoring natural resources to an acceptable risk level for the society (Simion et al., 2011; Dauvin et al., 2018; Fernández-Fernández et al., 2019). In addition, remediation of contaminated hot spots may have negative impacts from local to global scales (Suèr et al., 2004). To support reclamation process, qualitative, semi-quantitative and quantitative tools are assessed and under development (Lemming et al., 2010). Thanks to these tools, it is possible to analyze a reclamation procedure by breaking it down into phases, analyzing environmental, economic and social impacts. After Volkwein et al. (1999), several authors compared several techniques and remediation options for specific contaminated sites including Life Cycle Assessment (LCA) (U.S.S.R.F., 2009; Wen-Yen et al., 2016). Since the green remediation program was launched by Environmental Protection Agency (EPA), various authors have started to evaluate and implement tools to apply sustainability criteria on remediation of contaminated sites. Remediation action can be performed in many different ways, depending on the level of contamination (EPA, 2005; U.S.S.R.F., 2009). The SuRFItaly Group (Sustainable Remediation Forum Italy; <http://www.surfitaly.it/organization.html>) has defined sustainable remediation “The process of management and remediation of a contaminated site, aimed at identifying the best solution that maximizes the benefits of its execution from an environmental, economic and social point of view, through a process decision-making shared with stakeholders”. This definition introduced for the first time the concept of sustainability in the field of remediation that meets the needs of the present without compromising the ability of future generations to meet their needs (Brundtland, 1987).

Multi-Criteria Decision Analysis (MCDA) is an environmental management tool that can be adopted to choice suitable remediation technologies and for prioritization of intervention planning (Guerra et al., 2010; Linkov et al., 2006). Environmental assessment is however difficult to implement as the choice of technologies are often driven by profit and not by the desire to implement a sustainable process and or public services (Gebert et al., 2019). Addressing the social impacts is also often challenging because as many companies and authorities are often reluctant to increase the initial capital spend for intervention even if it could potentially become profitable in the long term. The first quantitative investigation of environmental assessment on a soil washing process for the remediation of a Pb-contaminated shooting range site was conducted by Kim et al. (2013), using a green and sustainable remediation tool (SiteWise™). Before this present

study, the use of SiteWise™ has never been applied to dredging and remediation of contaminated sediment. In the present study, we used Favignana Harbor as a case study to assess the environmental impact of different options for sediment management.

2. Objectives

The aim of the present study is to test and provide a sustainable footprint of different options for sediment management. We provided support to local authorities aiming at more sustainable remediation by using SiteWise™ (Ferdos and Rosen, 2013; <http://www.sustainableremediation.org>). Our goal was to implement a management model of harbor sediments, which, after characterization (Ausili et al., 2012), may be reused avoiding landfilling (Cappucci et al., 2011).

In the present study, sediment management options from a small port layout located in Southern Italy have been used in order to: compare the footprint of different scenarios proposed by local authorities; and determine which has the lowest impact. The main objective was to evaluate which of the possible management options has the lowest impact. Based on the complexity of sediments management and the need to guarantee periodical dredging, as well as to sustain tourism in the area (by harbour, sportive activities and versus municipalities and tourist operators), a simple and straightforward analysis was implemented. A preliminary sediment characterization carried out before the new legislation criteria, suggested a low level of contamination that could compromise the use of sediment for beach replenishment (Ferrantini, 2012).

To avoid disposal into landfill, technical and economic analyses were carried out under the hypothesis that an average volume of 22,000 m³ (estimated according to preliminary characterization carried out by local authorities; Ferrantini, 2012) must be periodically dredged and excavated to:

- create beach volleyball fields (to improve touristic services);
- restore the coast (by replenishment of the emerged beach and back shore);
- enlarge the port layout (with a CDF made with the dredged material).

In this context, the assessment of different scenarios was proposed by using the SiteWise™ software, providing individual alternatives, with relevant information related to the different options (i.e. GHG emissions; total energy used; water consumption; use of electricity; NO_x emissions onsite; SO_x emissions onsite; PM₁₀ emissions onsite; total NO_x emissions; total SO_x emissions; total PM₁₀ emissions).

3. Study area-Favignana Harbor

The Favignana Harbor (Fig. 2) is in the sheltered inlet of Cala Principal (north central area of Favignana Island), and is located in a Marine

Protected Area. The harbour is equipped with a pier about 110 m long, which extends North-west. The smaller Molo S. Bernardo stretches for about 85 meters in a southerly direction. About 100 berths are available, 30 of which are dedicated to boats of travellers/navigation. On the seabed the *Posidonia oceanica* meadow is located (Marbà et al., 2014).

Frequent siltation of the structure is due to the anticlockwise circulation inside the harbor generated during mistral winds (Cappucci et al., 2017) and dredging activities must be carried out to guarantee navigation and safety (maintainance of navigation depth). Navigation is limited, especially close to the docks, due to sand transport under the effect of anticlockwise circulation that reduces the depth of the seabed. It needs periodic dredging of about 22.000 m³.

4. Methodology and assumptions

Sediment characterization of the site was undertaken (Ferrantini, 2012) through a deterministic strategy. The characterization of superficial sediments allowed determination of the physical properties of the particles. The grain size revealed a percentage of silt and clay of about 1% and a D₅₀ of approximately 0.215 mm. The analytical tests detected a moderate contamination (Ferrantini, 2012), exceeding the thresholds set by the Legislative Decree no. 152/2006, for Cadmium, Arsenic, Lead, Tin, PAH and TBT (Tributyl tin compounds used in anti-fouling paints). The outcomes from this preliminary study were used to identify the positioning of various sampling stations in areas where accumulation of pollutants takes place due to deposition of coastal sediments in front of Piazzale Marina.

Due to the contamination level, local authorities required technical support to assess the best option to manage the dredged material. Sediment must be removed to guarantee the navigation and we assumed that they should be isolated from direct contact with marine organisms and coastal water in case they are not treated to reduce concentration of contaminants. The main hypothesis is that after the characterization, the entire volume of sediment will be managed according to the following three scenarios (Table 1):

1. Sport, to build beach volleyball fields;
2. Coast, to replenish the coasts;
3. Harbor, to fill CDF.

The study of the different sustainability assessment was carried out by using SiteWise™ (Bhargava and Sirabian (Battelle), 2013).

A detailed technical and economic analysis of treatment and remediation technologies was conducted (i.e. (GHGs), use of energy, electricity from renewable and non-renewable sources, criteria on air pollutants (NO_x, SO_x, PM), use of water, consumption of resources (soil consumed), and safety of worker (risk of accidents, lost hours)), on the basis of literature review, budget estimation (market research) and results of tests carried out on other contaminated sites (Ferrantini, 2012).

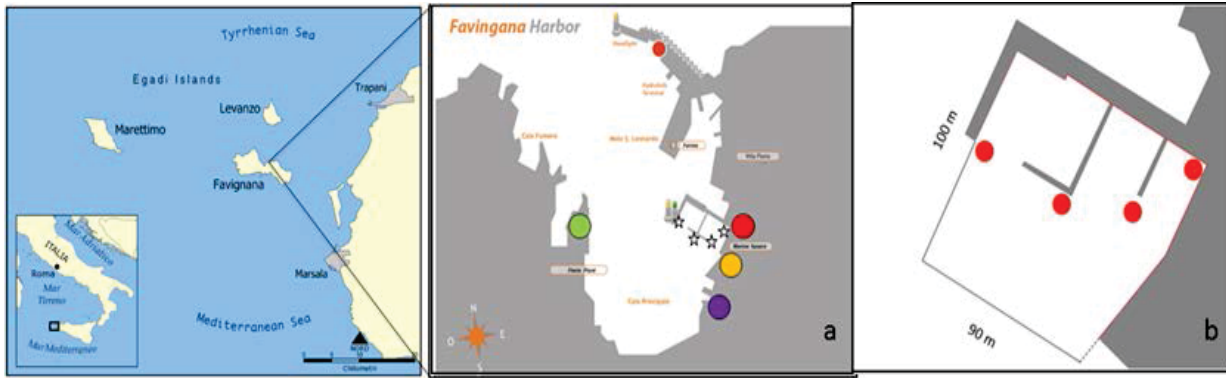


Fig. 2. Egadi archipelago and Favignana Harbor. (a) Florio’s factory (green); Gas station (red); Wastewater discharge (purple); Boat repair (orange). (b) Red dots indicate sampling station for characterization of seabed

Table 1. Scenarios for sediment management and their phases

Scenario	Volume to be dredged	Volume to be treated	Phases
N	m ³	m ³	
1 sport	22,000	22,000	Characterization → Dredging → Transport → Storage → Treatment → Installation of the fields
2 beach	22,000	22,000	Characterization → Dredging → Transport → Storage → Treatment → Requalification of coastal area
3 harbor	22,000	0	Characterization → Dredging → Harbor infrastructure

The most reliable sources of data and information have been catalogued, classified and, for each technology, a range of costs was determined.

4.1. Technical analysis of remediation technologies

Due to the low level of contamination of the Marine Protected Area and the limited extension of the Harbor layout (i.e., volume to be excavated), dredged sediment could be subject to:

- treatment before their reuse on coastal areas (beach nourishment);
- treatment and reuse on-shore (in order to ignore the bioavailability and effects of contamination on marine organisms as material will not be in contact with coastal marine water bodies);
- direct disposal within confined disposal facilities (isolated from marine organisms and water). The total footprint is calculated by integration of all the activities. The data required to model each stage of remediation scenarios in Table 2 were different, but the following inputs are mandatory to run SiteWise™ simulations:
 - material required for each stage of remediation;
 - transportation of both personnel and material (machinery, etc.);
 - all activities to be performed off shore and on shore (including mobilization and de-mobilization of devices);
 - management of sediment produced by dredging and disposal on-off-shore.

4.2. SiteWise™ simulation for different management option

The evaluation of each intervention alternative is performed by breaking it down into individual blocks that can represent the individual phases of the alternative (or their combination/aggregation). The dredging process, for example, consists of separated blocks/phases: study and design, dredging, equipment transport, personnel transport. The environmental footprint is calculated for each block, and these footprints are then combined to provide the output related to the whole alternative. In this way it is possible to determine which alternative produces the highest environmental footprint, or how to reduce it by using energy from renewable sources. In order to insert the inputs in the easiest way and reconstruct the modules and the phases that make up the alternatives in the program, the following subdivision was used (Fig. 3):

- component 1 = characterization and dredging;
- component 2 = transport (to the treatment facility or to the CFD);
- component 3 = treatment;
- component 4 = final destination of sediments.

The electrical production of California State was selected as it is the most similar to percentage of renewable electricity (26 vs 24 %), CO₂ emissions per MWh (800 vs 680 pounds/MWh) and electricity production efficiency (0.426 vs 0.445 gross electricity yield per unit of fuel energy content) compare to the study area. In Scenarios 1 and 2, pretreatment (storing and drying) of 22x10³ m³ (59x10⁶ kg) of sediments with the use of earth-moving machines was

considered. Then, a soil washing treatment plant with a potentiality of $5 \times 10^4 \text{ kg/h}$ was considered. Unit conversion and assumptions used for environmental footprint analysis are respectively reported in Table 3 and Table 4.

4.3. Economic specification and assumption of management scenarios

The costs of the three different scenarios were estimated based on the approach of Bortone and Palumbo (2007), executive projects and through quotations to companies.

The cost analyses took account of identical activities to be carried out for each different scenario, including: characterization, dredging, transport, storage and treatment. A second group of costs were then considered in relation to specific activities to be carried out for the following three different scenarios:

- installation of beach volleyball field (for sediment reuse within a beach volleyball field);
- requalification of coastal area (for sediment management to be reused along the coast);
- construction of CDFs (to enlarge the port layout).

Cost for taxes and safety were considered and included within each different scenario.

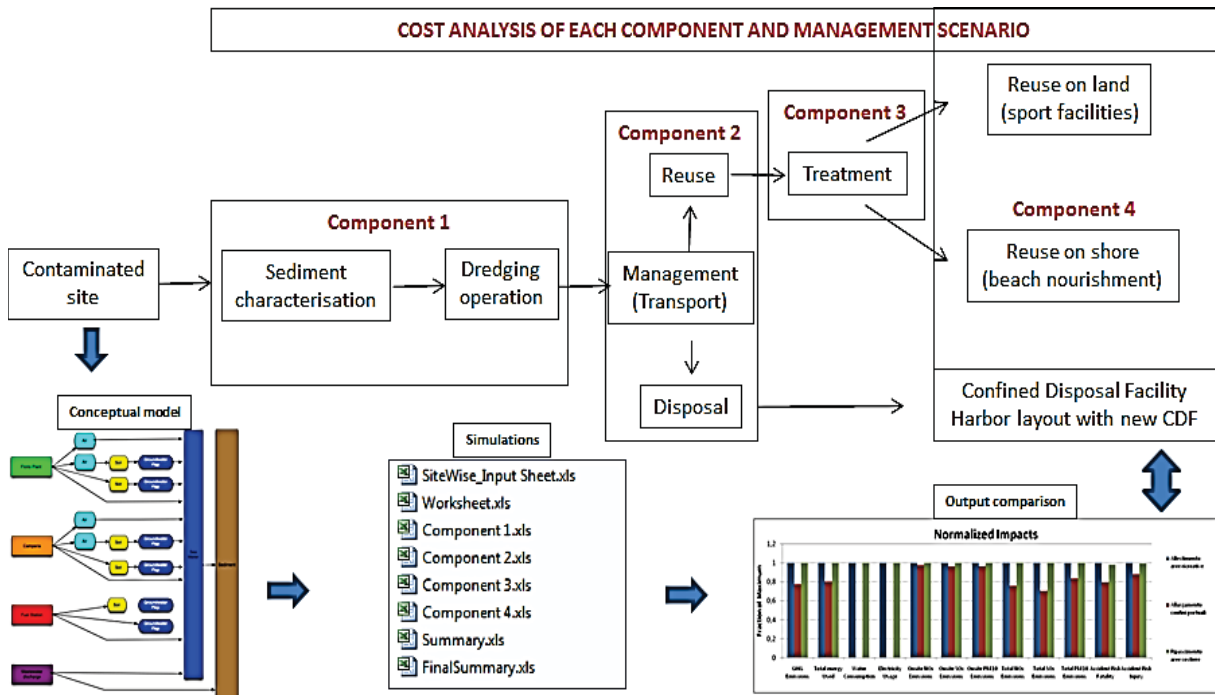


Fig. 3. Flow diagram of the simulated scenarios (conceptual model by Ferrantini, 2012)

Table 3. Conversion from Site Wise™ units to international system

Value	Conversion units (from/to)
3,412	BTU/kWh
947.867	BTU/Mj
0.001	Mj/BTU
1.055	kJ/BTU
0.746	kW/hp
33,013	ft lbs/min hp
2.204	lb/kg
0.454	kg/lb
2,204.6	lb/metric ton
1,000	kg/metric ton
3.785	L/gal
8.34	lbs H ₂ SO ₄ /gal

Table 4. Technical specifications and assumptions adopted for each scenario

COMPONENT 1 - Characterization and dredging				
Transport for "characterization"	Number/quantity	Distance [km]	Round trip	Vehicle
Personnel for study area	2	22	6	diesel car
Personnel for study area	1	500	2	airplane
Personnel for sampling	2	22	2	auto diesel

Sediment samples	4	500	1	airplane/ferry
Transport for "dredging"	Number/quantity	Distance [km]	Round trip	Vehicle
Personnel for dredging operation	4	3	12	diesel car
Dredge	1	100	2	ferry
Silt curtains	1480 kg	100	1	ferry
COMPONENT 2 - Transport of sediment				
Transport of dredged sediment	Number/quantity	Distance [km]	Round trip	Vehicle
Port - treatment volume	22,000 m ³	3.6	1,100	truck 20 m ³
treatment area – port	0 m ³	3.6	1,100	truck 20 m ³
treatment area - destination area	21,350 m ³	4	1,070	truck 20 m ³
Waste from treatment area – port	650 m ³	3.6+22	35	truck 20 m ³ + ferry
Truck drivers	2	3	368	diesel car
COMPONENT 3 - Treatment - soil washing				
Transport of personnel	Number/quantity	Distance [km]	Round trip	Vehicle
Personnel for treatment facilities	2	3	590	diesel car
Personnel for moving soil	2	3	590	diesel car
Personnel for installation	4	3	40	diesel car
Personnel for CLS transport by trucks	2	3	12	diesel car
Facilities	Number/quantity	Distance [km]	Round Trip	Vehicle
Facilities for moving soil	1	3	2	Truck
Pumps for soil washing	2	100	2	ferry + trucks
Hydrocyclons	2	100	2	ferry + trucks
Conveyor belts	1	100	2	ferry + trucks
Water treatment	1	100	2	ferry + trucks
Pre- treatment (sifts)	1	100	2	ferry + trucks
Dehydration of treated sediments	1	100	2	ferry + trucks
Pumps for hydrocyclons	2	100	2	ferry + trucks
Materials	Number/quantity	Distance [km]	Round Trip	Vehicle
Concrete - treatment area	300 m ³	22+3	30	truck 20 m ³
Concrete - storage area	960 m ³	22+3	96	truck 20 m ³
Concrete - treatment area	0	22+3	30	truck 20 m ³
Concrete - storage area	0	22+3	96	truck 20 m ³
PVC for prefabricated - treatment area	4,000 kg	100	1	ferry + trucks
PVC for roof - storage area	7,000 kg	100	1	ferry + trucks
Steel for covering PVC support	2,500 kg	100	1	ferry + trucks
Materials for treatment	Number/quantity	Distance [km]	Round Trip	Vehicle
Water	0.1 m ³ /t soil	\	\	Pumps
ETAC (ethyl acetate)	0.18 kg/t soil	100	1	Ferry + trucks
COMPONENT 4 - Recreational area facilities (4400 m²)				
Playground' s construction	Number/quantity	Distance [km]	Round trip	Vehicle
Personnel staff area	4	3	40	diesel car
Sediment	2,200 m ³	\	\	\
TNT fabric	440 kg	100	1	ferry
PVC for enclosed spaces	4,000 kg	100	1	ferry
Protection networks	81 kg	100	1	ferry
Lighting system (steel)	174 kg	100	1	ferry
COMPONENT 4 - Beach nourishment				
Sediment movement	Number/quantity	Distance [km]	Round Trip	Vehicle
Vehicle to be used for land	19,225 m ³	3	1	truck
Personnel for moving sediment	2	3	53	diesel car
COMPONENT 4 - Harbor				
Dredging and CDF	Number/quantity	Distance [km]	Round Trip	Vehicle
Dredger	22,000	100	2	crawler crane (25t)
Personnel staff area	4	3	64	diesel car
General concrete	14,790,000 kg	22	1	ferry+ trucks
Steel	102,000 kg	22	1	ferry+ trucks
Scow tenders	2	0.5	32	zodiac

5. Results

5.1. Environmental footprint of the three scenarios

Results for each alternative are provided in Table 6 where data refer to m³ of dredged sediment. The enlargement of Harbor layout facilitates the

deposition of contaminated sediment over a confined facility that is isolated from the marine habitat. As a consequence, there is no consumption of electricity and water, and GHG and other emissions are lower compared to other options (total NO_x emissions -25%; total SO_x emissions -30%; total PM₁₀ emissions -15%). The total energy used for recreational areas is

much higher (263.743 kWh) compared to other solutions (+20%) (Table 2). In Fig. 4 an example of SiteWise™ outputs is reported to show the GHG emissions, the total energy used and the water consumption of each component for the scenario 1 (recreational area for sport activities). The highest impact component is "sediment characterization and dredging" (C1). Also the "treatment" component (C3), has a relevant footprint. On the other hand, the "transport" and "sediment repositioning" components require less resources. Component C2 (Transport of sediment) has a lower footprint, and the impact of the components "transport" and "treatment" are null. The alternatives "recreational areas" and "coastal restoration" lead to similar impacts (e.g. GHG emissions of 11.632 kg/m³ and 11.623 kg/m³ respectively). While those relating to the "port layout" alternative are significantly lower (e.g. GHG emissions of 90.42 kg/m³).

GHGs emission is higher during the "treatment" than "transport" and "sediment repositioning" (transport / C3) to the storage area (which respectively are equal to total amount of 46.000 kg and 21.000 kg and are almost irrelevant (Fig. 4 top). Different values were obtained for PM₁₀ emission: characterization and dredging (C1) is significantly higher (1.230 kg) than minor contribution of transport (1.8 kg), treatment (277.3 kg) and sediment repositioning (10.9 kg).

The total amount of onsite PM₁₀ emission is lower in case of direct placement of dredged sediment into a CDF, while an increase of about 3% is observed with other scenarios.

5.2. Economic analysis of remediation technologies

According to the preliminary analysis by Ferrantini (2012), which showed the costs for different treatment processes, in this study we adopted soil

washing as the most convenient and effective solution. The alternatives have an economic impact that is influenced by the processing chain for clean-up operation and installation of the destination area. Such differences in the management option produce differences in the estimated expenditure (Table 7). The alternative "Preparation of recreational areas" has a higher cost (\$ 1,566,745, i.e. € 1,368,500) due to the work for the preparation of the beach volleyball fields (the estimated cost is around € 200,000). The cost of preparation increases the intervention cost of "Coastal areas restoration", as "Preparation of recreational areas" (the preparation of the beach volleyball fields) have to be added. Table 8 summarizes the impact of each scenario on the environment. It can be seen that the Harbor scenario is the only one having low impact in terms of water use and electricity consumption. More detailed information of environmental impacts of the studied scenarios is provided by Fig. 5, where the impacts are normalized. All metrics are normalised (the alternative with the highest impact is assigned a value of 1.0 and impacts for the other alternative metrics are presented as ratios to that unit). The alternatives "Sport Area" and "Coastal restoration" are very similar.

6. Discussions

Dredging within Favignana's Harbor is periodically needed to ensure the safety of navigation and an adequate depth of the sea floor. This study evaluated three possible types of interventions on the territory: Sports facilities; Coastal restoration; Harbor, as the large part of dredged material is slightly contaminated. In this study, SiteWise™ (v3) was used to evaluate a dredging project not only to determine the impact of different stages of the remediation, but also to understand which management options has the lower environmental footprint.

Table 6. Comparison among the scenarios (data refer to m³ of sediment)

<i>Scenario</i>	<i>GHG emission</i>	<i>Total energy used</i>	<i>Water consumption</i>	<i>Electricity used</i>	<i>Onsite NOx emission</i>
	metric kg/m ³	kWh/m ³	m ³ /m ³	kWh/m ³	metric kg/m ³
recreational areas	116x10 ²	346.35	294x10 ⁻²	1.38	105
port layout	90x10 ²	279.75	0	0	103
coastal restoration	116x10 ²	346.35	294x10 ⁻²	1.38	105
<i>Scenario</i>	<i>Onsite SOx emissions</i>	<i>Onsite PM₁₀ emissions</i>	<i>Total NOx emissions</i>	<i>Total SOx emissions</i>	<i>Total PM₁₀ emissions</i>
	metric kg/m ³	metric kg/m ³	metric, kg/m ³	metric, kg/ m ³	metric, kg/m ³
recreational areas	160x10 ⁻¹	516x10 ⁻²	148	27	7x10 ⁻²
port layout	150x10 ⁻¹	516x10 ⁻²	113	19	6x10 ⁻²
coastal restoration	160x10 ⁻¹	499x10 ⁻²	149	27	7x10 ⁻²

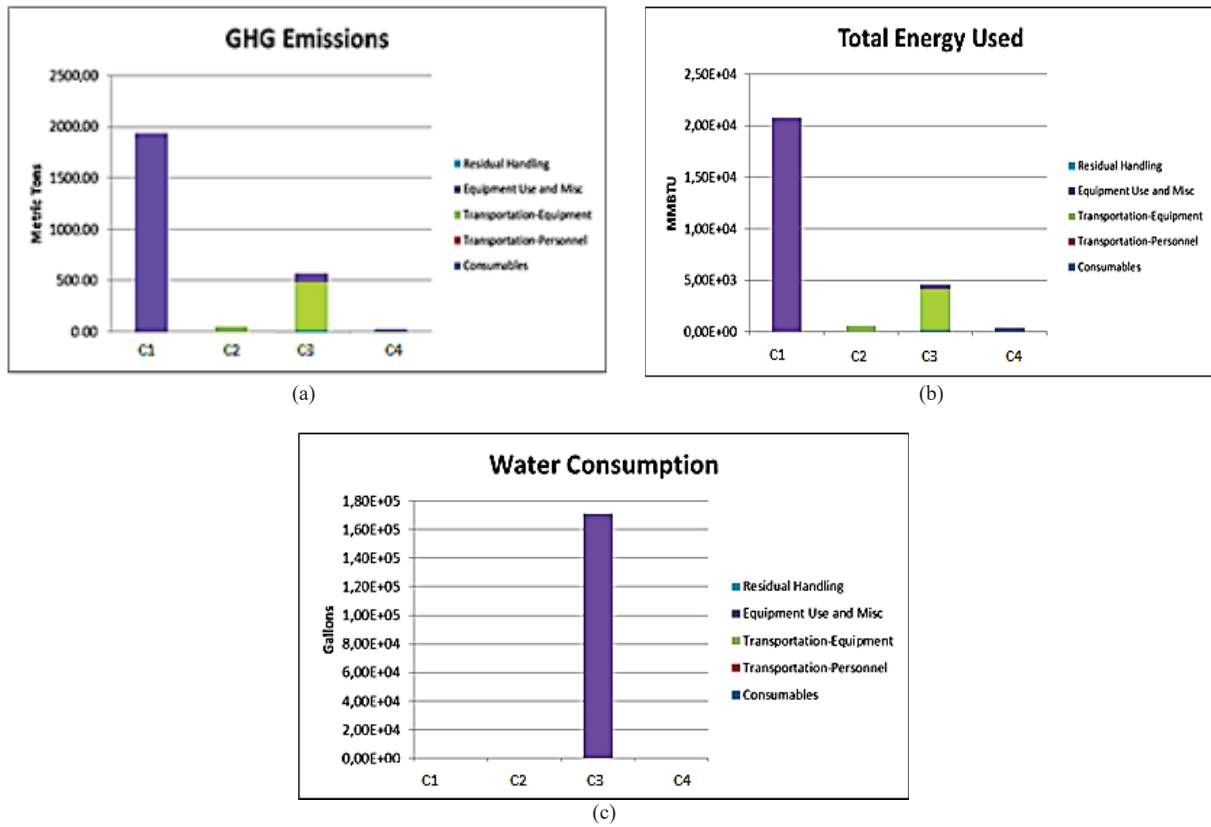


Fig. 4. Example results of Scenario 1 (recreational area for sport activities) produced by SiteWise™. GHG emissions, total energy consumption and water consumption are plotted for each component (C1: Sediment characterisation and dredging operation; C2: Transport; C3: Treatment; C4: Coastal intervention) with US\$ Unit

Table 7. Results of the economic evaluation

Scenario	Stages of remediation and reuse of sediment	Cost	Total cost
N	€	€	€ (\$)
1 Sport	Sed. Characterization:	25,000	1,368,500 (1,566,745)
	Sed. Analysis:	6,000	
	Sed. Dredging (6.7 €/m ³):	148,000	
	Sed. Transport:	13,500	
	Sed. Treatment:	994,000	
Installation of beach volley:		200,000	
2 Coast	Sed. Characterization:	25,000	1,211,000 (1,368,000)
	Sed. Analysis:	6,000	
	Sed. Dredging (6.7 €/m ³):	148,000	
	Sed. Transport:	13,500	
	Sed. Treatment:	994,000	
Requalification of coastal area:		16,000	
3 Harbor	Sed. Characterization:	25,000	1,079,000 (1,218.89)
	Sed. Analysis:	6,000	
	Sed. Dredging (6.7 €/m ³):	148,000	
	Sed. Transport:	0	
	Sed. Treatment:	0	
Infrastructure (new harbor layout):		900,000	

The present study confirms the findings of Kim et al. (2013) related to the relevant contribution of soil washing treatment compare to the other stages of remediation projects. The consumable chemicals, electric energy consumption for system operation, and equipment use are the major sources of environmental pollution to occur during the soil washing process. The results of our study demonstrated that the footprint of a Confined Disposal Facility (CDF) option is lower

because the reuse of dredged contaminated sediment on coastal marine environment certainly need a lower concentration of contaminants and ecotoxicology level compare to dredged material that can be spilled over a completely isolated coastal infrastructure. The total energy used for recreational areas is much higher compared to other solutions (+20%), because the installation of beach volleyball facilities requires additional materials and components (Table 2).

Table 8. Impact categories Table of normalized impact. The outputs reported with red (high), yellow (medium) and green colour (low) are based on a 30% difference (if the two data points are within the 30% difference then both the alternatives are assigned the same high, medium, or low index)

Scenario	GHG Emissions	Energy Usage	Water Usage	Electricity Usage	Onsite NOx Emissions
Sport	High	High	High	High	High
Coast	High	High	High	High	High
Harbor	High	High	Low	Low	High
Scenario	Onsite SOx Emissions	Onsite PM10 Emissions	Total NOx emissions	Total SOx Emissions	Total PM10 Emissions
Sport	High	High	High	High	High
Coast	High	High	High	High	High
Harbor	High	High	High	High	High

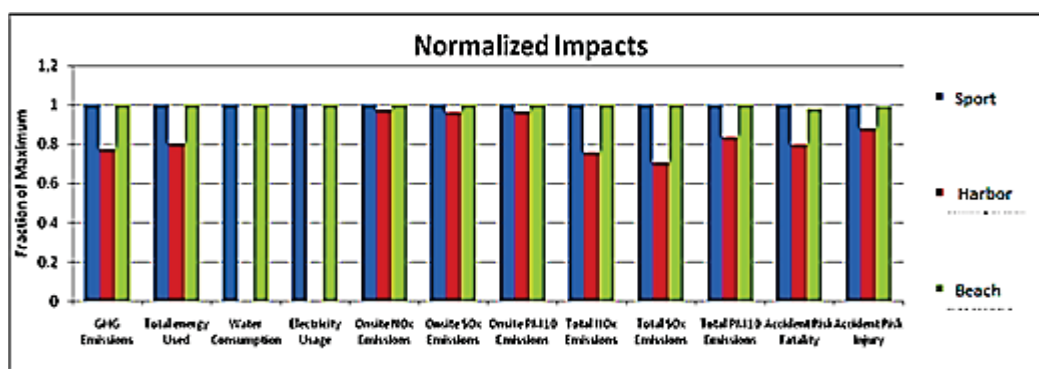


Fig. 5. Normalized impact category diagram generated in the Final Summary Sheet of Site Wise for each scenario

The highest impact component was "sediment characterization and dredging" (C1), as it involves the use of specific equipment with high power and high consumption. Also the "treatment" component (C3), has a relevant footprint as it consumes a lot of water. On the other hand, the "transport" and "sediment repositioning" components require less resources. Component C2 (Transport of sediment) has a lower footprint because the distances to be covered on the small island is reduced and the equipment uses technologies with lower impacts compared to the other components. The impact of the components "transport" and "treatment" are null due to the enlargement of the Harbor layout and direct discharge of sediment from the dredger into the CDF. The alternatives "recreational areas" and "coastal restoration" led to similar impacts, while those relating to the "port layout" alternative were significantly lower.

As shown in Table 8 and Fig. 5, the alternatives "Sport Area" and "Coastal restoration" are very similar because the components "characterization and dredging" and "treatment" are mandatory for those scenarios and the transport component is not considerably different as the island is small and, as a consequence, distances are very limited. Moreover, the "storage" component (within a temporary sediment deposit) has a lower impact compare to "characterization and dredging" and "treatment" component. The lowest impact is generated by the enlargement of Harbor layout scenario. In fact, the two components of "transport" and "treatment" are

avoided with this option. However, it has some impact during the immediate intervention, as the use of reclaimed tanks into which contaminated sediment is located, does not allow omitting future monitoring and intervention. Moreover, in that scenario the sediments are considered as waste and not as a resource, like in the other two alternatives.

A CDF (Confined Disposal Facility) was the cheapest option and presents lower water and energy consumption. Beach replenishment along the coast is the beneficial use that most authorities should follow. The on-shore recreational activities (like beach volleyball fields) is an innovative promising option, indicated for reduced dredged volume of sediment because it favour the reuse and increases the amount of goods and tourism services. However, the possibility to realize beach replenishment will be strongly influenced by the eco-toxicology of dredged sediment and the presence of the *Posidonia oceanica* meadow in shallow water (close to the shore line and within the active zone of the submerged beach).

From the economic point of view, the parameters that influence the operations of the considered process have a direct influence also on the respective costs. Those parameters are: volume of sediments to be dredged, seagrass meadow, and extension of the storage area to be waterproofed. It is important to observe how the scenario that involves the construction of a CDF within the Favignana Harbor as well as lower consumptions of water and electricity (generated by the lack of treatment) also generates lower emissions of GHGs in the

atmosphere. Global climate change is actually one of the major environmental issues of the present and the future (GEO-5, 2012). Evidence for global climate change is increasing and there is a growing consensus that the most important cause is humankind's interference in the natural cycle of GHGs (Crutzen and Stoermer, 2000; 2013). This study confirms that human activities enhance and influence the emission of natural greenhouse effect by adding other GHGs such as carbon dioxide and nitrous oxide to the atmosphere (Pallottini and Cappucci, 2009; Pascucci et al., 2008; Stoddart et al., 2019).

Even if green and sustainable terms are sometimes interchanged, green remediation can be associated to environmental components, while sustainable remediation can be associated with environmental factors, social responsibility and economic aspects. In fact, these two notions can be connected as Green and Sustainable Remediation (GSR), addressing a broad range of environmental, social and economic impacts during all remediation phases (Reddy and Adams, 2010). The results obtained in our study revealed that enlargement of the port layout is the more cost and time effective option even if sediments (as un-renewable natural resources), are taken out of the coastal system and used to fill the new infrastructure.

Furthermore, from a regulatory point of view, there are still no laws to manage in a quantitative way the application of the sustainability concept and apply it to practical cases. In addition, the complexity of the sediment regulations for water and waste management, increase the complex application of sustainability criteria to the sediment management options.

7. Conclusions

Dredging of sediment with low contamination levels harbors is often required to facilitate recreation and beach formation. However, the sustainability of land reclamation is a major challenge and presents many obstacles, including lack of economic incentives, use of lower impact technologies and limitations in acceptance of new technologies by companies. In addition, technical restrictions must be resolved (i.e. data input should be defined and standardized at national and international level). The methodological approaches and evaluations as well as the ongoing research of technologies need to deliver quantitative analysis and must be carried out considering a conjoint protocol. At the same time, the high costs of low impact technologies lead to the choice of traditional and established equipment and know-how, such as, in the case of excavation sediments, removal and disposal

Reuse of sediment often meets the waste status of dredged sediments. When a sustainable approach could lead to regeneration and reuse of material in various sectors, the legislation does not always support a sustainable approach to managing the sediments. Waste regulations often hamper reuse

projects with environmental constraints which were not designed for sediments reuse. They are often country specific and make EU-wide projects difficult. However, reuse can provide environmentally beneficial options for site restoration, for fresh (river) and coastal (marine) water good status as indicated by the Water Frame Directive 60/2000 and for climate change mitigation. The Italian legislation is continuously evolving, but still does not consider risk analysis as a decision tool for dredging the contaminated sediment. Italian national legislation recently set out the criteria for the classification of sediments and their possible reuse or disposal offering a more comprehensive regulatory framework for contaminated sites. However, areas outside of the contaminated sites of national interest still require specific attention.

The use of SiteWise™ seems a promising tool in applied sedimentology and coastal engineering even if the suggested alternative scenarios are not necessarily always accepted or welcomed by local authorities and stakeholders. Stakeholder could adopt the results to manage the dredged sediment in other sites considering different options and related environmental footprint (e.g. consumption of natural resources and energy, and emissions into atmosphere).

Results of the present study are useful for the decision-making phase and for the competent authorities, but also decisive for the scientific community as they integrate results of conventional techno-economical assessment studies. Specific attention should be placed on implementing low energy demand technologies, particularly if electricity mix relies on fossil fuel and this scenario may impact the footprint of sediment management options.

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REAL TIME MONITORING OF WATER QUALITY IN AN AGRICULTURAL AREA WITH SALINITY PROBLEMS

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Abstract

Agriculture is a highly water-demanding sector. Developed in recent years, the precision farming approach allows to optimize irrigation without compromising crops productivity. WSN networks are a key element of this approach because they allow to monitor continuously large number of parameters providing the possibility of a real-time intervention on field management practices. The WSN networks can be used to measure traditional parameters such as precipitation, soil moisture, or irradiation and others such as the quality of irrigation water and groundwater. The qualitative monitoring of these parameters is essential when the cultivation is carried out under complex conditions such as those represented by soils with salinization problem. This work fits this context by presenting the results of the first 13 months of an experimental campaign aimed at the measurement of soil, water (quality of irrigation and drainage water of the fields) and groundwater parameters by a WSN system. This paper analyzes results of this activity and provides practical suggestions to ensure a more efficient system.

Keywords: Monitoring Precision Farming; salinity; water management; WSN

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1. Introduction

Agriculture, and irrigated agriculture in particular, is by far the sector with the highest use of water. Globally it accounts for around 70% of world water withdrawal (WWAP, 2015). Irrigated agriculture represents 20 % of the total cultivated land but contributes 40 % of the total food produced worldwide (www.fao.org). Irrigation water withdrawal greatly exceeds the need for irrigation water due to significant losses in both fields and distribution systems. Consequently not all water taken from a source reaches the root zone of the plants and so irrigated land does not fully meet its production target (Afrasiabikia et al., 2017). In many countries including Italy (Canone et al., 2015) and others in the Mediterranean area (Iglesias et al., 2007; Levidow et al., 2014), efficient water use and management are

today's majors concerns. In recent years this has pushed farmers to investigate the possibility of using moderately saline water for irrigation purposes (Wang et al., 2017), however by adding salts to the soil via irrigation, it may lead to soil salinization and crop yields reduction. It should also be considered that especially in the Mediterranean region, many aquifer systems, that naturally contain vast quantities of brackish water, have limited possibilities for exploitation for human or agricultural uses, imposing so, additional demand stress to neighboring aquifers with higher water quality. Also saline intrusion is an important concern in aquifers, where as a result of the high seasonal water demand, mainly for tourism, they have been over pumped (Iglesias et al., 2007).

New technologies (e.g. soil moisture, water table depth, electrical conductivity and canopy sensors) can allow scheduling irrigation by following

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plant needs. This together with good agricultural practices will consent to reduce water withdrawal and chemicals without compromising crop productivity (Levidow et al., 2014). The use of information technologies (IoT) in agriculture is frequently known as "Precision farming" (Auernhammer, 2001). The key component of this farm management approach is the use of IoT and a wide array of items such as control systems, sensors, robotics, drones, autonomous vehicles, variable rate technology, GPS-based soil sampling, automated hardware, telematics, and software to optimize the growing of crops (Barnes et al., 2019; Zamora-izquierdo et al., 2018).

Sustainable irrigation is a key element of precision farming and it mainly relies on the efficient use of water avoiding soil degradation. A sustainable use of water resources, for the irrigation of soils suffering of problems connected with salinization, must take into account different factors such as: the quality of irrigation water, crop requirements, and salt concentrations in soils (Libutti et al., 2018; Peragón et al., 2018) The measurement of all these parameters through a sensor network offers the possibility to optimize irrigation while protecting the overall environment. Wireless sensor networks (WSNs) can be used in agriculture to provide farmers with a large amount of information. Jawad et al. (2017) provided a detailed review of the WSN-based agriculture applications by comparing communications protocols, energy harvesting techniques and presenting the most used sensors and actuators. However, this document does not contain any information concerning the use of WSNs for monitoring parameters related to water salinity.

Salinity problems exist when the concentration of salt accumulated in the crop's roots zone causes a loss in yield. It may be caused by 2 main factors: a) primary salinity due to natural causes; and b) secondary salinity due to irrational land use and inappropriate agricultural practices. The first occurs in both soils and waters, and it is often associated with certain types of relief, geomorphological and

hydrogeological conditions such as a high groundwater table and impeded drainage or poor drainage. Secondary salinity is caused by an excessive water inputs via irrigation that, in the absence of appropriate drainage systems, leaches the soils causing a rapid raising of the groundwater table (Tables 1, 2)(Vargas et al., 2018).

The accumulation of salt in the root zone causes the impossibility of extracting enough water from the salty soil solution by roots, resulting in a water stress (Ayers and Westcot, 1985). Salts that contribute to a salinity problem are water soluble and readily transported by water. The electrical conductivity (EC) is the parameter used to measure the water and soil salinity, and it is usually reported in deciSiemens per meter at 25°C (dS/m).

Waters and soils salinity classes generally recognized are given in Tab. 1 and Tab 2 respectively, while a detailed description of the grade of soil salinity as a function of the chemistry of salinization is presented in Vargas et al. (2018). Usually water sourced from snow-fed rivers, has a total salinity of less than about 0.5 to 0.6 dS/m, groundwater in semi-arid region has a salinity in the range 1-15 dS/m, and sea water has an average total soluble salts content of about 35 g/l corresponding to an electrical conductivity of about 50 dS/m. As a result of this irrigation water ranges between a wide range of salinity values. The higher the total salinity of an irrigation water, the higher is its salinity hazard for the crops if the soil and climatic conditions and the cultural practices remain the same.

When farmers deal with problems connected to salinity, it is important to evaluate all the factors that caused them such as: soil salinization, poor quality of irrigation water, unfavorable climatic conditions, seawater intrusion, and poor management; in order to identify the factors on which to intervene. The precision farming approach combines perfectly with this process because it allows to monitor all the variables and therefore to understand where, how, and when to act.

Table 1. Classification of saline waters, adapted from Rhoades et al. (1992)

<i>Water class</i>	<i>Electrical conductivity dS/m</i>	<i>Salt concentration mg/l</i>	<i>Type of water</i>
Non-saline	<0.7	<500	Drinking and irrigation water
Slightly saline	0.7 - 2	500-1500	Irrigation water
Moderately saline	2 - 10	1500-7000	Primary drainage water and groundwater
Highly saline	10-25	7000-15 000	Secondary drainage water and groundwater
Very highly saline	25 - 45	15000-35 000	Very saline groundwater

Table 2. Classification of saline soils (adapted from Rhoades et al. (1992))

<i>Soil Salinity Class</i>	<i>Conductivity of the Saturation Extract (dS/m)</i>	<i>Effect on Crop Plants</i>
Non-saline	0 - 2	Salinity effects negligible
Slightly saline	2 - 4	Yields of sensitive crops may be restricted
Moderately saline	4 - 8	Yields of many crops are restricted
Strongly saline	8 - 16	Only tolerant crops yield satisfactorily
Very strongly saline	> 16	Only a few very tolerant crops yield satisfactorily

Integrated in this context, the LIFE AGROWETLANDS II research project – SMART WATER AND SOIL SALINITY MANAGEMENT IN AGRO-WETLANDS – aims to counteract the soil degradation and the wetlands natural ecosystems alteration through a targeted and efficient management of the water resources (precision farming approach). The project provides for the implementation of a smart irrigation management system - SMART AGROWETLAND - that, by monitoring weather, soil, groundwater, channel water and crops parameters will formulate irrigation recommendations (decision support systems, DSS) to support farmers’ decisions (Masina et al., 2019).

In this frame, this paper will present the architecture and the results obtained after 13 months of monitoring activity of the wireless sensor network (WSN) developed within the described project, highlighting benefits, limits of applicability, possible improvements, and strategies to optimize the operational costs.

2. Material and methods

2.1. Project architecture

The overall architecture of the SMART AGROWETLANDS II is depicted in Fig.1. It is essentially organized into three modules: the monitoring system, the data cloud and analytics, and a Decision Support System (DSS) into a web environment which provides irrigation recommendations.

The monitoring system consists of two subsystems: a) a monitoring via WSN, b) and a traditional manual monitoring (Fig. 1). The first deals essentially with real-time monitoring of environmental data (soil, ground water, canal water, irrigation water); the other consists of a manual data collection of field data, the post processing and the upload into the cloud. This last sub-system includes measurements of agricultural (agricultural workings, fertilization, canopy cover, etc.) and ecological parameters (William et al., 2001).

The WSN is an innovative on-line system composed by a group of spatially dispersed and dedicated sensors for monitoring and recording the physical parameters of soil, ground water, surface water and weather. The WSN is based on IEEE standard 802.15.4 (Adams, 2006), which focuses on a low-cost and low-speed communication between nearby devices with little to no underlying infrastructure, and lower power consumption.

The WSN is composed by different nodes, which basically are measurement points. There can be three type of nodes (Fig. 2): the “S-node” that is equipped only with sensors for soil monitoring, the “P-node” which is equipped with a soil sensor and a water sensor inserted in a nearby piezometer; the “I-node” which is located close to a canal and has only water quantity and quality sensors. Each WSN node

can serve as router or gateway. A router is a node which collects and transmits information to another router or to a gateway. A gateway is a coordinator node, and usually it integrates a weather station “M-node”, and it is responsible to the sending of monitoring data to the cloud.

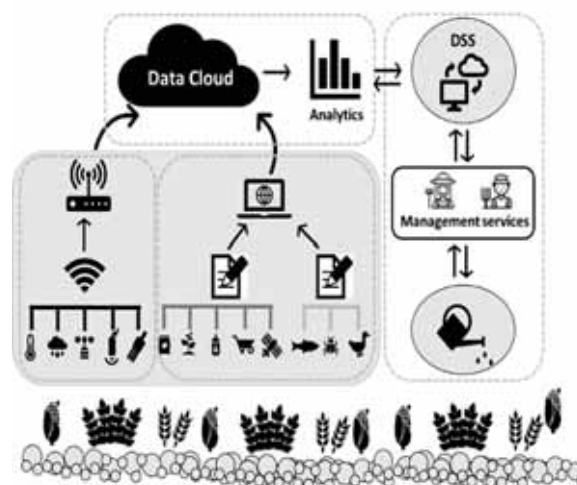


Fig. 1. Overall architecture of the SMART AGROWETLANDS platform

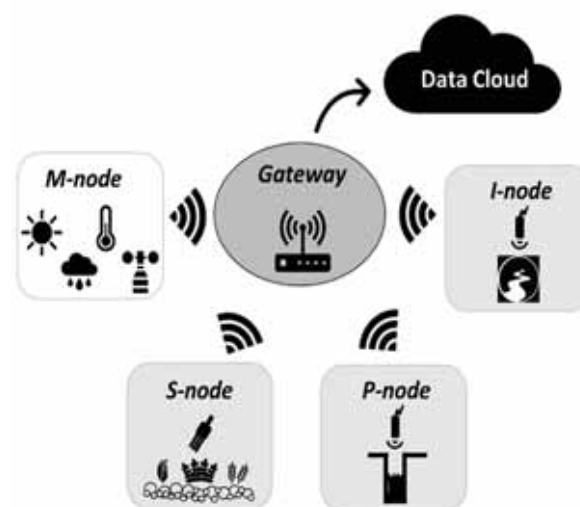


Fig. 2. Overall structure of the Wireless Sensor Network (WSN)

Nodes have been equipped with the following sensors:

- **Decagon CTD-10** - The Decagon CTD-10 sensor is a low cost, accurate tool for monitoring water level, electrical conductivity, and temperature in both ground water and surface water. The sensor utilizes a vented pressure transducer to obtain an accurate water level measurement from 0 to 10 m while removing the effects of barometric pressure. With a range of 0 to 120 dS/m, the CTD sensor has the ability to make accurate electrical conductivity measurements in a broad range of applications.

- **Decagon GS3** - The GS3 soil moisture, temperature, and EC sensor is built with an epoxy body and stainless-steel needles. The internal circuitry is the same cutting-edge design that you can find in other Decagon soil moisture sensors, but the form factor has been optimized for use in soilless substrates or harsh environments, giving it a wider range of EC measurement and an increased temperature range. Not only do the steel needles improve sensor contact, but they also improve the sensor's ability to measure EC in porous substrates such as peat or perlite.

2.2. Case study

The study area (Fig. 3) is located in the northern part of Italy, between the Reno River to the north, the Lamone River to the south, and the coastline of the Adriatic Sea. It includes rural and agricultural land, with a high landscape value, as well as a significant number of coastal wetlands, brackish and otherwise where salinity is a fundamental controlling factor for wetland water chemistry and biodiversity (Antonellini and Mollema, 2010; Smith et al., (2007); Turnbull et al., 2007).

The pilot site is composed by 5 farms managed by a co-operative (Agrisfera, 2019) for a total surface of 609 ha mostly located close or below the sea level. The area is affected by soil salinization, salt water intrusion, and it has a shallow water table (Antonellini and Mollema, 2010; Giambastiani et al., 2007; Lamberti et al., 2018). This is essentially due to the fact that, during the second half of the 19th century, the area was converted from a wetland to an agricultural zone through hydraulic land reclamation. Soil texture ranges from clay loam to sandy loam with poor

internal drainage. There is a shallow water table present within 2.5 metres from the surface in most of the study area. The climate is humid subtropical and rainfall ranges between 800-900 mm per year (Felisa et al., 2013).

The drainage system consists in 69 km of canals of different sizes (the lower the width the higher the order of the canal indicated in Fig. 3) and two dewatering pump systems (the main characteristics are summarised in Table 3) which guarantee the minimum depth to water table in the fields, it means that drainage is carried out almost exclusively mechanically. Canals have a primary function of drainage and, some of them, a secondary of irrigation (Cipolla et al., 2018a).

Table 3. Characteristics of the pump systems

ID Pump systems	Drained area [km ²]	Head [m]	Flow rate [m ³ /s]
1° Bacino Mandriole	18.99	4.35	6.00
2° Bacino CasalBorsetti	47.38	2.96	0.87

Among all canals, only the “Canale di Bonifica Destra Reno (CBDR in Fig.3) which runs through the study area in an east-west direction and it is parallel to the Reno river, drains naturally to the Adriatic Sea. It is dammed on both banks along its whole extension and it equips a water control gates to avoid conveying seawater inland (red line in Fig. 3). During the summer season the water control gate is closed to guarantee a higher upstream water level, and so the possibility to use the water for irrigation purposes (Cipolla et al., 2018b).

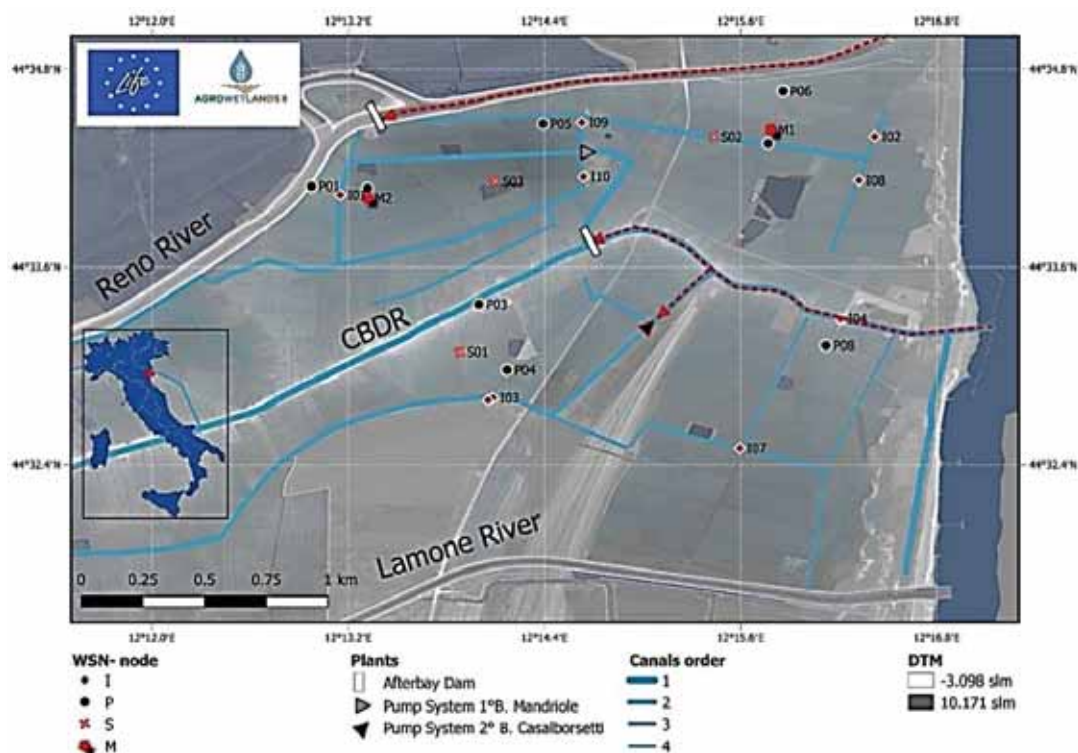


Fig. 3. Case study area

The study area is mainly cultivated with summer crops such as: maize, alfalfa, sorghum and sunflower both in traditional and organic way. Rainfall does not play a significant role in meeting crop water demand or leaching requirement and then irrigation season begins on April and finishes on the end of July/August depending on the crop.

Irrigation water comes from surface water and it is withdrawn from the Reno River and the CBDR. The first source serves, through two pump systems, a pressurised irrigation networks called "distretto irriguo in pressione", and a gravity open pipe called "Canaletta Mandriole". The second source is the CBDR and the water withdrawn through a pump and a complex systems of sluice gates is sent to the Rivalone canal. The most used irrigation systems are: traveling sprinklers and centre pivots with drop sprinklers.

2.3. Wireless Sensor Networks

The WSN is composed by 19 nodes organised into 6 subnetworks. It means that 6 gateways guarantee the transmission of monitoring data to the cloud once per hour. Fig. 3 shows the positions of the 11 *I-nodes*, 8 *P-nodes*, and 3 *S-nodes*, while Table 4 illustrates the type of sensors installed in each node and the date of installation. Monitoring data are acquired with a 10 minutes time step. The CTD10 sensors allow measuring the temperature, the water depth and electrical conductivity. They are installed in both *P-node* and *I-node*. In-canal installations were carried out by positioning the sensors in the centreline

of the channel, when possible, or near a bank otherwise. *P-node* installation of CTD10 sensors was realised between 2 and 3 meters below the ground level. GS3 soil sensors were located 50 cm below the ground level.

3. Results and discussion

3.1. M-node and weather data

The WSN network is equipped with 2 weather stations 3.2 km away from each other. The M2 and M1 weather station are respectively 5 and 1.5 km from the Adriatic Sea. The traditional wind rose plots, illustrated in Fig. 4, show how wind speed and direction are distributed at M1 (a) and M2 (b) weather stations. The prevailing winds recorded on M1 come from the NW and NE with maximum speeds reaching 130 m/s. M2 station is more sheltered from the wind, the maximum speed measured is less than half (52 m/s) and the prevailing winds come from the SE.

This is likely because the right bank of the Reno river and the weather station are only 400-500m distant and the first is 4-5m higher than the second, sheltering the weather station from winds coming from NE and NW. Fig. 5 shows the cumulative daily rainfall (a and c) and the average, maximum and minimum daily air temperature (b and d) recorded at M1 and M2 stations respectively.

It may be observed that there are almost no differences in terms of temperature. On the contrary, the rainfall variability between the two stations is really accentuated.

Table 4. Characteristics of the WSN nodes

<i>SUB-NETWORK</i>	<i>ID</i>	<i>Type</i>	<i>CTD-10</i>	<i>GS3</i>	<i>Weather</i>	<i>Role</i>	<i>Date Installation</i>
GATTOLO INFERIORE	P02	P-node +S-node +M-node	1	1	1	G	10/11/2017
	P01	P-node +S-node	1	1	-	R	10/11/2017
	I01	I-node	1	-	-	R	01/01/2018
AUGUSTA	S01	S-node	-	1	-	G	29/03/2018
	P03	P-node +S-node	1	1	-	R	29/03/2018
	P04	P-node +S-node	1	1	-	R	06/04/2018
	I03	I-node	1	-	-	R	29/03/2018
	I11	I-node	1	-	-	R	29/03/2018
MARCABO' EAST	P07	P-node +S-node +M-node	1	1	1	G	10/08/2017
	P06	P-node +S-node	1	1	-	R	10/08/2017
	I02	I-node	1	-	-	R	01/01/2018
	I08	I-node	1	-	-	R	16/03/2018
	S02	Soil	-	1	-	R	06/04/2018
MARCABO' WEST	I10	I-node	1	-	-	G	06/04/2018
	I09	I-node	1	-	-	R	06/04/2018
	P05	P-node +S-node	1	1	-	R	06/04/2018
	S03	S-node	-	1	-	R	06/04/2018
BARONIA	P08	P-node +S-node	1	1	-	G	16/03/2018
	I04	I-node	1	-	-	R	04/12/2018
	I07	I-node	1	-	-	R	16/03/2018
S. ALBERTO	I05	I-node	1	-	-	G	04/12/2018
	I06	I-node	1	-	-	R	04/12/2018
TOTAL			19	11	2	6 G+16 R	

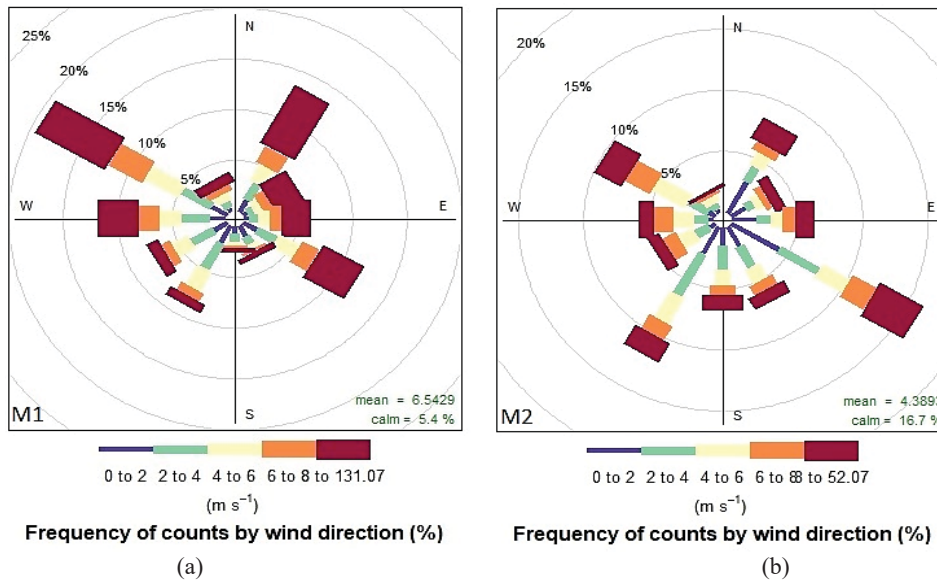


Fig. 4. Wind rose plot for the M1 (a) and the M2 M-node (b)

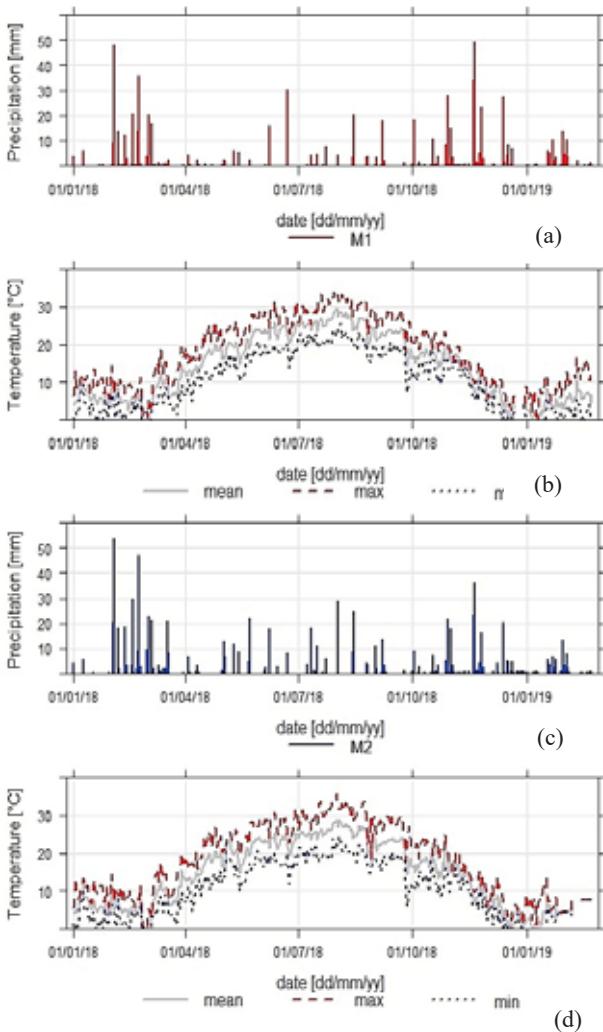


Fig. 5. Representation of: (a) the daily cumulative rainfall depth and (b) the minimum, average and maximum air temperature for the weather station M1; and of the daily cumulative rainfall depth (c) and minimum, average and maximum air temperature (d) for M2 weather station

Both rain gauges installed in the two weather stations are "tipping bucket" and have a tolerance depth of 0.1 mm. The cumulative rainfall recorded during the observation period (01/01/2018-22/02/2019, 417 days) for M1 and M2 was respectively equal to 715.3 and 854.3 mm, which corresponds to a percentage variation of 16% (Table 5).

The measurement of precipitation is very sensitive to exposure, and to wind. The differences in terms of wind exposure described above could therefore be the main cause of this difference.

Table 5. Cumulative monthly rainfall depth for weather station located on M1 and M2 and differences between them

NODE-ID	M1	M2	M1-M2
SU	mm	mm	Mm
Jan-18	11.0	10.5	0.5
Feb-18	162.0	210.8	-48.8
Mar-18	49.0	103.5	-54.5
Apr-18	8.5	11.5	-3.0
May-18	19.8	68.5	-48.8
Jun-18	46.5	32.5	14.0
Jul-18	18.0	41.5	-23.5
Aug-18	35.8	70.3	-34.5
Sep-18	25.5	32.0	-6.5
Oct-18	76.8	53.8	23.0
nov-18	141.3	111.5	29.8
Dec-18	51.3	46.5	4.8
Jan-19	48.8	45.3	3.5
Febr-19	21.3	16.3	5.0

Making an analysis only during the irrigation period (Apr-Aug) the differences sharpen considerably. The cumulative rainfall recorded in this period is equal to 128.5 and 224.3 mm on M1 and M2 respectively, which corresponds to a percentage variation of 42%. The high variability of rainfall data

between the two stations, particularly during the irrigation period, suggests the importance of installing a dense network of rain gauges in the area that is grown using the precision farming approach. In the near future the use of precipitation radar data, which in Emilia Romagna region are supplied free of charge with a resolution of 500*500m could be exploited to reduce the costs associated with the installation of multiple rain gauges (Cipolla et al., 2019).

3.2. Water level and salinity in I-node

All sensors located in *I-nodes* allow estimating the quality of water returned to the sea by the canal system, while some of them (I03, I05, I06), located in canals used for both irrigation and drainage purposes, provide information also on the irrigation water quality.

Fig. 6 depicts the monitoring data collected by the *I-node* of the WSN. Water levels in canals generally vary proportionally to rainfall volume, rising during intense meteoric events, and lowering in dry

weather. However, many canals (I01, I09, I07) show an artificial level variation which is caused by the pump system downstream. Moreover, during the irrigation season, the water levels are kept high thanks to the introduction of fresh water into the network through the irrigation systems, following the purpose of countering the shallow water table. This management practice is clearly visible in node I01, I02, I07 and I09.

EC values are strongly variable. Generally, the highest values are in winter and the lowest in summer, as showed in Fig.7 for nodes I01, I03, I07, and I10. The highest EC values were recorded almost in each sensor in winter 2017/2018 probably because 2017 was much drier than 2018.

This behaviour may be mainly caused by 4 factors: a) in winter the canals collect the waters that leach the soils; b) since all the canal beds range between -2.39 and -0.34 s.l.m., they collect also saline groundwater; c) in summer a large amount of fresh water is pumped in canals; 4) irrigation water has a good quality.

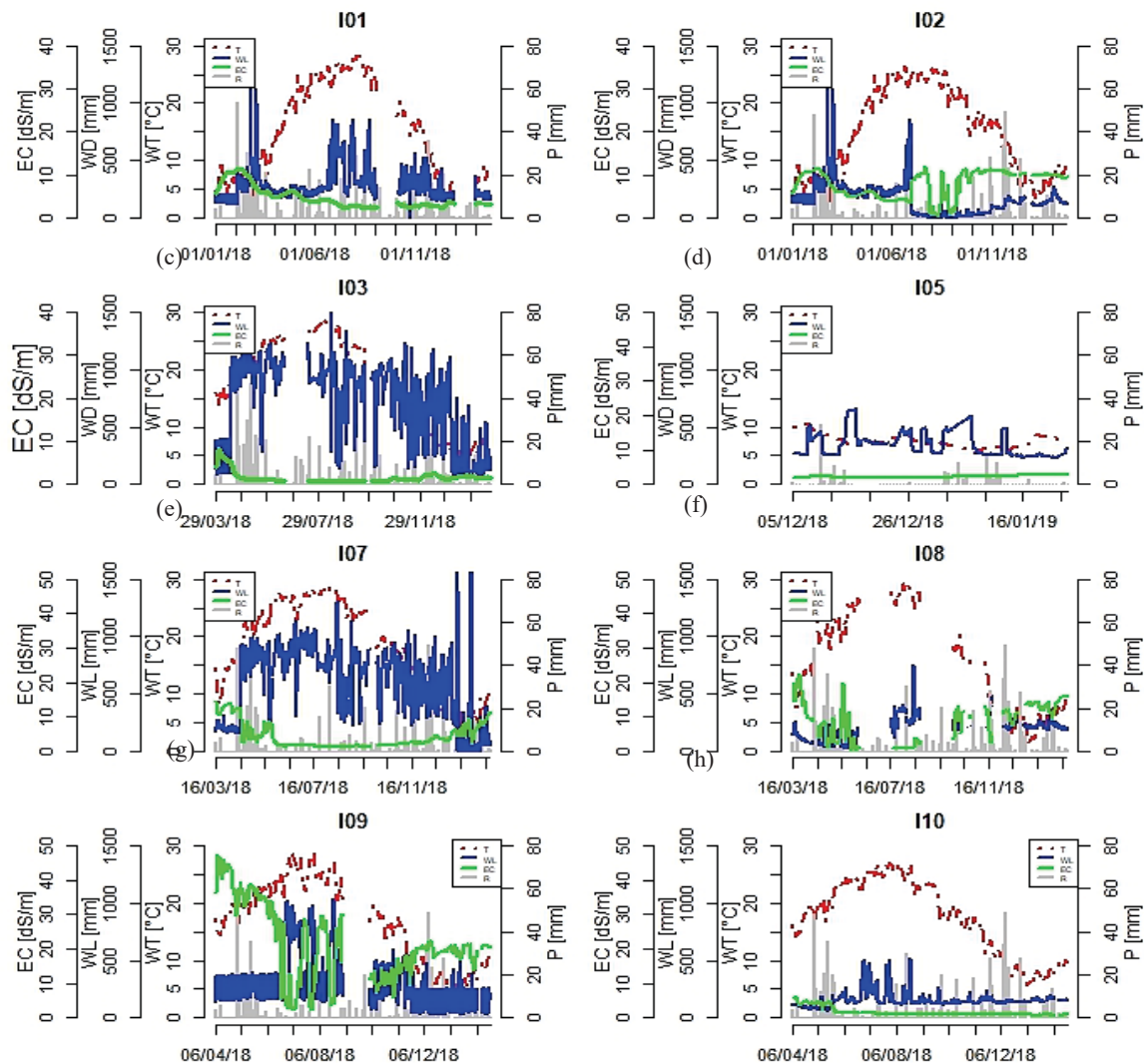


Fig.6. Daily cumulative rainfall depth of the weather station closer to the I-node (P, gray), average daily air temperature (T, red); average hourly water level in canal (WL, blue); average hourly electrical conductivity (EC, green) measured in nodes I01 - I10 during the monitoring period

The use of a real time control system, such as the one provided by the WSN, makes it possible to monitor the operation of the sensors in each moment. This allows to highlight both punctual anomalies and long-term anomalies of the data acquired. For example, the nodes I02, I08 and I09 present anomalies in terms of EC. Regarding the first two nodes, these anomalies are found between July and September 2018 and in all the months except July 2017 for node I08. I02 presents very uneven EC values during the summer, this is because the presence of water depth close to zero, as often happens in summer, the sensor measures the EC value of stagnant water, and these values should be analyzed with caution.

The behavior of node I08 is the opposite, during the winter the level is almost always close to zero and then EC rises, while during storms it drops. Upstream of the I08 there is the outfall of the “Canaletta Mandriole” irrigation system, and the low EC value indicate that during July and August a good amount of fresh water was discharged into the canal. Such water may be used by farmer for irrigation purposes. The I09 hydrometer, whose EC values reach peaks above 50 dS/m as well as an important monthly and daily variability provides an alert. Through punctual data withdrawals and inspections, the origin of such anomalies could be understood.

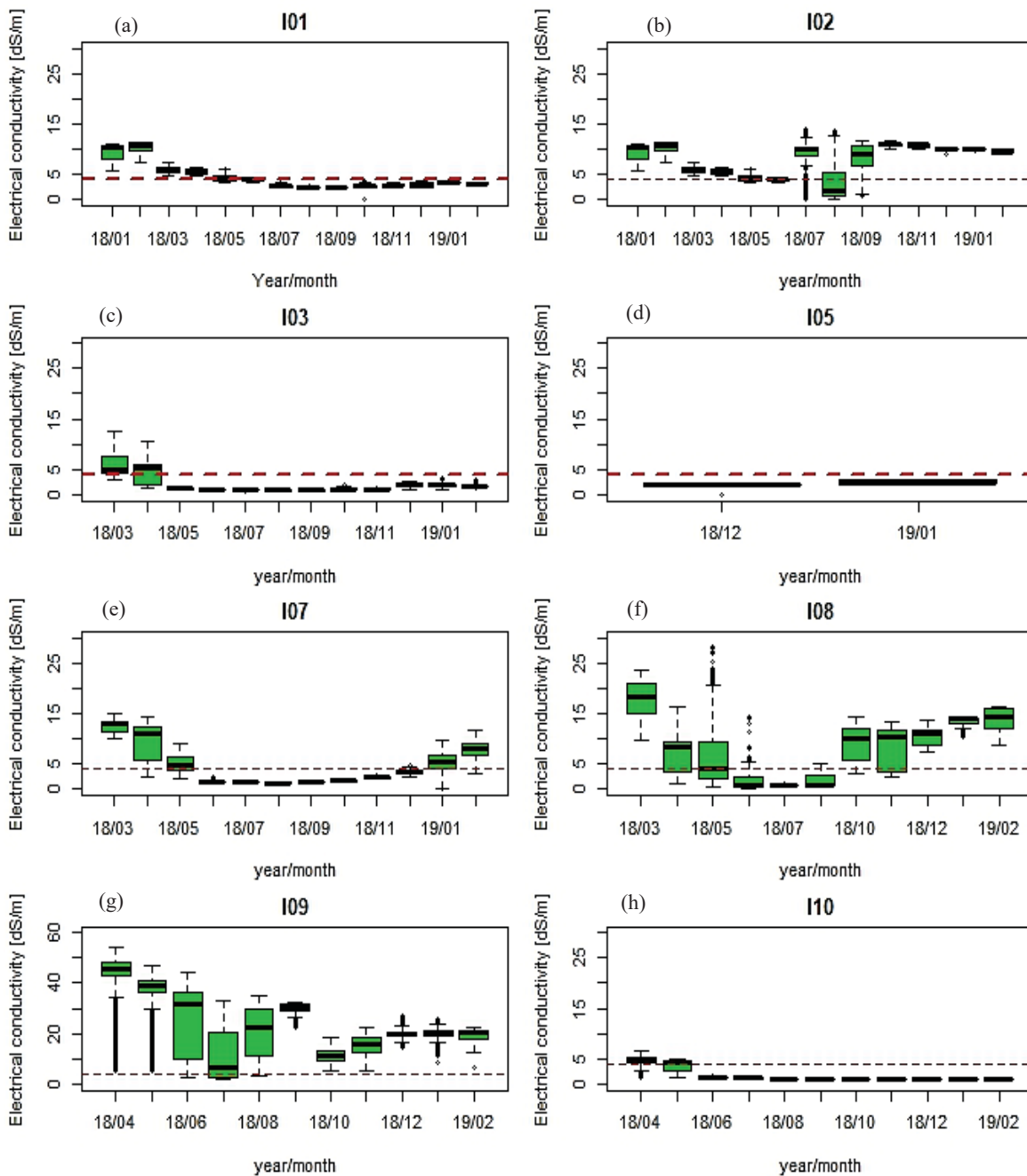


Fig. 7. Monthly boxplots of EC values in nodes I01 (a), I02 (b), I03 (c), I05 (d), I07 (e), I08 (f), I09 (g), and I10 (h) during the monitoring period

3.3. Water level and salinity in P-node

Groundwater table has been monitored in terms of depth from the ground level and EC by 8 P-node and 8 piezometers (see Fig. 3 for their positions). Fig. 8 shows the monthly box plots of the level and EC values for 4 of the 8 monitored piezometers. Piezometers show a marked seasonality in the watertable depth pattern and a low monthly variability of EC values since sensors have a fixed position inside the piezometer.

Rising brackish groundwater level, as the case of almost all the monitored piezometers (the piezometer P01 is in fact close to the Reno river), is a

major indicator of the risk of salinity. Once the watertable rises to within 2 meters of the soil surface there is large risk of soil salinization. The fixed depth of installation of the sensors greatly affects the measurement of EC so it must be selected with due attention. Table 3 sums up the monthly mean values of the depth, temperature, and electrical conductivity of water. The red line of each graphs shows the sensor position and the brown one the ground level. In conclusion all groundwater monitored are strongly saline. Lowering the watertable is the first step to effectively reclaim a saline site, and this the motivation that, during the monitoring period, has pushed farmers to install agricultural drains.

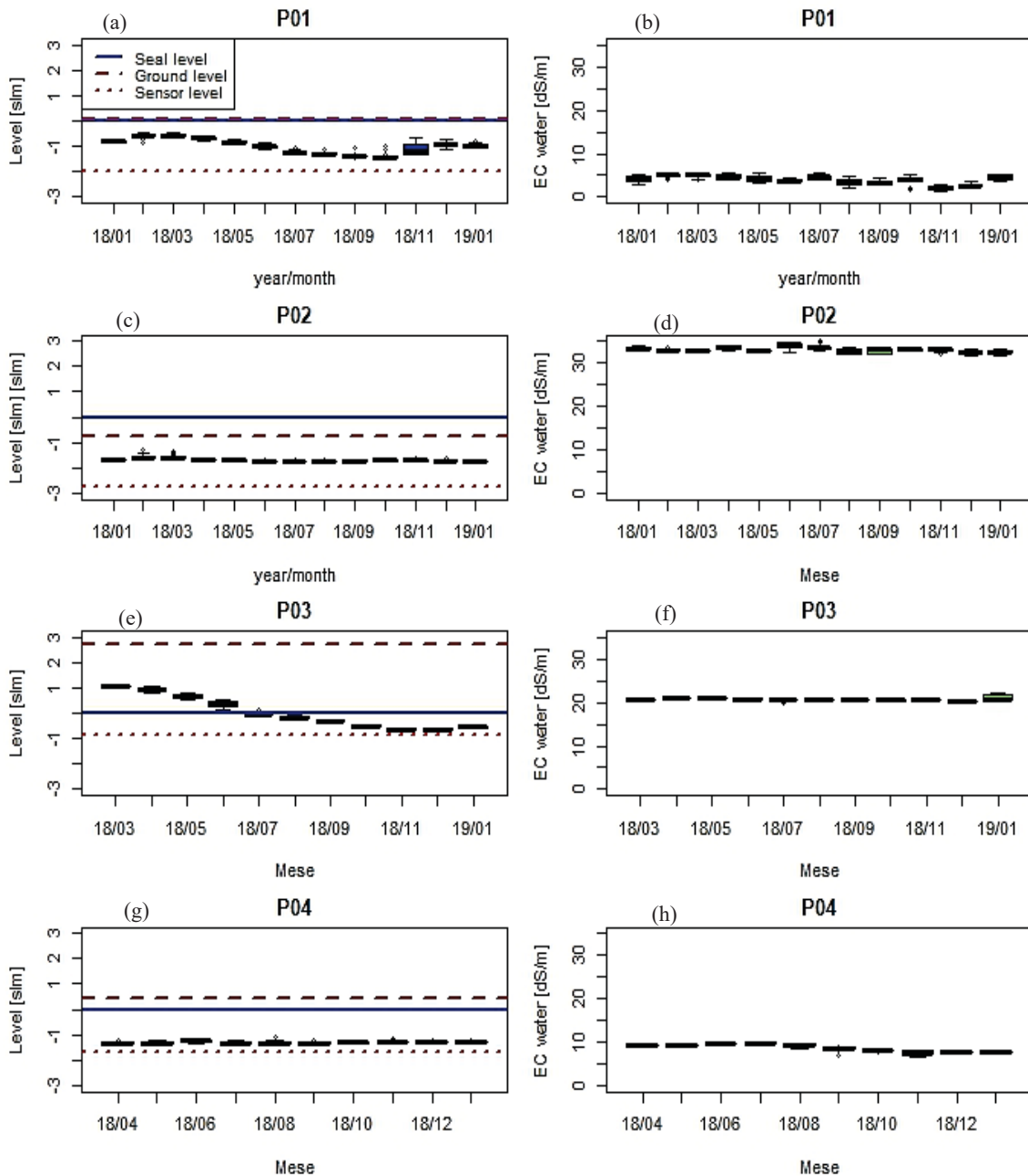


Fig. 8. Monthly boxplots of water depth (left) and EC (right) values in P01 (a and b), P02 (b and c), P03 (e and f), and P04 (g and h). The brown line of each figure represents the ground level, the red line is the level in which the sensor has been installed, and the blue line corresponds to the sea level

Table 6. Average monthly EC, water level, and water temperature values and sensor altitude for each P-node

ID	Sensor	01/18	02/18	03/18	04/18	05/18	06/18	07/18	08/18	09/18	10/18	11/18	12/18	01/19
P01	CTD10 Ew mS/cm	4.2	5.0	5.0	4.7	4.2	3.8	4.6	3.5	3.4	3.9	1.9	2.7	4.5
	Water Table [slm]	-0.8	-0.6	-0.6	-0.7	-0.8	-1.0	-1.2	-1.3	-1.4	-1.4	-1.1	-0.9	-1.0
	CTD10 Tw $^{\circ}$ C	12.8	11.8	10.9	12.0	14.2	16.3	19.1	20.7	21.2	20.2	18.2	14.9	13.1
	Level [slm]	-1.95	-1.95	-1.95	-1.95	-1.95	-1.95	-1.95	-1.95	-1.95	-1.95	-1.95	-1.95	-1.95
P02	CTD10 Ew mS/cm	33.3	32.9	32.8	33.4	32.7	34.0	33.5	32.5	32.8	33.1	32.9	32.5	32.5
	Water Table [slm]	-1.7	-1.6	-1.6	-1.7	-1.7	-1.7	-1.7	-1.7	-1.7	-1.7	-1.7	-1.7	-1.7
	CTD10 Tw $^{\circ}$ C	12.6	11.7	11.0	11.9	13.4	15.4	17.3	19.1	20.0	19.6	17.9	15.4	13.3
	Level [slm]	-2.75	-2.75	-2.75	-2.75	-2.75	-2.75	-2.75	-2.75	-2.75	-2.75	-2.75	-2.75	-2.75
P03	CTD10 Ew mS/cm	NA	NA	20.7	20.9	21.0	20.8	20.6	20.6	20.6	20.5	20.5	20.3	21.0
	Water Table [slm]	NA	NA	1.1	0.9	0.7	0.3	0.0	-0.2	-0.3	-0.5	-0.6	-0.6	-0.6
	CTD10 Tw $^{\circ}$ C	NA	NA	12.8	12.6	13.1	14.0	15.1	15.9	16.8	17.3	17.3	16.9	16.1
	Level [slm]	NA	NA	-0.83	-0.83	-0.83	-0.83	-0.83	-0.83	-0.83	-0.83	-0.83	-0.83	-0.83
P04	CTD10 Ew mS/cm	NA	NA	NA	9.3	9.3	9.5	9.8	9.2	8.5	8.1	7.5	7.6	7.8
	Water Table [slm]	NA	NA	NA	-1.3	-1.3	-1.2	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3	-1.3
	CTD10 Tw $^{\circ}$ C	NA	NA	NA	12.2	14.3	15.8	19.4	20.7	21.1	20.2	18.7	16.3	14.1
	Level [slm]	NA	NA	NA	-1.66	-1.66	-1.66	-1.66	-1.66	-1.66	-1.66	-1.66	-1.66	-1.66
P06	CTD10 Ew mS/cm	31.3	30.5	32.2	31.3	30.5	30.8	29.9	26.5	20.1	19.2	19.1	13.8	11.9
	Water Table [slm]	0.13	0.37	0.35	0.12	0.04		-0.23	-0.62	-0.51	-0.35	-0.33	-0.23	-0.16
	CTD10 Tw $^{\circ}$ C	14.1	13.2	12.5	12.3	12.6	14.8	16.7	19.1	19.8	19.0	17.5	15.1	12.3
	Level [slm]	-2.34	-2.34	-2.34	-2.34	-2.34	-2.34	-2.34	-1.81	-1.81	-1.81	-1.81	-1.81	-1.81
P07	CTD10 Ew mS/cm	NA	36.2	36.1	36.3	36.5	36.5	29.9	16.2	17.6	17.9	18.3	18.6	18.2
	Water Table [slm]	NA	-0.9	-0.9	-1.2	-1.2	0.0	-1.3	-1.6	-1.5	-1.5	-1.5	-1.5	-1.4
	CTD10 Tw $^{\circ}$ C	NA	14.0	12.8	12.4	13.1	14.1	15.9	18.1	18.9	18.7	17.8	16.3	14.0
	Level [slm]	-2.72	-2.72	-2.72	-2.72	-2.72	-2.72	-2.72	-1.85	-1.85	-1.85	-1.85	-1.85	-1.85
P08	CTD10 Ew mS/cm	NA	NA	1.90	4.10	4.10		6.70	9.90	6.30	9.40	12.00	10.30	3.90
	Water Table [slm]			-0.48	-0.89	-0.96		-1.03	-1.19	-1.46	-1.43	-1.36	-1.18	-1.02
	CTD10 Tw $^{\circ}$ C			10.10	11.30	13.80		16.20	18.00	19.50	20.00	19.30	17.90	15.00
	Level [slm]	-1.82	-1.82	-1.82	-1.82	-1.82	-1.82	-1.82	-1.82	-1.82	-1.82	-1.82	-1.82	-1.82

3.3. Moisture and salinity in S-node

S-nodes allow estimating the moisture content (U_s) and measuring the bulk conductivity (EC_b). The pore water EC (EC_w) has been then estimated, as a function of the previous illustrated parameters, based on an empirical equation provided by the company that made the sensors. EC_w provides information about the soil solution, and then of the water that the plant roots actually experience during the transpiration process. Salinity sensors may be used for continuously monitoring electrical conductivity of soil water at selected depths over relatively long periods of time, as illustrated in Table 7. Soil moisture content generally

decreases during the summer period and in fact all the sensors show this trend. An exception is represented by S03 sensor, which is located in the middle of an irrigated field and moreover it is close to an artificial wetland.

As the soil moisture decreases, the concentration of the salts is increased, causing an increase in the EC_w , and this causes a poor crop yield. During the monitoring period the field located near the P02 was cultivated with sunflower, and the low yields achieved are certainly attributable to elevated EC_w measured. On the contrary, the sorghum cultivated near the P08 has obtained a good yield demonstrating to better tolerate the high values of EC_w .

Table 7. Average monthly moisture content (US), bulk conductivity (EC_b), pore water conductivity (EC_w) and relative statistics for some S-node

ID	S01			S03			P02			P08		
	GS3_E C _b	GS3_E C _w	GS3_ U _s	GS3_ EC _b	GS3_E C _w	GS3_ U _s	GS3_ EC _b	GS3_E C _w	GS3_ U _s	GS3_E C _b	GS3_E C _w	GS3_ U _s
SU	mS/cm	mS/cm	%	mS/cm	mS/cm	%	mS/cm	mS/cm	%	mS/cm	mS/cm	%
01/18	-		-	-		-	1.460	6.421	46.060	-	-	-
02/18	-		-	-		-	1.630	6.425	48.010	-	-	-
03/18	0.320	2.301	36.840	-		-	1.410	5.611	47.840	0.630	4.151	38.580
04/18	0.330	2.321	37.220	0.860	3.326	47.980	1.240	4.886	47.640	0.660	4.505	37.550
05/18	0.380	2.825	35.840	0.930	3.160	49.790	1.380	4.570	50.110	0.730	4.449	39.130
06/18	0.340	4.614	26.710	1.010	3.073	51.210	1.370	4.790	48.880	0.810	5.148	38.110
07/18	0.220	5.033	21.090	1.160	3.230	52.330	0.840	11.456	26.170	0.570	8.160	25.640
08/18	0.310	4.673	25.480	1.030	3.153	50.780	0.710	10.371	25.080	0.650	9.073	25.830
09/18	0.220	4.858	21.140	0.980	3.236	49.660	0.460	7.091	25.150	0.720	7.960	29.130
10/18	0.200	4.819	20.850	0.900	3.949	45.180	0.110	1.561	26.170	0.630	7.163	28.820
11/18	0.250	4.686	23.330	1.100	3.870	49.370	0.670	4.928	32.880	0.690	6.656	31.730
12/18	0.320	3.172	31.680	1.110	4.293	48.340	1.390	6.304	45.320	0.650	5.095	35.410
01/19	0.370	3.060	33.780	1.080	4.368	47.540	1.350	6.444	44.010	0.590	4.773	35.250
Average	0.296	3.851	28.542	1.016	3.566	49.218	1.078	6.220	39.486	0.666	6.103	33.198
Max	0.380	5.033	37.220	1.160	4.368	52.330	1.630	11.456	50.110	0.810	9.073	39.130
Min	0.200	2.301	20.850	0.860	3.073	45.180	0.110	1.561	25.080	0.570	4.151	25.640
Var	0.004	1.219	44.409	0.010	0.252	4.229	0.218	6.320	109.925	0.005	3.072	26.561

4. Conclusions

This study shows the results of 13 months of monitoring activity realized by means of a wireless sensor network in an area affected by water and soil salinization. The WSN system is equipped with *M-nodes* to monitor the weather parameters, *S-node* to monitor moisture and electrical conductivity of soils; and *P-node* and *I-node* to monitor the water table and the electrical conductivity of groundwater and surface water respectively.

The network, currently set up with a 10-minute acquisition time step, is able to provide a wide range of data through which irrigation can be optimized. Furthermore *I-nodes* may allow optimizing the management of both irrigation and drainage systems by reducing for example the amount of fresh water get into the system to reduce the EC in canals with irrigation functions, or by optimizing the operation of pumping systems during wet weather.

Overall the network worked without major concerns, except for P05 node in which cables have been cut out by a farmer during plowing, and for I08 node that had a problem of data transmission caused by vegetation growth. In conclusion, the network as a whole turns out to be an excellent tool to support the precision farming, however during the installation of the sensors it would be advisable to take the following precautions:

- 1) The high variability of precipitation, in particular during the irrigation season, suggests the need of installing an adequate number of rain-gauges;
- 2) The sensors located in canals should always be covered by a minimum water depth, and water stagnation should be avoided.
- 3) Water density rises proportionally to salt content. In the piezometer water column, there is often a clear interface between the fresh and the salt water. The depth of this interface depends on the volume of fresh water in the piezometer, which in turn depends on rainfall and irrigation. However, it often happens that the probes placed at lower depth measure highest EC values. For this reason, the continuous measurement of EC at a given fixed level must be integrated with measurements along the water column to evaluate the salinity gradient.

In the near future, in situ measurement through the WSN must be integrated with satellite data (e.g. rainfall, soil moisture, NDVI etc). Those last family of measurements are frequently free of charge, and moreover, the resolution is continually improving in terms of both space and time. This will provide distributed information that will allow to extend the information acquired by a wireless sensor network system.

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START-UP OF THE DOOR-TO-DOOR MUNICIPAL SOLID WASTE SEPARATE COLLECTION SERVICE IN A LARGE METROPOLITAN AREA

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Abstract

The study describes the start-up phase of the door-to-door separate collection service of municipal solid waste (MSW) in a large metropolitan area, analysing not only the performance in terms of separately collected waste but also the feedback from citizens on how to improve it. For the scope, the case study of the city of Bari (Southern Italy) was considered. The methodological approach involved primarily the subdivision of the entire municipality into eight homogeneous territorial zones (HTZ) considering population density and space availability. Additionally, each HTZ was decomposed into unitary areas, which in turn were classified according to the degree of feasibility in implementing door-to-door separate collection. During the first year of operation, results showed excellent performance in terms of separately collected waste (>80%) highlighting the goodness of the adopted technical approach as well as the convenience in acquiring feedback from users during the start-up of the service. While expressing positive satisfaction about the door-to-door system, users consider the adopted sanctioning and control system to be critical. The same was considered insufficient to deal with the well-known phenomenon of “waste tourism”.

Keywords: citizen involvement, door-to-door collection, municipal solid waste, service performance, start-up

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1. Introduction

In Europe today separate collection (SC) of each municipal solid waste (MSW) fraction is considered a prerequisite for promoting high quality recycling and high recycling rates. Article 11(1) of the Directive 2018/251 amending Directive 2008/98/EC on waste sets out the general obligation for SC by requiring Member States to set up SC schemes at least for paper, metals, plastics and glass, and by 1 January 2025, for textiles. Article 11(1) of the same Directive requires European Member States to take measures to promote high quality recycling through SC. Technical literature shows a wide variety of ways to collect

different MSW streams such as bring points, door-to-door, co-mingled door-to-door and civic amenity sites. In general, with the bring points system, citizens have to transport waste from the point of production to the point of collection. With the door-to-door system, it is the operator of the collection service who goes to the individual producer users to collect the separated waste (De Feo et al., 2012). Door-to-door collection ranges from one container to six separate containers/bags (including the container for residual waste) while co-mingled door-to-door involves the harvesting of some fractions in a single container such as metal and plastic. Civic amenity sites are used as additional collection schemes, usually accepting the

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same fractions collected in transport containers. For some countries such as Czech Republic, civic amenity sites are the primary collection system for metals and bio-waste. The EU report “Assessment of separate collection schemes in the 28 capitals of the EU” (EC Directive, 2015) showed that practical implementation of the Waste Framework Directive obligations differs significantly across 28 EU Member States. In complex cities such as those in Table 1 it is possible to have multiple waste collection systems simultaneously. In all, 25 cities operate a door-to-door separate collection system, 9 cities collecting each fraction in a separate bin and 16 cities including co-mingled bins in their door-to-door collection infrastructure. Regarding the yield of the separate collected materials, on average, only 19.7% of generated municipal waste is collected separately in EU-28 capitals; this means that 80.3% of the waste still ends up in the residual waste bin.

Literature focusing on different aspects of waste collection clearly agree on the advantages of SC, even if opinions regarding the optimal design of collection systems differ. De Feo and De Gisi (2010) stressed the importance of the technical infrastructure of the collection system and how important it is to inform and motivate users of the service. Bertanza et al. (2018) pointed out how the percentage of recyclables as well as their quality increases with the door-to-door; the collection costs are higher than alternatives although collection rates and consequently revenues are usually higher with a

consequent reduction in waste rates and treatment costs. Giacetti et al. (2009) highlighted that the bring points system encourages inhabitants to produce waste with a higher percentage of impurities. However, it was a reasonable solution for some fractions such as the glass.

Co-mingled collection of recyclables (e.g. plastics and metals) is a widespread practice that tends to reduce costs; however, mixing multiple streams can result in a higher incidence of cross contamination, and the quality of recyclables tends to be lower and rejection rates higher. Furthermore, Giacetti et al. (2009) highlighted how a system based on the SC of organics generates an increase in the dry collection fraction. With reference to the case study of a Greek city (Xanthi, Thrace) undergoing a change in its waste collection system from the existing kerbside to a door-to-door SC system, Tsalis et al. (2018) showed how the most of the respondents were willing to participate in a future door-to-door recyclables collection programme; the factors that influenced the respondents’ attitude with regard to such a programme were associated with level of education, their beliefs about the effectiveness of the current recycling system and also their attitudes towards recycling issues in general. Age and religion significantly affected recycling frequency. Haupt et al. (2018) pointed out how a higher percentage of waste collection did not always imply greater economic and environmental benefits.

Table 1. MSW separate collection schemes for 28 EU-Capitals (EC Directive, 2008)

City	Applied collection schemes				% separate collection considering all systems
	Door-to-door separate 	Door-to-door co-mingled 	Bring points 	Civic amenity sites 	
Amsterdam	x		x	x	12.4
Athens	x	x	x		16.1
Berlin	x	x	x	x	27.4
Bratislava			x	x	14.2
Bucharest			x		2.9
Budapest	x	x	x		7.6
Brussels	x	x	x	x	20.9
Copenhagen	x		x	x	23.7
Dublin	x	x	x		36.6
Helsinki	x		x	x	38.6
Lisbon	x		x	x	11.5
Ljubljana	x	x	x	x	55.4
London	x	x	x	x	25.4
Luxemburg	x	x	x	x	28.4
Madrid	x	x	x	x	11.6
Nicosia	x	x	x	x	6.1
Paris	x	x	x	x	11.6
Prague			x	x	14.3
Riga	x		x	x	18.3
Rome	x	x	x	x	16.3
Sofia	x	x	x	x	4.0
Stockholm	x		x	x	21.5
Tallinn	x	x	x	x	47.2
Valetta	x	x	x	x	7.9
Vienna	x		x	x	29.2
Vilnius	x		x	x	5.5
Warsaw	x	x			4.5
Zagreb	x		x	x	1.0
Average		-			19.7

The optimal value of the collection rate was site-specific and would be determined by both economic and environmental analysis (e.g., based on the Life Cycle Assessment), as reported in De Feo and Malvano (2012). The literature review showed limited information on the methodological approaches to be adopted to monitor the effectiveness of a waste collection service. The latter must also take into account both the technical and social component of the issue addressed. Referring only to the case of a collection system already in operation, Calabrò and Komilis (2019) proposed a semi-qualitative inspection method to evaluate both the door-to-door SC system and the conventional curbside system. The method was based on the combined evaluation of waste collection using a set of indicators and the assessment of the perception of the citizens towards collection and street cleaning services using behavioural questionnaires.

The present paper describes the start-up phase of the door-to-door collection service of municipal solid waste (MSW) in a large metropolitan area, analysing not only the performance in terms of separately collected waste but also the feedback from citizens on how to improve it. The intent was to provide new methodological insights to be applied in similar territorial contexts.

2. Methodological approach

2.1. Experimental plan

The MSW door-to-door SC service was designed and started up in June 2017 in the municipality of Bari (Apulia Region), third city in Southern Italy after Naples and Palermo, characterized by a population in 2017 of 316,656 inhabitants.

The pre-existing collection system described below had lower performances than the Italian Law (LD, 2006), which sets a limit value of 65%. Since the project has to switch to door-to-door, it envisaged the adoption of an innovative methodological approach specially defined by CONAI (Consorzio Nazionale Imballaggi) based on the following main phases: (i) identification of the start-up areas, (ii) start of service for each start-up area and (iii) contextual survey of the start-up area citizen's public perception with the goal of highlighting and resolving any critical issues. The main elements of this developed methodological approach were described below.

2.2. Inlet waste characterization and present mode of waste collection

The inlet MSW consists of compostable material (40 %), recyclable (51.5 %), WEEE (Waste Electrical & Electronic Equipment) and bulky items (2%), sanitary textile such as diapers (1.3 %), hazardous material such as expired pharmaceuticals

(0.2%) and collection residue (5 %).

The major component of the compostable (and in absolute terms) was the organic fraction from food waste (34 %). The main components for recyclables were paper and cardboard (25 %) and plastic (12 %). Such a product composition was in line with that reported in ISPRA (2017) with reference to the period 2008-2017 (Table 2). It was possible to observe how the percentage of paper and cardboard was higher than the Italian average value as already highlighted by De Feo et al. (2017).

Bari has always been a positive anomaly in this regard. The pre-existing MSW collection system was "bring-points" type based on public and collective containers (bins, or other types of containers) where users can deliver the waste (Bertanza et al., 2018). The collection points consisted of a fixed container for undifferentiated residue (mixed waste), a container with wheels for multi-material, a container with wheels for paper, a green bell for glass, a container for used clothing and brown wheeled bins with a lock for organics.

2.3. Methodology for the identification of the door-to-door start-up areas

The design of the door-to-door system involved an in-depth analysis of the territory, its urban and socio-demographic characteristics. First of all, homogeneous areas were identified from the urbanistic point of view on the basis of the analysis of the following variables: population density, type of dwellings, availability of condominium space, availability of space on the sidewalks. Called "Homogeneous Territorial Zones" (HTZ), these areas were delimited by physical, urbanistic or administrative boundaries. The HTZs were then hierarchized according to their vocation for door-to-door collection, starting from the principle that this type of collection can be carried out anywhere but with increasing constraints and penalizing factors depending on the urban context.

Therefore, the HTZs were classified into the following six classes (and colours) (UNI EN, 2017): (i) vocation areas (green); (ii) areas with a predominant vocation (yellow); (iii) areas with penalizing factors (orange); (iv) areas with a poor vocation (red); (v) mixed areas (blue); (vi) agricultural, artisanal and industrial area with low residency level (grey). This led to the creation of a map of the areas with a vocation for door-to-door SC for the municipality of Bari (Fig. 1). Instead, the number of households and inhabitants of each HTZ is shown in Table 3.

It was observed that, excluding the areas classified red and orange (which represent 39.45% of the population with 124,910 inhabitants), the rest of the territory could quickly move to a door-to-door SC, involving 60.55% of the population for a total of 191,756 inhabitants.

Table 2. Characteristics of the investigated inlet MSW

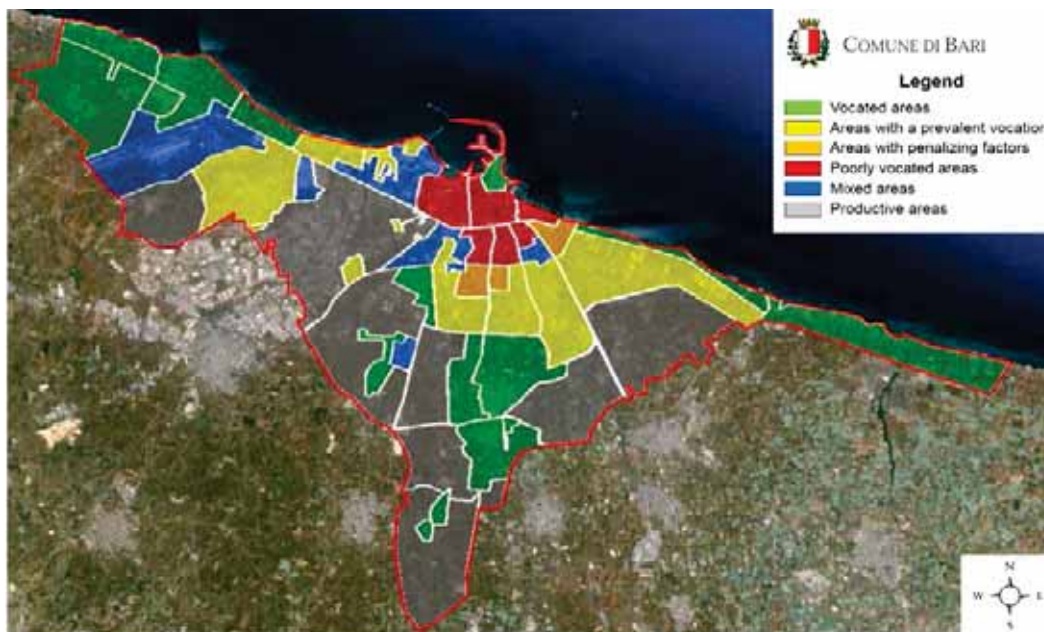
Composition		Unit	Value	
			Our study	Italy (ISPRA, 2017)
Compostable	Organics	% w/w	34.0	35.7
	Cellulosic material	% w/w	6.0	-
Recyclable	Paper and cardboard	% w/w	25.0	22.6
	Plastic	% w/w	12.0	12.8
	Glass	% w/w	7.0	7.6
	Metals	% w/w	2.0	2.6
	Wood	% w/w	2.0	3.0
	Clothing and textiles	% w/w	3.5	3.6
Other	WEEE and bulky items	% w/w	2.0	0.9
	Sanitary textile (diapers)	% w/w	1.3	3.5
	Hazardous	% w/w	0.2	0.3
Residue	Undifferentiated fraction or mixed	% w/w	5.0	7.4

The most suitable areas to switch to door-to-door in the short term were all those with green, yellow, blue and grey colour. Areas with red and orange colours were included among those that had to pass to the door-to-door. Unlike the other areas, these areas would have required “preparatory” mechanisms with the goal of encouraging the separation of waste at family level.

In this regard, while the transition to door-to-door would take place in the most suitable areas, users

of disadvantaged areas (with red and orange colours) would be involved in continuous and widespread communication campaign aimed at increasing the knowledge and awareness of citizens for the proper separation and delivery of waste.

Subsequently, starting from the map in Fig. 1, the so-called “Start-up Zones” of the MSW door-to-door SC service were identified (see paragraph 3.1), representing the areas of the municipality of Bari interested in the launch of the service.

**Fig. 1.** Homogeneous Territorial Zones (HTZ) of the municipality of Bari relating to the vocation to the door-to-door SC scheme**Table 3.** Number of households and inhabitants for each HTZ

Homogeneous Territorial Zones	Colour	Household users	Population	
			Inhabitants	%
Vocated areas	Green	29,351	77,632	24.52
Areas with a prevalent vocation	Yellow	42,359	113,143	35.73
Areas with penalizing factors	Orange	11,065	27,123	8.56
Poorly vocated areas	Red	41,423	97,787	30.88
Mixed areas	Blue	110	288	0.09
Agricultural, artisanal and industrial area with low residency level (productive areas)	Grey	256	683	0.22
Total	-	124,564	316,656	100.00

2.4. Description of the the door-to-door SC service

The door-to-door system was based on teams of vehicles operating in synergy. Different types of satellite vehicles were provided as visible from Table 4. Lower-capacity vehicles are used in areas with small-sized roads and as such are not directly accessible by larger vehicles. These satellite vehicles only collect the waste and then unload their contents into a larger compacting vehicle; the latter transports the waste to the municipal collection center. Depending on the number and qualification of the operator, 5 teams have been identified, the description of which is given in Table 4. Among the main design parameters of a collection system, a time of 300 min per turn was set for intermediate collection and discharge, plus a time of 60 min for movement between the operational site and the collection area.

For household's users (a total of 124,564), the

door-to-door system also provided for the automatic detection of deliveries by means of RFID tags for all fractions. The collected fractions were as follow: (i) paper and cardboard; (ii) light multi-material (plastic, aluminium and ferrous materials packaging); (iii) glass packaging; (iv) organics; (v) dry residual fraction not differentiable; (vi) sanitary textiles (diapers, only to the users who have requested the service). The collection of used textiles and clothing was unchanged (bring-points) although the availability of specific road containers (anti-intrusion towers), already widespread on the territory, was increased. The selection and attribution of bins to households was based on the average per capita waste productivity and, in the case of apartment buildings, on the number of users per civic number. The colors and characteristics of the bins identified for the various collections were in accordance with the UNI 11686:2017 standards (Table 5).

Table 4. Vehicles and operators used for the door-to-door SC

Typology of team	Team description	Vehicle description	Typology of vehicle
Team A	1 Operator/vehicle driver	Truck with 2.5-3 m ³ tank collection equipment	Satellite
Team B	1 Operator/ vehicle driver + 1 Operator collecting	Truck with simple 5 m ³ tank collection equipment	Satellite
Team C	1 Operator/ vehicle driver + 1 Operator collecting	Trucks with 7 m ³ tank capacity with compactor	Satellite
Team D	1 Vehicle driver + 1 Operator collecting	Truck with double tank collection equipment, with simple tank of 2.5-3 m ³ and compacting tank of 7-8 m ³	Satellite
Team E	1 Vehicle driver + 2 Operators collecting	16/18 m ³ compactor on two axis frame	Main vehicle

Table 5. Collection equipment for household and non-household utilities (Fr = collection frequency)

Collected material	Household			Non household	
	Fr.	Single utility	Multiple utility	Fr.	Utility
Paper and cardboard (joint collection)	1/7	 40 litres	 40 + 360 litres	1/7	 40, 120, 1100 litres
Cardboard (selective collection)	-	-	-	3/7	 in bulk or roll containers
Light multi-material (plastic, steel, tinsplate and aluminium packaging)	1/7	 PE 100 litre bag with alphanumeric code	 PE bag 100, 360 litres	3/7	 80-100 litre bag or 360 litre wheeled bin
Glass	1/14		 40, 240 litres	3/7	 40, 120 litres

Organics (food waste)	3/7	 biodegradable bags, 10 litre holed bin, 20-24 litre closed bin	 biodegradable bags, 10 litre holed bin, 240 litre closed bin	6/7	 120, 240 litres
Undifferentiated waste (also called residue or mixed waste)	2/7	 40 litres	 40, 360 litres	2/7	 40, 120 and 240 litres
Sanitary textiles (diapers)	4/7	 20, 24 litres on request	 20, 24 litres on request	-	-

Single users, i.e. those located in buildings with only one dwelling or up to 8 units per building (for a total of 43,906 single users, 32% of the total), delivered the purpose-separated waste directly on the road, through the relative bin or bag, on days and times fixed by the municipal administration. Apartment buildings users, i.e. those located in buildings with more than 8 residential units (for a total of 93,359 condominium users, 68% of the total), were equipped with kits similar to those of single users, with the exception of the 20-24 liters bin for the organics and without RFID tags. Moreover, apartment buildings users did not need the endowment of self-adhesive labels with a unique identification code for the bags of the multi-material. These users delivered their separate waste to the condominium containers (wheeled bins) located in a private area, with no restrictions on days or hours. Kits distributed for single and multiple domestic users were shown in Table 5. Non-domestic users (a total of 18,905) were treated in the same way as domestic ones. For some wastes, collection occurred simultaneously with that of household users (paper, mixed waste); for others, a specific service was provided with different calendars and frequencies. For non-domestic users, containers equipped with RFID tags were distributed.

Table 5 also showed the frequency of collection. Paper collection was carried out simultaneously for household and non-domestic users. On the other hand, cardboard was collected only for non-domestic users. The frequency of collection of light multi-material was 1/7 and 3/7 for households and non-domestic users, respectively. The remaining frequencies for each fraction and user were shown in Table 5. An element of excellence, automatic waste detection with RFID tags allowed to identify (spatially and temporally) and quantify the single fractions delivered by users.

2.5. Methodology for calculating the percentage of separately collected waste

The calculation of the percentage of separate collection (SC%) of a given area (a zone or the whole

city) was carried out according to the ISPRA-ONR method (ISPRA, 2010), by means of the Eq. (1):

$$SC(\%) = \frac{\sum_i SC_i}{(\sum_i SC_i + UW_{undiff} + B + S_{SC})} \times 100 \quad (1)$$

where:

$\sum_i SC_i$ is the sum of the different collected materials constituting the separate collection, excluding any residues (Table 5);

UW_{undiff} is the sum of the amount of residue (“unsorted municipal waste”, 20.03.01 ERC code) and waste from street cleaning (“waste from street cleaning”, 20.03.03 ERC code);

B is the amount of bulky waste for disposal (“bulky waste”, 20.03.07 ERC code);

S_{SC} is the amount of scraps/residues from separately collected waste sorting facilities.

2.6. Survey to assess the user’s customer satisfaction

Questionnaires were administered to citizens involved in the door-to-door in order to assess the performance of the service and to highlight any critical issues. The questionnaire included a two-part format generally adopted in studies of this type (De Gisi et al., 2017). The first part referred to age, gender, educational qualifications and number of family members. The second part contained 13 questions concerning the public perception of users of the new service of which, the first 12 multiple-responses and the last with free response (Table 6).

The questionnaire was produced in print and digital format. The print format was distributed at the start-up offices where citizens went to collect the annual supply of waste bags. Instead, the digital format was spread online on several websites and social networks supported by the Municipality of Bari, citizens ‘committees, environmentalists, etc.

In total, 305 questionnaires were administered, most of them in digital format (~89%).

Table 6. The submitted questionnaire (English translation and adaptation)

<i>Social aspect</i>	<i>No.</i>	<i>Question</i>	<i>Answers</i>
Personal attributes	-	Age	18-30; 31-40; 41-50; 51-60; 61-70; >70
		Sex	Male, female
		Family members	1, 2, 3, 4, 5, >6
		What is your education level?	First level (primary); Second level (secondary); Third level (high); Fourth level or more (degree, Ph.D.)
		What is your neighbourhood?	Sub-zone 1: S. Spirito; Sub-zone 2: Palese; Sub-zone 3: San Pio, Catino, Palese centro, Palese Macchie; Sub-zone 4: Marconi, San Girolamo, Fesca.
Behaviour	Q1	Why do you think SC is useful?	For environmental protection; for economic savings; other.
	Q2	If you throw away a waste you do not know the classification of, what do you do?	You find out where to dump it; You throw it into the undifferentiated; Other.
	Q3	Have you any difficulty sorting your waste at home?	Yes, the containers take up too much space; Yes, I have doubts about how to sort some waste; Yes, I waste too much time in separating; No; Other.
	Q4	With which type of waste do you have the most difficulty in separating?	Organics; Paper and cardboard; Plastic; Metals; Glass; Used cooking oils; Anybody.
	Q5	What types of waste do you usually separate from the undifferentiated waste?	Organics; Paper and cardboard; Plastic; Metals; Glass; Used cooking oils.
	Q6	Do you think that the current collection frequency for each type of waste is satisfactory?	Yes; No, I believe that the frequency of the organics must be increased; No, I believe that the frequency of plastic and metals must be increased; No, I believe that the frequency of undifferentiated must be increased; No, I believe that the frequency of paper and cardboard must be increased.
Opinion	Q7	Collection methods shown on the information material are clearly exposed?	Yes; No.
	Q8	Do you find the supplied bins suitable for SC?	Yes; No.
	Q9	Do you think that the number of SC bags provided is sufficient for your household's consumption?	Yes; No, I believe that the number of bags for organics should be increased; No, I believe that plastic and metal bags should be increased.
Customer satisfaction	Q10	Are you satisfied by the work done by the collection operators?	Yes; No.
	Q11	Are you satisfied by the work done by road cleaners?	Yes; No; Enough.
	Q12	How do you evaluate the overall door-to-door collection service?	Excellent; Good; Satisfactory; Scarce.
User feedback	Q13	How do you think the door-to-door collection service could be improved?	It is an open question with answers from the user.

3. Results and discussion

3.1. Identification of the door-to-door start-up zones

The map of the Start-up Zones for the city of Bari was shown in Fig. 2. These areas were mainly characterized by comparable populations. The start-up zones were separated by natural boundaries, demarcation barriers and in general “buffer zones”. At the time of their identification, the intention was to

contain the well-known phenomenon of “waste tourism”. As reported De Feo and De Gisi (2010), it represents the passage of (waste) flows between one area and another of the city and in the different phases of the start-up of the new collection system.

The territory of the Municipality of Bari was therefore divided into 8 start-up zones gradually and sequentially involved in the start of the service. The first area was called Start-up Zone 1 (Fig. 3), where the door-to-door SC service has begun in June 2017.

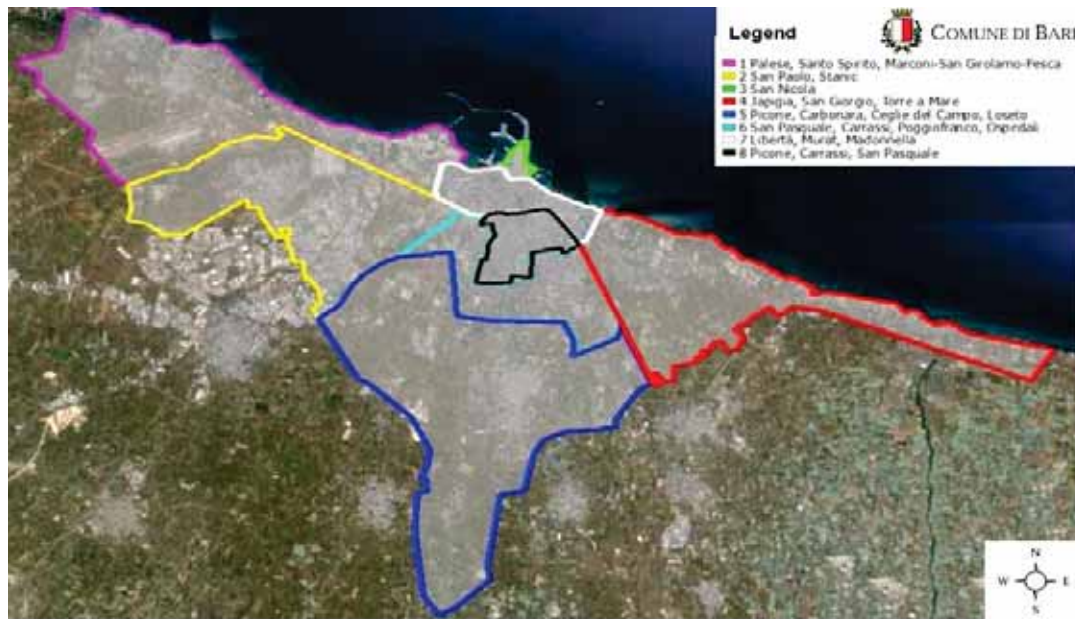


Fig. 2. Start-up zones of the municipality of Bari



Fig. 3. Details of the Start-up zone 1 of the door-to-door SC service

3.2. Service performance in start-up zone 1

The amount of sorted waste collected in the reference year (June 2017 – June 2018) in start-up zone 1 was 8,848 tonnes (Fig. 4a), corresponding to a percentage of separately collected waste of 82.6% (Fig. 4b). Separately collected waste showed a growing trend over time and then stabilised in the last 4 months both for total waste collected separately and for residue. The SC percentage was always higher than the minimum limit of 65% (LD, 2006) (Fig. 4b).

These values were significantly higher than those achieved in the whole municipality of Bari with the previous bring-points system, equal to 36.9% for

the year 2016 (http://ecologia.regione.puglia.it/portal/portale_orp). Mostly collected waste was organics, residue, paper and cardboard, multi-material, glass and lastly diapers (Fig. 4c). The reasons behind the excellent performance of the service were herein described.

3.3. Service evaluation by users

Most of the respondents were women aged between 30 and 50. 48% had a high school diploma and 33% a university degree or higher; therefore, the percentage of those who had a medium-high educational qualification was 81%.

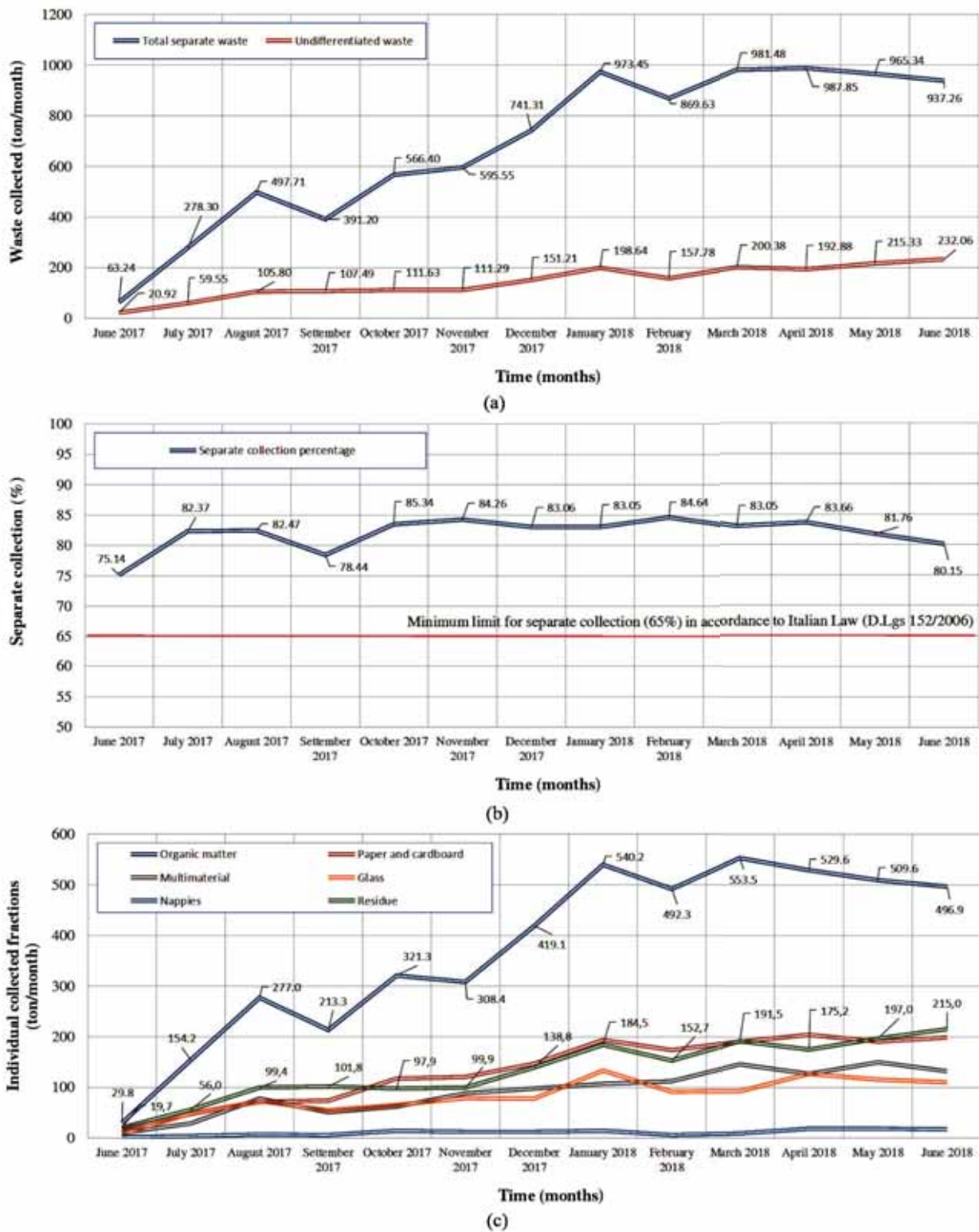


Fig. 4. Door-to-door SC performance for start-up zone 1 in terms of: (a) Amount of collected waste; (b) SC percentage; (c) Amount of collected individual fractions

This sample well represented the population of the start-up zone 1, made up mainly of young families. The typical household was composed of 3-4 members. The results of Fig. 5a allowed to outline the behaviour of the respondents as well as their approach to waste sorting. 92% of respondents were well aware of the importance of SC for the protection of the

environment rather than for economic savings. If they were to dispose of a waste of which they did not know its classification, they were informed of where to dispose of it (68%) (via the Internet, Facebook groups, Junker Apps and information material provided) rather than disposing of it in the undifferentiated bin (28%) (Fig. 5b).

Most of the users of the service did not find it difficult (72%) and a small part (14%) felt that a reason that makes separation complex is the lack of space in the house for the placement of containers (Fig. 5c). This confirmed the positive trend of SC in start-up zone 1 described in the previous paragraph.

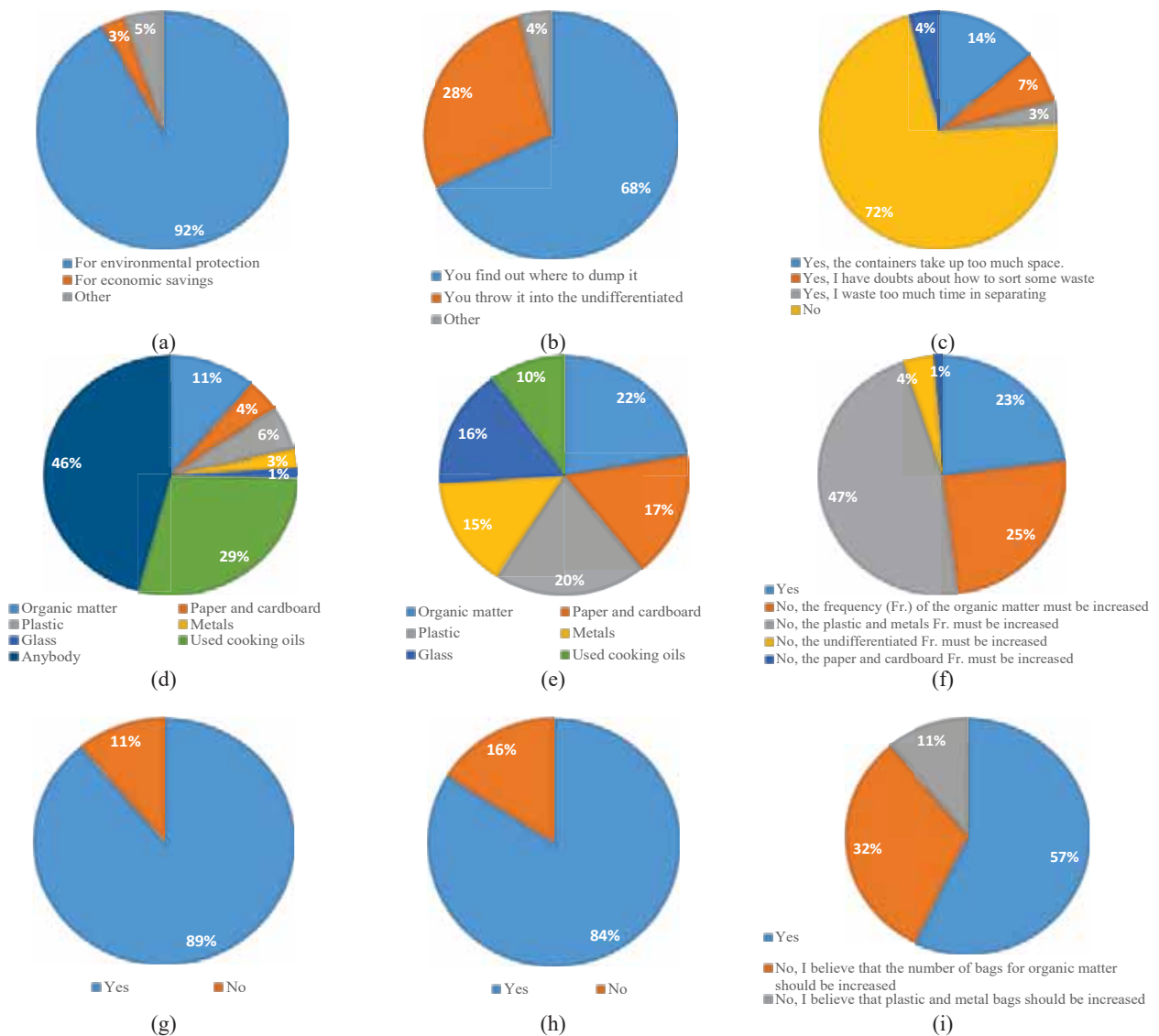
Further confirmation was given by the answers to the Q₄ question “With which type of waste do you have the most difficulty in separating?” where almost half of the respondents (46%) indicated that they had no difficulty with any fraction. On the other hand, the rest found it more difficult to differentiate used cooking oils (28.7%) and organics (11%) (Fig. 5d). Analysing the answers to question Q₅ “What types of waste do you usually separate from the undifferentiated waste?” it was observed that most of the respondents usually separate all the product fractions (Fig. 5e). Subsequently, it was investigated how users evaluated the SC service. The results showed that changes could be made to the collection calendar; for almost half of it (47%), the collection frequency for plastics and metals had to be increased

(Fig. 5f). It was interesting to note that only 4% wanted the frequency of collection of the undifferentiated to be increased. This suggested that the users were already educated to sort waste well.

With regard to the equipment supplied by the Municipality of Bari, a large number of users (89%) considered the collection instructions reported on the information material to be clear (Fig. 5g), and also found the containers supplied to be suitable (84%) (Fig. 5h). As far as bags are concerned, about 57% believed that they were sufficient for their own household, while 32% would have liked to have greater availability (Fig. 5i).

When users were asked to give their opinion on service satisfaction, they expressed a good rate for collection operators (42% answered Yes, 40% answered Enough) (Fig. 5j) and a very bad satisfaction rate for road cleaning operators (70%) (Fig. 5k).

The door-to-door collection service in start-up zone 1 was generally appreciated by users. Only 7% and 21% (in total 28%) of respondents considered the system scarce and satisfactory, respectively (Fig. 5l)



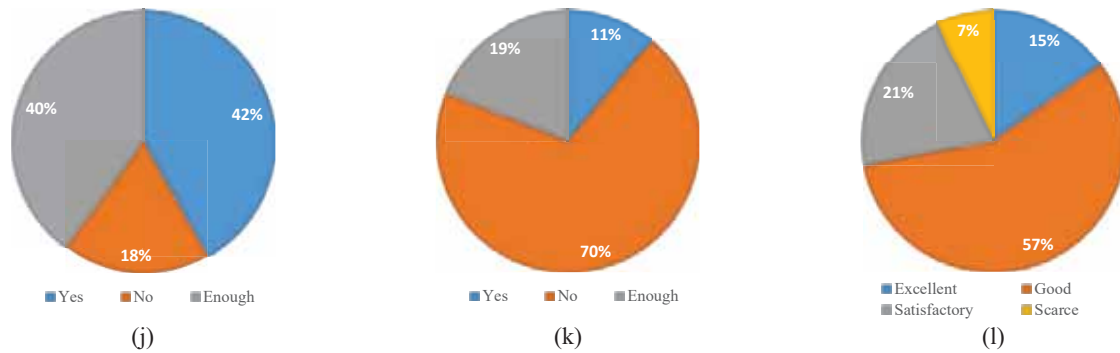


Fig. 5. Results of the sociological survey for the start-up zone 1: (a) Q₁ = Why do you think SC is useful? (b) Q₂ = If you throw away a waste you do not know the classification of, what do you do? (c) Q₃ = Have you any difficulty sorting your waste at home? (d) Q₄ = With which type of waste do you have the most difficulty in separating? (e) Q₅ = What types of waste do you usually separate from the undifferentiated waste? (f) Q₆ = Do you think that the current collection frequency for each type of waste is satisfactory? (g) Q₇ = Collection methods shown on the information material are clearly exposed? (h) Q₈ = Do you find the supplied bins suitable for SC? (i) Q₉ = Do you think that the number of SC bags provided is sufficient for your household's consumption? (j) Q₁₀ = Are you satisfied by the work done by the collection operators? (k) Q₁₁ = Are you satisfied by the work done by road cleaners? (l) Q₁₂ = How do you evaluate the overall door-to-door collection service?

The questionnaire for the evaluation of the door-to-door service along with user feedback were the main source for the identification of the criticalities of the system during the first year of operation. The results (Fig. 6) made it possible to identify the following main criticalities: (i) the need for higher frequency of collection for some waste (19.3%); (ii) the working methods of ecological operators (17.7%); (iii) the need to intensify controls and increase penalties (15.6%). In detail, 47% and 33% of the respondents would have liked the frequency of collection of plastics/metals and of organics to be increased, respectively. Such feedback was also in line with the answers to question Q₆. The second critical point was the bad work by ecological

operators; in particular, 37% of users considered the work of roads cleaners to be insufficient. Again, such feedback was confirmed by the answers to question Q₁₁.

The last critical point concerned the control and sanctioning system put in place by the Municipality of Bari. Users considered inadequate the implemented system of sanctions. This feedback highlighted a criticality already found in the first months of the service's start-up, that was the irregular abandonment of waste either in the countryside or in the bins of nearby neighbourhoods not yet reached by the door-to-door SC service. This was the most damaging aspect of the service, in line with findings already available in the literature (De Feo, 2014).

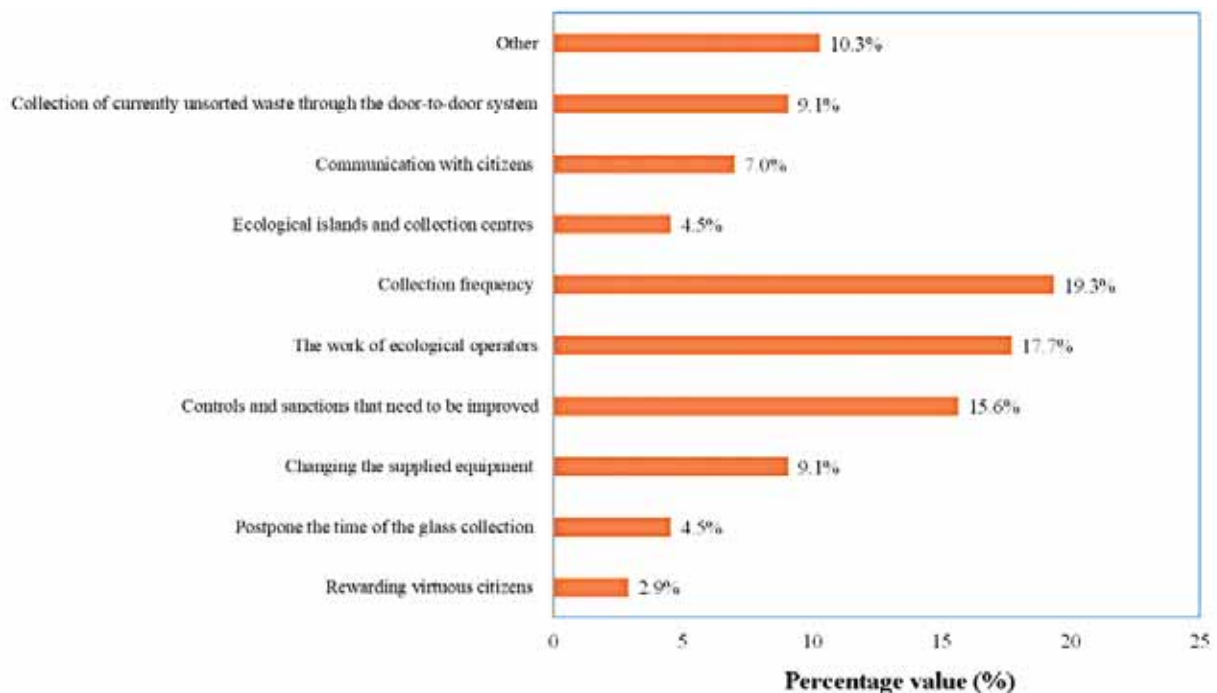


Fig. 6. Criticalities highlighted by the users during the first year of operation (Q₁₃)

4. Conclusions

Based on the obtained results, the following outcomes can be pointed out:

- The heterogeneous nature of a metropolitan area from an urban point of view required careful planning and implementation of the MSW separate collection service, especially in the case of the door-to-door. Among the preliminary steps there was the identification of areas with homogeneous urban characteristics such as population density, type of dwellings, availability of condominium space, availability of space on the sidewalks. The elaboration of a map of the Homogeneous Territorial Zones (HTZ) allowed to classify the areas in terms of vocation to the door-to-door collection; the HTZ map was then used to identify the Start-up Zones together with other criteria such as comparable population distribution as well as respect for the physical/administrative boundaries of the territory.

- Once the door-to-door system was designed in its main variables (e.g. equipment, collection vehicles, personnel), it was advisable to start the system in successive steps, starting from the most vocated areas and providing simultaneous and continuous communication campaigns for those less vocated.

- With reference to the case study of the municipality of Bari (Southern Italy), the excellent performance in terms of waste separately collected (>80%) highlighted the goodness of the adopted technical approach, as well as the convenience in acquiring feedback from users during the starting of the service. While expressing positive satisfaction with the door-to-door system with an overall percentage of 72%, users consider the adopted sanctioning and control system to be critical. The same was considered insufficient to deal with the well-known phenomenon of “waste tourism”.

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NITROGEN AND *Escherichia coli* REMOVAL IN FACULTATIVE FINISHING LAGOONS RECEIVING TREATED URBAN WASTEWATER

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Abstract

The scarcity of water resources, especially in the agricultural sector, is an increasing problem both in developing and industrialized countries. Recent studies and reports have shown the high potential of wastewater reuse mostly in South European countries. In this paper, we study the possibility to reuse wastewater from the Santerno full-scale wastewater treatment plant located in Imola (Bologna, Italy). Specific monitoring campaigns have been carried out in Basin 1 of the natural finishing treatment of the plant and these data are analysed and discussed. The Nitrogen and *Escherichia coli* degradation has been analysed with respect to the nitrification/denitrification and disinfection processes in the water volume. Furthermore, we have implemented a prediction model for the *Escherichia coli* degradation in Basin 1 and compared the results with the measured data. The comparison results are encouraging, showing that a future implementation of the model on Basin 1 is possible. Finally, the first data collected on a pilot plant designed and realized near Basin 1, are discussed. The *Escherichia coli* data collected in pilot plant and Basin 1, show that the main part of the disinfection process occurs in the upper layer of Basin 1 (around 60 cm). Consequently, this layer is crucial in order to define future management policies that can be tested on the pilot plant and then adopted on the full-scale plant.

Keywords: disinfection; finishing lagoon; nitrogen removal; solar radiation; wastewater reuse

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1. Introduction

The lack of water resources availability for human activities is a very critical and current problem all over the world. This problem is decisive in agricultural sector where the water consumption is very high referred to the annual global freshwater withdrawals (Tran et al., 2016). In this context water reuse has a key role. In particular, the developing of smart wastewater reuse practises can be a very interesting future solution. The global wastewater reuse volume is increasing and this trend is expected to be confirmed in the coming years (Kirhensteine et al., 2016) (BIO by Deloitte, 2015)(Sanz and Bernd, 2014).

Therefore, the rising attention on wastewater reclamation pushed on the development of guidelines

and regulations both for the promotion of wastewater reuse and for human health and environment protection, by defining correctly the water quality indicators, their specific reuse and threshold values.

At European level, no specific directive for wastewater reuse has been implemented yet. Instead, several environmental directives used at member states and regional levels exist for the implementation of national laws and standards. Regulations are thus highly heterogeneous, especially in terms of intended uses, analytical parameters and permitted threshold values. This regulation heterogeneity represents one of the main barriers to the development of wastewater reuse at European level. Recently, the European Commission (EC) puts water reuse as key point of the Circular Economy action plan to overcome this problem. In 2016 the EC asked to the Joint Research

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Centre (JRC) to develop a technical report for water reuse in agricultural irrigation. After a first draft in October 2016, the JRC published a final version in June 2017 (Alcalde-Sanz and Gawlik, 2017) in which minimum quality requirements for water reuse have been proposed dividing the reclaimed water in four quality classes associated to different agricultural uses. Moreover, the EC requested an additional study to the Technical University of Munich who published a complete report in October 2017 (Drewes et al., 2017). Starting from the studies, on May 2018 the European Commission published a “Proposal for a Regulation Of The European Parliament And Of The Council on minimum requirements for water reuse” in order to overcome this regulation heterogeneity.

Table 1 shows the legal thresholds values for irrigation reuse in five European countries: Italy (DM 185, 2003), Spain (RD 1620, 2007), France (JORF 153, 2014), Greece (CMD 145116, 2011), Cyprus (Law 106, 2002) compared to the 2018 EC proposal (European Commission, 2018). The legal thresholds are referred to five main parameters: Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Total Nitrogen (TN), Ammonium (NH₄⁺) and *Escherichia coli* (*E. coli*). The development of wastewater reuse for irrigation deals with several barriers: cultural, economic, food security. Moreover, most of the existing Wastewater Treatment Plants (WWTPs) have been designed and built to respect the limits for discharge in natural water bodies. Thus, their adaptation for irrigation reuse will be an important challenge in the near future. This adaptation can be complicated in large plants due to the lack of large surface availability near them. Indeed, wastewater reuse for irrigation needs large surfaces for storage basins. In medium plants, less than 100000 Population Equivalent (PE), this availability appears more feasible even if the existing processes are verified and studied with respect to two main requirements: 1) to respect the legal thresholds and, 2) to guarantee the water storage in order to comply with irrigation needs.

Thus, the adaptation of existing WWTPs to irrigation reuse needs appears to be a very interesting option even if its feasibility is also connected to the achievement of very stringent legal thresholds, specifically *E. coli* (Table 1). On the other hand, the achievement of COD and TN limits could be easier as

they are similar with the ones for discharge in water bodies.

Nevertheless, Nitrogen discharge in soils must be taken into account because high concentrations can reduce the crops quality due to overstimulation, lodging or maturity delay (Lazarova and Bahri, 2005). Indeed, Nitrogen compounds could be already present in the soil due to addition as fertilizer. Besides, a reduction of Nitrogen concentration enables discharging effluent wastewater after irrigation in water bodies, with a high tendency for eutrophication phenomena (Mancini, 2004).

In this context, we studied the existing WWTP of the city of Imola (Bologna, Italy), called “Santerno municipal WWTP”, where a traditional municipal plant with predenitrification/nitrification is followed by five facultative lagoons as tertiary finishing treatment. The final aim of the study is to adapt the existing plant for irrigation reuse diverting a part or the entire effluent flow from the first facultative lagoon (Basin 1) fulfilling the seasonal irrigation requirements. This adaptation can be implemented only if the effluent from Basin 1 will respect the legal thresholds. We refer on the Italian thresholds (DM 185, 2003) as they are very stringent, in some cases (*E. coli*) more stringent than the ones proposed by EU countries (Table 1).

In wider terms, the implementation of natural wastewater treatment systems allows for Nitrogen reduction, thanks to two main processes due to algal and biomass activities (Malschi et al., 2018), as well as for natural disinfection that depends on the solar irradiation, pH and temperature and takes place both in aerobic and anoxic conditions (Liu et al., 2016; Pozo-Morales et al., 2014).

We studied more particularly the nitrification/denitrification and natural disinfection processes occurring in Basin 1 through the analysis of the data collected during specific monitoring campaigns conducted from May 2016 to July 2017. Furthermore, we have implemented the *E. coli* degradation model based on the dispersed flow equation from Wehner and Wilhelm (Wehner and Wilhelm, 1956) in order to test its usability in full scale case. We studied the disinfection capacity of Basin1 with respect to solar irradiation variations both on the water surface and in the water column.

Table 1. BOD, COD, TN, NH₄⁺ and *E. coli* legal limits for irrigation reuse in five European countries and EU Proposal in 2018

Parameter	Italy (DM 185, 2003)	Spain (RD 1620, 2007)	France (JORF 153, 2014)	Greece (CMD 145116, 2011)	Cyprus (Law 106, 2002)	EC proposal (European Commission, 2018)
BOD (mg/L)	20	-	-	10-25	10-70	10-25
COD (mg/L)	100	-	60	-	70	-
TN (mgN/L)	15	10	-	30	15	-
NH ₄ ⁺ (mgNH ₄ ⁺ /L)	2	-	-	-	-	-
<i>E. coli</i> (CFU/100mL)	10 ^a	0-10 ⁴	250-10 ⁵	5-200	5-10 ³	10-10 ⁴

a. It is the limit for 80% of the samples while 100 CFU/100mL is the maximum limit for all cases. The limit is higher using natural systems (phyto-depuration or lagoons) becoming: 50 for 80% of the samples while 200 CFU/100mL is the maximum limit for all cases.

In particular, the presence of the aquatic macrophyte *Lemna minor* (*Lemna*) on the water surface has been estimated during the monitoring campaigns and taken into account in the model implementation as it strongly influences the light penetration in the water column.

Finally, a pilot plant has been designed and realised in the Santerno plant area in order to study the natural disinfection process in the upper layer of Basin 1 measuring the *E. coli* concentration.

The study has been developed in the frame of a partnership project between the Department of Civil, Chemical, Environmental and Materials Engineering (DICAM) of the University of Bologna and the multiutility HERA S.P.A., responsible of the water and wastewater management in Bologna region. The general aim of the project was the management optimization of the Santerno plant with the specific aim of the implementation of the lagoon basins for irrigation reuse by analyzing, for the first time in this site, chemical and microbiological phenomena.

2. Materials and method

The study is based on data collected during measurement campaigns conducted from 25 May 2016 to 24 July 2017 both in the first lagoon (Basin 1) of the tertiary natural treatment phase of the Santerno full scale WWTP and in the pilot plant located in the plant area. The full-scale plant is fed on urban wastewater from the city of Imola and hinterland, 75000 PE, with an average influent flow rate of 25000 m³/day. As shown in Fig. 1, the overall plant scheme can be divided in two main parts: primary/secondary treatments and natural finishing treatments. After the primary treatment (screening) without primary sedimentation, the influent sewage goes to the secondary treatments made by denitrification and

nitrification tanks as active sludge process and secondary sedimentation. Finally, five natural treatment basins provide for the finishing and natural disinfection treatment before the final discharge into the Santerno river.

Basin 1 follows the secondary treatments by treating half the effluent flow that can easily be bypassed for irrigation reuse (see dotted arrow in Fig. 1). Its volume is around 23000 m³ with a water surface of 14000 m², consequently its Hydraulic Retention Time (HRT) is around 2 days. Besides, *Lemna minor* grows in this basin occupying its whole surface during summer and leading to an equilibrium between phyto-treatment and Free Water Surface (FWS) lagoon.

In a first step of the study, we divided the basin into four sections perpendicular to the main flow direction. Hence, we measured the water depth at each black point for each section obtaining the cross-sections profiles (Fig. 2). Afterwards, we measured Temperature (t) and Dissolved Oxygen (DO) along the water column at each point marked with black/white dots in Fig. 2, using the multiparameter system YSI 556. Moreover, we collected samples in the middle of the water column of the same points in order to measure Ammonium Nitrogen (NH₄⁺-N), Nitrate Nitrogen (NO₃⁻-N), Total Nitrogen (TN) and *E. coli*. Ammonium Nitrogen has been measured with the Ion Selective Electrode Crison 9663C, NO₃⁻-N with Ion Chromatograph DX-120 and Total Nitrogen according to the APHA methods for water and wastewater (APHA, 1998).

E. coli were enumerated by membrane filtration method using membrane-Thermotolerant *Escherichia coli* Agar (modified mTEC) according to the EPA Method 1603 (Usepa, 2009). After filtration, using a Whatman system, the filters were placed on the mTEC in Petri plats and incubated at 35°C for 2 hours and, afterwards, at 44.5°C for 22 hours.

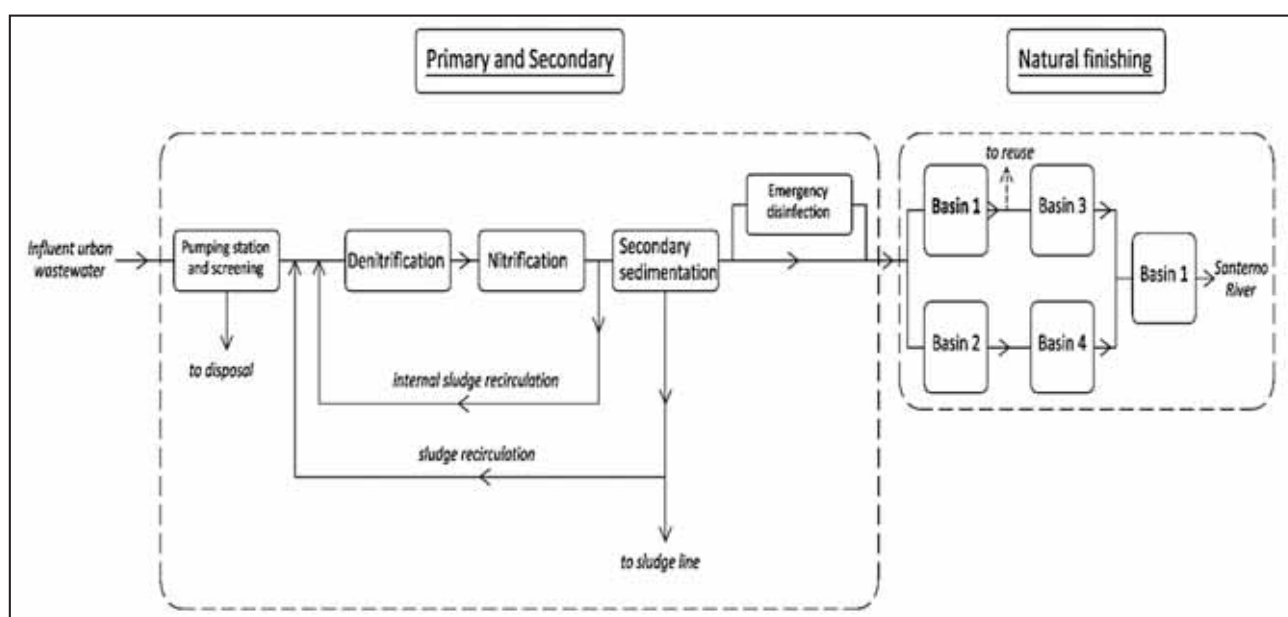


Fig. 1. Treatment scheme of the Santerno WWTP in Imola (Bologna, Italy)

We implemented a model for *E. coli* degradation in Basin 1 using the measured input data. We then compared the simulated results with the data measured in section D-D'. The model is based on the most common solution for the steady-state differential equation (Eq. 1) in its dimensionless form (Wehner and Wilhelm, 1956). The Equation is commonly used for chemical reactors design. In 1969 Thirumurthi proposed its application to BOD removal modelling (Crites et al., 2006) in facultative ponds under the hypothesis that they can be considered something between Plug Flow Reactor (PFR) and Completely Mixed Flow Reactor. Recently, the equation has been adapted to *Fecal Coliform* and *E. coli* degradations in constructed wetlands by Khatiwada N.R and Polprasert C. (Hamaamin et al., 2014; Khatiwada and Polprasert, 1999) and under the same hypotheses. Consequently, we implemented the dispersed flow equation under the following hypotheses:

- 1) hydraulic behaviour between Plug Flow Reactor and Complete Mixed Flow Reactor;
- 2) the mechanism of pathogen removal is due to the effects of temperature, solar radiation, sedimentation, adsorption and filtration.

$$\frac{C_e}{C_0} = \frac{4a_i e^{\frac{1}{2d}}}{(1+a)^2 e^{\frac{1}{2d}} - (1-a)^2 e^{\frac{1}{2d}}} \quad (1)$$

where: C_e = influent *E. coli* (CFU/100 mL); C_0 = effluent *E. coli* (CFU/100 mL); $a = \sqrt{1+4kTd}$ (-); $k = K_t + K_f + K_i$ overall removal rate coefficient (day⁻¹); $K_t = K_{t,20} * \Phi^{t-20}$ removal rate coefficient due to temperature at °C (day⁻¹); $K_i = \varphi * I_{av}$ removal rate coefficient due to solar radiation (day⁻¹);

$$I_{av} = \frac{I_0}{\tau \cdot h} (1 - e^{-\tau h}) \text{ average solar radiation (cal/m}^2\text{day);}$$

$$K_f = \frac{4}{\pi} \eta \alpha \frac{u(1-\vartheta)}{d_c} \text{ removal rate coefficient due to}$$

adsorption, filtration and sedimentation (day⁻¹);

$$\eta = 0.9 A_s^{1/3} \cdot \left(\frac{K_B T_a}{u d_c d_p u} \right)^{2/3} + \frac{2}{3} A_s \left(\frac{d_p}{d_c} \right)^2 + \frac{\rho_p - \rho}{18 \mu u}$$

$$\text{removal efficiency (-); } A_s = \frac{2(1 - \epsilon^5)}{2 - 3\epsilon + 3\epsilon^5 - \epsilon^6}$$

parameter accounting for the effect on adjacent media grains on the flow about a collector (-); $\epsilon = (1 - \theta)^{1/3}$ parameter accounting for the porosity (-).

Table 2 shows the parameter values used in the model implementation. They are based on literature review and a previous study of the authors (Fiorentino et al., 2016). In this case, a specific measurement campaign on the solar irradiation (I_0) has been carried out in the Santerno WWTP site. The solar irradiation was measured with a PIRSC-GEOVES pyranometer located near Basin 1 and the data were acquired and stored with a data logger stand-alone, dataTaker DT 80, at a frequency of one per minute.

In order to implement the model to Basin 1 we have followed different steps. First, the flow velocity in each chosen section has been calculated with Chézy formula, considering the influent flow rate and the cross area measured. Second, Basin 1 has been schematized as four rectangles, where flow velocity (v) and water depth (h) remain constant (Fig. 3). Third, Basin 1 has been discretized into 10m spaced sections and calculated the corresponding HRT. Finally, (Eq. 1) has been solved assigning the *E. coli* input concentration as C_0 in the first section. The natural disinfection process mainly depends on the solar radiation and its capacity to pierce through the water.

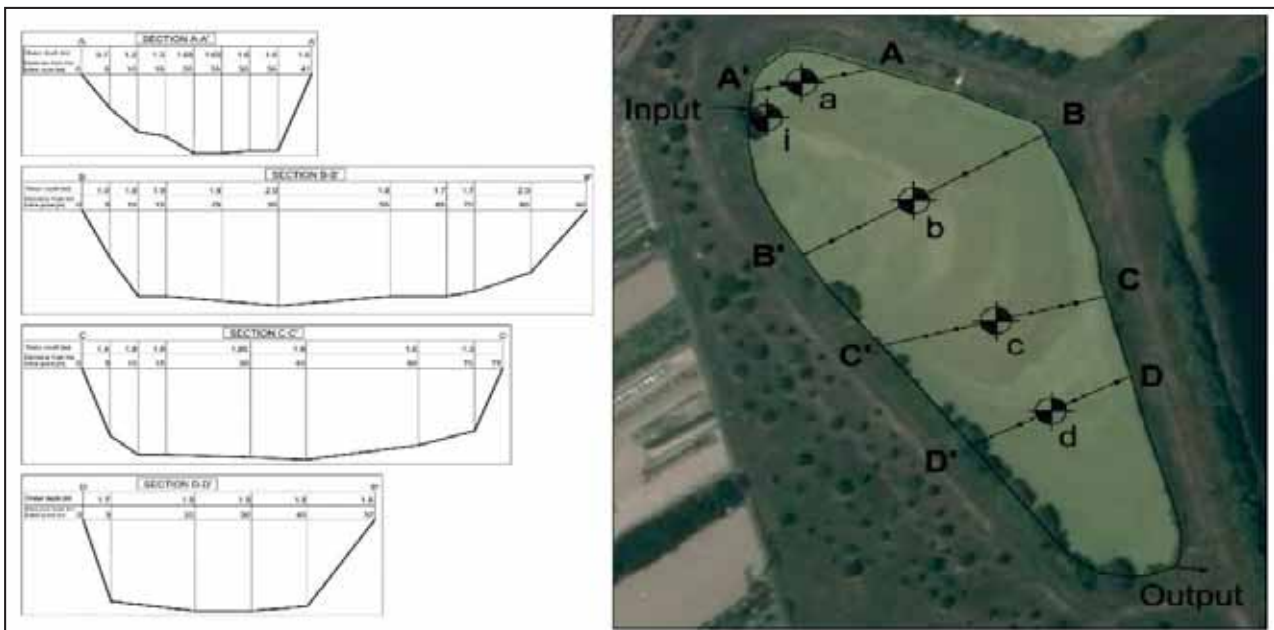


Fig. 2. Basin 1: cross-sections profiles (left) and aerial view (right) with sampling points (i, a, b, c, d) marked with black/white dots and water depth measurement points marked with black dots

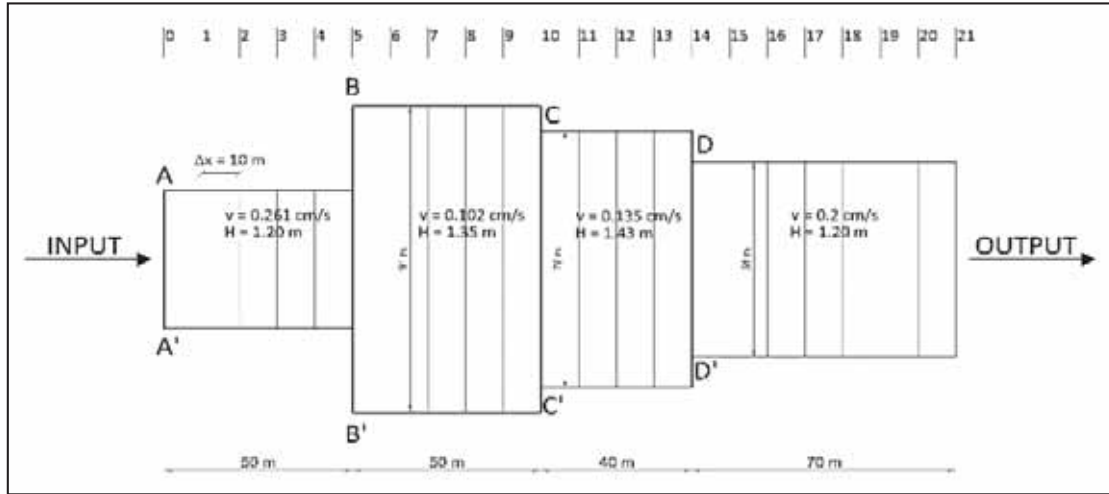


Fig. 3. Basin 1: scheme for *E. coli* model implementation

It is possible that the solar radiation pierces only through the upper layer of Basin 1 so the disinfection effect should not interest the entire water column. Moreover, the presence of *Lemna* on the water surface shades the solar radiation reducing this capacity and its thickness intensify the effect. Starting from these hypotheses, we first verified the *E. coli* degradation in the pilot plant realized in the Santerno WWTP area, by collecting samples in two points: near the input (PP_input) and output (PP_output). These sampling points are marked with black/white dots in Fig. 4.

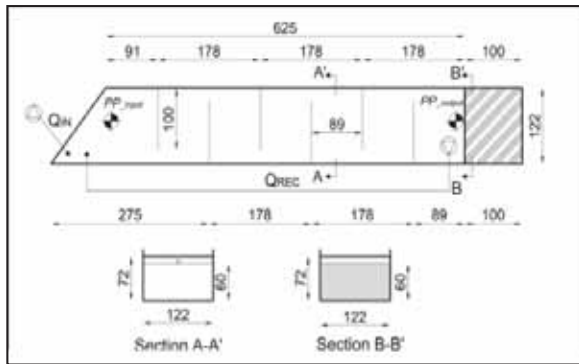


Fig. 4. Pilot plant: plan and sections (left) and picture from the outlet (right). Measures are in centimetres and the black and white dots represent the sampling points

The pilot plant has been designed to be a plug flow reactor and to represent the upper layer of Basin 1 (around 60 cm). The tank is equipped with six baffle walls and two pumps: the first for inlet flow (Q_{IN}) and the second for recirculation flow (Q_{REC}). The input and recirculation flow rates have been fixed to 0.02 l/s and 1.15 l/s in order to have HRT equal to 2 days. The water surface was covered by a constant thickness of *Lemna minor* during all the experimental period.

3. Results and discussion

3.1. Nitrification/Denitrification processes on Basin 1

We present the results for the Ammonium Nitrogen, Nitrate Nitrogen and Total Nitrogen analysis in Table 3. The basin surface covered by *Lemna* has been evaluated in each measurement campaign to take into account its finishing effect (Fig. 5). Observing the NH_4^+-N data in section A-A', three periods can be identified:

- 1) from 25 May 2016 to 26 October 2016 the NH_4^+-N concentration is in the range 3.5 - 8.6 mgN/L;
- 2) from 30 November 2016 to 24 May 2017 with lower values (1.3 - 1.7 mgN/L).
- 3) the last NH_4^+-N value in section A-A' return in the range of the first period (6.0 mgN/L).

Table 2. *Escherichia coli* degradation model: parameters used

Parameters	Unit	Value	Reference
d	Dispersion number	(-)	0.15 (Polprasert et al., 1998)
$K_{t,20}$	Removal rate coefficient at 20°C	(day ⁻¹)	0.047 (Khatiwada and Polprasert, 1999)
ϕ	Temperature coefficient	(-)	1.07 (Mancini, 1978)
φ	Light mortality constant	(cm ² /cal)	0.0103 (Sarikaya et al., 1987)
τ	Vertical light extinction coefficient	(m ⁻¹)	25 (with <i>Lemna</i>) 1 (without <i>Lemna</i>) (Khatiwada and Polprasert, 1999)
α	Sticking efficiency	(-)	0.003 (Khatiwada and Polprasert, 1999)
θ	Porosity	(-)	0.52 (Khatiwada and Polprasert, 1999)
d_c	Duckweed root diameter	(m)	1.76×10^{-4} (Cedergreen and Madsen, 2002)
d_p	<i>E. coli</i> diameter	(m)	1×10^{-6} (Khatiwada and Polprasert, 1999)
ρ_p	<i>E. coli</i> density	(kg m ⁻³)	1050 (Khatiwada and Polprasert, 1999)

These variations are due to different management policies in the Primary/Secondary treatments, especially for the denitrification/nitrification processes. As the aim of this study is to investigate the possibility to reuse the wastewater coming from a common and existing urban WWTP, the capacity of the natural treatment basin to face such variations is crucial to respect the legal thresholds.

The 25 May 2016 data show ammonium nitrogen decrease while nitrate nitrogen increases and TN remains under the Italian legal thresholds for irrigation reuse. Indeed, the input data are typical of a partial nitrification of the secondary treatment. The ammonium nitrogen reduction in the basin is only due to the nitrification process and there is no evidence of photosynthetic activity. Moreover, *Lemna* did not influence the process as it occupied only a small area near the inlet section.

On 15 June 2016 the conditions started to change with a decrease of NO₃⁻-N in section D-D' as well as a lower NH₄⁺-N removal efficiency even if NH₄⁺-N in input is comparable with the previous case.

In this case, the data reveal typical aerobic lagoon conditions with photosynthetic activity. The finishing effect of *Lemna* started to influence the process because it covered approximately one third of the basin surface near the bank.

On 13 July 2016 almost all the basin surface was covered by *Lemna*. We observe a maximum finishing effect as shown by the TN removal efficiency, around 40%. In particular, the highest TN decrease is observable in the middle of the basin, at sections B-B' and C-C', where *Lemna* is better established (Fig. 5). The TN removal efficiency increases on 26 October 2016 due to lower nitrification effect, while NH₄⁺-N reduction is minimal. Moreover, *Lemna* covers one fourth of the surface near the D-D' section, but its finishing effect is again minimal because its main part is not in vegetative phase. On 30 November 2016 and 22 February 2017, the finishing effect of the lagoon is much lower than before. Indeed, these analyses were conducted during the winter season when the photosynthetic activity was almost absent and there was not *Lemna* on the surface.

Table 3. Basin 1: Ammonium Nitrogen (NH₄⁺-N), Nitrate Nitrogen (NO₃⁻-N) and Total Nitrogen (TN) data in the middle of the sections A-A', B-B', C-C' D-D'

Meas. campaign	Sections													
	A-A'			B-B'			C-C'			D-D'			Removal efficiency	
	NH ₄ ⁺ -N	NO ₃ ⁻ -N	TN	NH ₄ ⁺ -N	NO ₃ ⁻ -N	TN	NH ₄ ⁺ -N	NO ₃ ⁻ -N	TN	NH ₄ ⁺ -N	NO ₃ ⁻ -N	TN	NH ₄ ⁺ -N	TN
(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(mgN/L)	(%)	(%)
25 May 2016	8.6	7.5	16.2	3.5	8.9	18	3.9	9.2	13.3	3.7	9.3	13.2	57	19
15 June 2016	6.7	6.8	13.7	4.2	7.3	14.5	4.4	7.3	11.8	3.7	7.2	10.9	45	20
13 July 2016	3.2	5.2	8.6	3.1	3.2	6.5	2.7	2.6	5.4	2.6	2.6	5.2	19	40
26 October 2016	3.5	7.9	11.9	3.5	8.3	12	3.2	8.6	11.8	3.2	8.5	11.8	9	1
30 November 2016	1.3	8.9	11.3	0.9	11.1	12.1	1.0	11.0	12.0	1.0	10.0	11.2	23	-
22 February 2017	1.4	20.1	21.9	1.4	19.2	22.0	1.4	18.7	20.1	1.6	18.7	21.0	-	4
22 March 2017	1.7	12.8	14.5	2.2	11.3	13.7	2.4	10.9	13.4	2.4	10.8	13.2	-	9
24 May 2017	1.7	12.9	14.8	1.6	14.3	16.0	1.5	13.8	15.3	1.5	14.6	16.4	12	-
21 June 2017	6.0	4.4	10.3	5.3	4.7	10.2	4.4	4.3	8.8	4.4	4.8	9.3	26	10

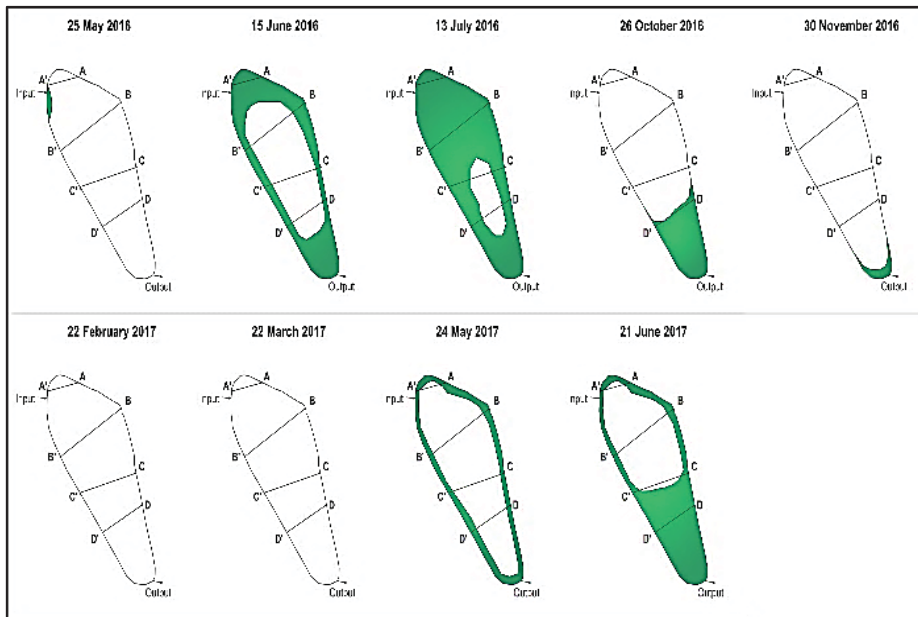


Fig. 5. Surface of Basin 1 occupied by *Lemna* during measurement campaigns

The Nitrate concentration in 22 February 2017, 22 March 2017 and 24 May 2017 measurement campaigns are higher than the others probably due to the efficiency reduction of the denitrification process, as explained before. However, in these cases the NO₃⁻-N reduction efficiencies were around 15% showing a finishing capacity of Basin 1 also towards the Nitrate. The last measurement campaign (21 June 2017) shows a behavior comparable to the first period with a decrease of ammonium and nitrate in input and a good finishing capacity of Basin 1. Temperature and Dissolved Oxygen have been measured along the water column in sections B-B' and C-C' and the results shown in tables from 4 to 6.

The 25 May 2016 data (Table 4) show temperature values around twenty degrees, typical for this season, without evidence of particular trends in depth. DO confirms the aerobic condition in all the

water volume and consequently nitrification process underway. Higher DO percentage in section B-B' is not due to photosynthetic activity but to the oxygen dissolved during the input. Indeed, sewage flows in Basin 1 through a free surface channel with a diameter of one meter and a length around three meters.

DO percentage on 15 June 2016 (Table 5) confirms the photosynthetic activity of phytoplankton, with higher values on the surface than on the bottom. The DO percentage does not show anoxic conditions so there is no denitrification in the bottom layer. Temperature are higher than on 25 May 2016 and approximately constant along the water column.

In addition to the presence of *Lemna* in Basin 1, the increase of Nitrogen removal efficiency observed on 13 July 2016 is also due to low DO concentration in the bottom layer of the water column (Table 6). Indeed, the anoxic conditions implied the denitrification process have taken place where the water depth was approximately more than 1.10 m. This is confirmed by the nitrate reduction efficiency around 50% (see Table 3).

The DO percentage indicates unsaturated conditions in the water column. This is due to the *Lemna* coverage, which reduced the oxygen transfer from the air to the water, and also to the reduction of the photosynthetic activity as the solar irradiation did not penetrate. Moreover, DO decrease in Summer can be also attributed to lagoon water temperature. Thus, water temperatures rise in Summer (Table 6) involves DO saturation decrease and in turn DO decrease.

Table 4. Data from the Basin 1: Temperature (t) and Dissolved Oxygen (DO) in the water column - 25 May 2016

Depth (m)	B-B'			C-C'		
	t	DO		t	DO	
	(°C)	(%)	(mg/L)	(°C)	(%)	(mg/L)
0.30	20.53	81.0	7.26	20.75	71.7	6.39
0.50	20.11	84.0	7.58	20.77	71.5	6.38
0.70	19.84	83.0	7.54	20.45	71.3	6.40
0.90	19.49	78.8	7.20	20.20	72.1	6.50
1.10	19.46	77.2	7.06	19.90	69.5	6.30
1.30	19.40	75.8	6.94	19.89	71.3	6.47
1.50	19.55	70.7	6.45	19.88	71.7	6.39

Table 5. Data from the Basin 1: Temperature (t) and Dissolved Oxygen (DO) in the water column - 15 June 2016

Depth (m)	B-B'			C-C'		
	t	DO		t	DO	
	(°C)	(%)	(mg/L)	(°C)	(%)	(mg/L)
0.30	22.12	112.4	9.77	22.30	98.0	8.53
0.50	22.10	113.9	9.90	22.30	92.1	8.02
0.70	22.10	111.0	9.65	22.23	89.0	7.74
0.90	22.10	110.8	9.63	22.22	87.2	7.56
1.10	22.11	110.1	9.57	22.21	86.2	7.47
1.30	22.14	111.1	9.65	22.08	85.2	7.38
1.50	22.12	111.7	9.70	22.01	82.7	7.16

This effect is less relevant in Spring (Tables 4-5) than in Winter. The presence of phytoplankton and organic matter, from bacterial and *Lemna* degradations, especially in Summer, enriches the water environment with organic matter which promotes bacterial growth and in turn effects DO levels.

3.2. Disinfection effect

The natural disinfection capability of Basin 1 has been tested measuring the *E. coli* concentration in different sampling points shown in Fig. 2 (i, b, c, d).

Starting from similar values in input, the overall efficiency changed significantly from 25 May 2016 to 13 July 2016 (Table 7) mainly due to the *Lemna* growth that covered the surface preventing the

solar irradiation from penetrating the water. The single measurement in autumn shows a good removal efficiency (87%) but the *E. coli* concentration is not under the Italian legal thresholds yet. During Winter/Spring seasons, from 22 February 2017 to 24 May 2017, the *E. coli* removal efficiency was over 96% because there was not *Lemna* coverage and the solar irradiation permits the natural disinfection. This behaviour is evident in February, when the disinfection efficiency was 98% even if the solar radiation was not maximum. Only two output values (22 February 2017 and 24 May 2017) are under the legal thresholds showing that this goal is very hardly achievable. Finally, the last measurement campaign conducted in July (24 July 2017) shows an unexpected increase of *E. coli* concentration from input (2.4×10^2) to output (3.88×10^2).

Table 6. Data from the Basin 1: Temperature (t) and Dissolved Oxygen (DO) in the water column - 17 July 2016

Depth (m)	B-B'			C-C'		
	t	DO		t	DO	
	(°C)	(%)	(mg/L)	(°C)	(%)	(mg/L)
0.30	26.00	44.01	3.55	26.67	50.65	4.10
0.50	25.93	43.27	3.50	26.18	48.59	3.93
0.70	25.83	43.94	3.56	26.20	46.35	3.76
0.90	25.67	44.84	3.64	25.94	45.20	3.67
1.10	25.36	18.82	1.54	25.79	42.58	3.43
1.30	25.19	15.96	1.20	25.75	42.19	3.39
1.50	25.15	12.30	0.48	25.74	27.88	2.22

Table 7. *E. coli* (CFU/100 mL) in Basin 1 - standard deviations in bracket

Measurement campaign	Sections					Removal efficiency
	A-A'	B-B'	C-C'	D-D'	A-A'	
25 May 2016	2.7×10^3 (3.2×10^2)			6.77×10^2 (3.01×10^2)	3.67×10^2 (1.86×10^2)	86%
15 June 2016	1.4×10^3 (2.2×10^2)		9.27×10^2 (1.95×10^2)		5.63×10^2 (1.70×10^2)	59%
13 July 2016	2.8×10^3 (1.9×10^2)		1.85×10^3 (1.31×10^2)		1.76×10^3 (1.04×10^2)	36%
30 November 2016	3.2×10^3 (2.9×10^2)		8.77×10^2 (3.86×10^2)		4.05×10^2 (7.07×10^0)	87%
22 February 2017	2.2×10^3 (4.9×10^2)	4.33×10^2 (1.06×10^2)	3.30×10^2 (5.93×10^1)	4.75×10^1 (2.50×10^1)	3.75×10^1 (1.71×10^1)	98%
22 March 2017	1.2×10^4 (8.0×10^2)	1.57×10^3 (9.45×10^1)	8.30×10^2 (7.07×10^1)	5.23×10^2 (4.04×10^1)	2.28×10^2 (2.50×10^1)	98%
24 May 2017	7.5×10^2 (3.5×10^2)			3.00×10^1 (1.41×10^1)	3.33×10^1 (1.15×10^1)	96%
24 July 2017	2.4×10^2 (5.1×10^1)			3.88×10^2 (1.94×10^2)		-

This behavior could be due to abnormal increases of *E. coli* concentration in plant input during the previous days that should have caused *E. coli* accumulation. Consequently, Basin 1 HRT was not enough to reduce the concentration under the legal thresholds for reuse using only one lagoon.

Fig. 6 shows the results of *E. coli* degradation model implementation. The Input data (see Table 7) of each measurement campaign have been taken as starting values for the model and are shown in each picture with their respective standard deviations.

Moreover, the pictures show the D-D' values with their standard deviations to compare them with the modelled values in the same section.

The model describes adequately the natural disinfection in Basin 1 even if the discrepancy from the measured values is high when all the Basin surface is covered by *Lemma*. As said before, in such cases, the solar radiation penetration is influenced by the *Lemma* thickness that in turn depends on its accumulation rate. The model did not allow to consider the *Lemma* thickness

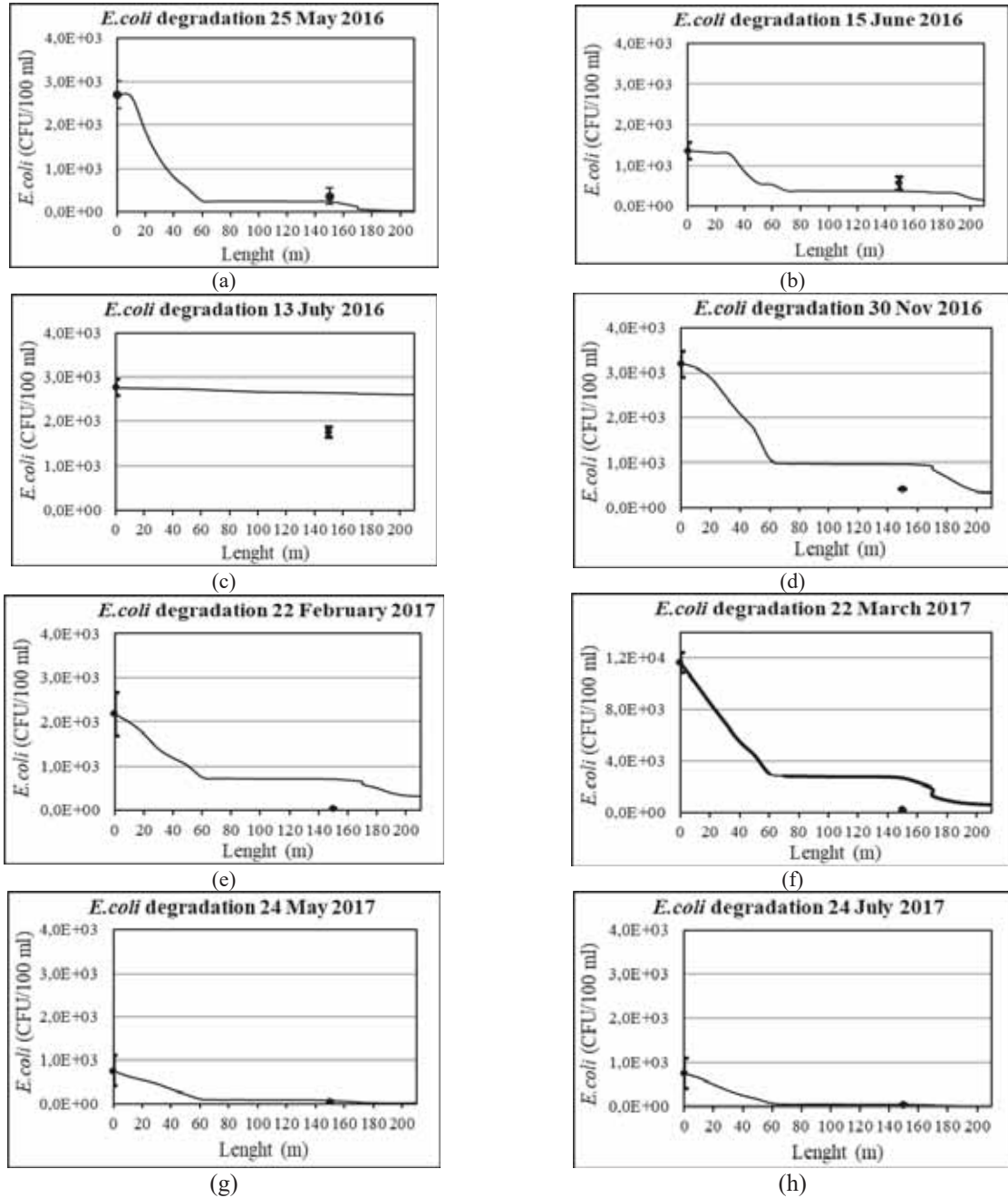


Fig. 6. Basin 1: comparison between *E. coli* measured in sections A-A' and D-D' (black points with standard deviations lines) and *E. coli* degradation modelled (black lines) in each measurement campaign: 25 May 2016 (a), 15 June 2016 (b), 13 July 2016 (c), 30 November 2016 (d), 22 February 2017 (e), 22 March 2017 (f), 24 May 2017 (g), 24 July 2017 (h)

Between 13 July 2016 and 24 July 2017, we collected samples and analysed them to obtain the *E. coli* concentration in input (PP_input) and output (PP_output) sections of the pilot plant. The samples in Basin 1 and in the pilot, plant have been collected at the same time. Table 8 shows the results in terms of *E. coli* concentration with the standard deviations in bracket and the overall *E. coli* removal efficiency. Comparing the data in Table 7 and Table 8 we note that starting from comparable input data, the natural disinfection efficiency is more than 83% in all cases except on 30 November 2016, when the input concentration is lower (1.3×10^2). Moreover, the efficiencies reached in pilot plant (Table 8) and Basin 1 (Table 7) are comparable and the output *E. coli* concentration is over the legal thresholds in all cases. However, these concentrations are not so far from the Italian thresholds and we must consider that the pilot plant surface was covered by *Lemna* in all cases, this obviously contribute to decrease the disinfection efficiency.

4. Conclusions

This study deals with the need to adequate existing WWTPs for irrigation reuse respecting the legal threshold and the water volume needs. The results from fifteen measurement campaigns on the first natural finishing basin of Santerno plant (Basin 1) have been analysed in comparison with the legal thresholds from the Italian regulation for wastewater reuse (DM 185/2003).

In the first part of the study we discussed the nitrogen compounds reduction in terms of TN and Ammonium nitrogen. Results confirmed that the irrigation purposes are achievable in terms of TN in all cases apart from two cases when the Nitrate nitrogen concentration in input was very high (20.1 mgN/L and 12.9 mgN/L). Ammonium Nitrogen was above the legal limits in all cases but a very interesting removal efficiency, up to 57%, has been registered during Spring and Summer seasons. In these cases, the Ammonium Nitrogen reduction is mainly due to high DO levels on the water column that involve high nitrification effect along it. This DO increase is due to the already oxygenated input flow and the photosynthetic extraction due to microalgae as the solar radiation is maximum that produce oxygen released in the basin. In such a case, Basin 1 behaviour

can be considered as Free Water Surface lagoon. The presence of *Lemna* minor on the surface during summer reduces the solar radiation capacity to penetrate in the water column and consequently the microalgae activity, nevertheless the *Lemna* synthesis requires ammonium that is reduced on the surface layer. In such a case, Basin 1 behaviour can be considered as phyto-treatment lagoon. Furthermore, in this last case we note that denitrification conditions are possible in the deep layers of the basin (around 1.50 m), as observed during the 17 July 2016 campaign.

The second part of the study focused on the disinfection capacity of Basin 1 analysing the *Escherichia coli* concentration. Results show that *Escherichia coli* concentration in output does not permit the irrigation reuse in six cases on eight when those concentrations are under the stringent Italian legal limit (50 CFU/100mL). Anyway, we observe very interesting disinfection efficiency, up to 98% in two cases. We observed that the natural disinfection process is strongly influenced by the presence of *Lemna* on the surface as shown by the different removal efficiency along the section analysed. Moreover, the tests carried on the pilot plant shown that the top layer of the basin (around 60 cm) is the most important in terms of natural disinfection.

DO increase and *E. coli* decrease in Summer can also due to the presence of phytoplankton and organic matter, from bacterial and *Lemna minor* degradations which influences the bacterial activities and the disinfection capability of the system.

Finally, results show that is possible to achieve the irrigation reuse goals using existing WWTPs equipped with natural finishing lagoons only adopting adequate policies in order to manage the equilibrium between FWS lagoon and phyto-treatment due to *Lemna*. The *Lemna* extraction management in relationship with the seasonal solar radiation variations plays a key role as its presence influences both the nitrification/denitrification and disinfection processes. Those management decisions can be effectively supported by the *E. coli* degradation model tested on the full scale Basin 1 as long as the *Lemna* thickness, expressed as τ in the model, is carefully considered.

Acknowledgements

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Table 8. *E. coli* (CFU/100 mL) in pilot plant - standard deviations in bracket

Measurement campaign	Sections		
	PP_Input	PP_Output	Removal efficiency
13 July 2016	1.4×10^3 (4.9×10^2)	2.0×10^2 (1.5×10^2)	86%
30 November 2016	1.3×10^2 (1.7×10^2)	7.0×10^1 (5.2×10^1)	47%
22 February 2017	2.1×10^3 (2.9×10^2)	2.8×10^2 (1.1×10^2)	87%
22 March 2017	8.0×10^3 (1.1×10^3)	1.3×10^3 (2.0×10^2)	84%
24 May 2017	5.0×10^3 (1.3×10^3)	8.3×10^2 (3.1×10^2)	83%
24 July 2017	1.2×10^3 (5.5×10^2)	1.7×10^2 (7.2×10^1)	86%

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MEMBRANE SYSTEMS AND WATER STRUCTURES

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Abstract

Water is a vital resource for both human needs and the functioning of our ecosystem. Fresh water availability and water use have been a growing problem for centuries. This paper gives a brief overview on the current knowledge of water stress and describes possible solutions. In addition to a differentiated consideration of the various types of water stress, the special importance of water purification by membrane technology and the associated processes is discussed in detail. The economic provision of high-quality water is the focus of these considerations.

Keywords: bulk water, catalytically accelerated, catalyst, membrane filtration, catalytic water treatment, membranes, molecular water, mollik, RO, water stress, water structures

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1. Water stress – What is that?

Water stress is a topic that already filled scientific books for over 2000 years (Lee, 1951; Kenneth, 1997; Spitz et al., 2012; Reig et al., 2019). By looking deeper on this topic, three types of water stress can be differentiated (Fig. 1).

1.1. Quantitative water stress

Water covers more than 70 percent of the earth's surface - but less than one percent of the total mass is directly available for human consumption (Chauhan et al., 2015). Thus, high quality tap water is a precious commodity (Köbel et al., 2018).

According to BATES (Bates et al., 2008), quantitative water stress is present when water abstraction exceeds existing freshwater resources. That means, precipitation risk, evaporation and population growth act as direct factors on the water stress. Climate-induced dry periods have always led to an increase in the salinity and turbidity of surface water resources. This is associated with downstream

ecological and economic chain reactions (Bates et al., 2008; Menzel et al., 2007).

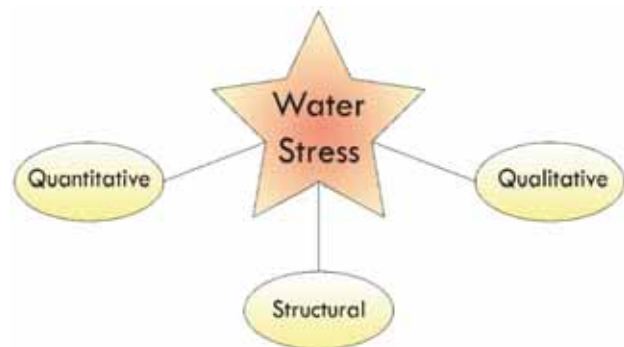


Fig. 1. Types of water stress

1.2. Structural water stress

Any energy input on the water, which is above the osmotic pressure, could end in structural changes within water structures. Drinking water - freshly drawn from a well - will have a different taste

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than water that has been pumped longer distances through pipelines. When creating long drinks, there are differences in taste between "shaking" and "stirring". In the field of pump technology, high energy input is accompanied by the formation of cavitation bubbles. Such processes lead to corrosion phenomena and to a reduction in the lifetime of equipment. Dissolving substances also cause structural changes by creating hydration shells. Structural changes in the water are temporary in nature and are mitigated by kinetic equilibrium reactions.

1.3. Qualitative water stress

For water stress of a qualitative nature, looking back at history, it has long been recognized that marginal and less affluent areas are less able to adapt to the changes associated with qualitative water stress. Without efficient technologies combined with appropriate policy instruments, climatic change leads to greater inequality (Smit et al., 2001). The focus here is on the economic provision of high-quality water together with an optimization of usage concepts. Today, as in the past, thoughtless treatment of high-quality water in water-scarce areas is further aggravating the situation (Küffner, 2018).

2. Water stress – Which solutions?

The question therefore arises: How can you remove salts / undesirable substances from the water at a reasonable cost to obtain enough water with a desired quality? For this purpose, filter systems and biomembranes have developed in the course of the evolution – without life, as we know it, would not be possible. Springs and wells are examples of natural filtration systems. If such drinking water resources are properly maintained, then these can be used for many years to provide high-quality water. In the Middle Ages, hints were given on how far manure piles should be moved from the source to ensure a certain quality standard of the drinking water (Hoppenberg, 2017).

However frequent occurrence of temporary peaks in groundwater usage (Navarro and Carbonell, 2007; Ortuño et al., 2010), as well as less-optimized fertilization (Ehlers, 2018), has a negative impact on natural drinking water resources. As a result, even in Europe, today some former groundwater reservoirs only provide brackish water. To overcome water stress of a qualitative nature, sailors had already in the days of ARISTOTLE a process for drinking water production via reverse osmosis (Lee, 1951). For this purpose, a jar of terracotta was submerged in the deep-sea water. Within a few hours, the jar was filled with purified sweet water. In addition, even at that time the more energy-consuming way of getting sweet water by evaporation was known.

To reduce structural water stress, there are suitable biocatalysts available in nature. In the field of technology, a combination of pump technology and the use of suitable heterogeneous catalysts has been proven successful. The first documented applications

are in the area of water supply in the Roman Empire. Here, the water was lifted instead of pumped. Another point is that volcanic ash was used at the viaduct construction as a catalytically active binder within the concrete. Today we know that this procedure has minimized the risk of deposits in distribution systems. This knowledge formed the basis for explaining the effects of technical mineral-metal catalysts for water treatment.

3. Production of high-quality water – How does it work?

The economic extraction of high-quality water by membrane filtration takes place via: pressure, temperature, electric field.

3.1. Importance of membrane technology

As natural water resources are unequally distributed, increasing qualitative water stress leads to the constant development of desalination technologies by resourceful engineers (Wünsch, 2019). More than 300 million people around the world are already getting their daily needs from desalinated water. For this purpose, currently 21,000 membrane plants in 120 countries are permanently in use (IDA Desalination Yearbook, 2017).

By the present membrane plants - can be provided in an economical manner - with a significantly reduced compared to the classic evaporation energy consumption (Lazarides and Katsanidis, 2003). Currently membrane installations for drinking water production are more economical when compared to classic evaporation processes due to the high energy consumption of the latter one. On the other hand, in membrane technology, pressures of up to 80 bar affect water structures and lead to the formation of deposits, which highly increase the operation and maintenance cost. As more chemicals added on the membranes facility as more chemicals will be found later in the concentrate discharged to the sea. Therefore, these desalination plants create an environmental impact on marine and plant life.

The use of suitable heterogeneous catalysts can suppress negative effects on water structures and at the same time extend cleaning intervals of the membranes (Kochan et al., 2019). As a result, the emitted chemical cargo from the concentrate is reduced in comparison to conventional way of operation. Thereby undesirable side effects on animal and plant life are minimized. To follow these views, let's take a closer look at the interaction of salts and water below.

3.2. Mode of action of a membrane system

What makes the processes of membrane technology so interesting? When looking more closely at membrane processes in aqueous systems, the following points are key:

- The operations are subject to isothermal conditions.

Only pressure changes are found

- Osmotic processes take place in aqueous systems

On the other hand, they are subject to the laws of the gas phase.

- Osmosis is related to ions consisting of solids.

At the molecular level, these ions are far removed from the gas phase.

- The osmosis is independent of the type of hydrated ions.

It depends only on the number of hydrated ions.

In order to understand these interesting aspects in more detail, the processes in the water are considered in more detail below.

3.3. Salts and water

What happens when salts are dissolved? The formation of hydration shells removes molecular water from the aqueous system and the atmosphere during salt dissolution - in conjunction with a decrease in entire volume (Koppe, 2017). At the same time, there is a “stress” on the aqueous system for providing molecular water. With reference to ARISTOTLE, it follows that aqueous systems containing a larger proportion of molecular water are under higher pressure and thus automatically flow to the low-pressure region (lower proportion of molecular water). The water of hydration proportion is thus directly proportional to the number of dissolved ions. Since water of hydration is cloaking dissolved ions, the specific nature of the ions plays virtually no role.

For osmosis, this means that membranes are impermeable to larger liquid agglomerates. Only gaseous molecular water and low molecular weight gases such as carbon dioxide and chloramines can pass through the texture of membranes.

3.4. Salts, water, membranes – development of the theoretical model behind

The writings of ARISTOTLE were intensively studied in the Renaissance - good 1800 years later.

During this time most probably also accessible to Leonardo da Vinci. However, the writings of LEONARDO DA VINCI were initially lost before they emerged towards the end of the twentieth century, so that even a wider public learned about it (Schneider, 2011).

In between, the basic views on membranes processes were not accessible and thus had to be re-worked within the last 250 years. More than 2000 years after the documentation of the osmotic effect of drinking water from seawater, JEAN-ANTOINETTE NOLLET described in 1748 membrane experiments in the field of experimental physics at the Collège in Paris. Only for 60 years are membrane systems available on the market, which can provide high-quality water in the technically relevant area.

At the end of the nineteenth century VAN'T HOFF succeeded in providing a theoretical model, which allowed for a calculation of the phenomenon of osmosis at the same time. The notable idea was the transfer of laws of gases to aqueous systems. For his pioneering work on the analogy of gas pressure and osmotic pressure, he was awarded with the first Nobel Prize in Chemistry in 1901. With VAN'T HOFF's model at the same time further effects, such as the boiling point increase, were mathematically developed.

3.5. Ion mobility and water structures

How does the water move through filters and membranes? Within the water, in the water clusters, which are characterized by hydrogen bonds, proton motions occur at a speed of more than 10,000 km/h. On the other hand, the slow ion migration rates in the water are a few centimetres per hour. At first glance, there is no direct correlation between the two processes.

The water structures are in a continuous thermodynamic controlled cycle. This “cycle of water structures” is determined by pressure and temperature (Samoilov, 1946; Vedamthu et al., 1994; Koppe, 2018). The energetic conditions are presented in Table 1.

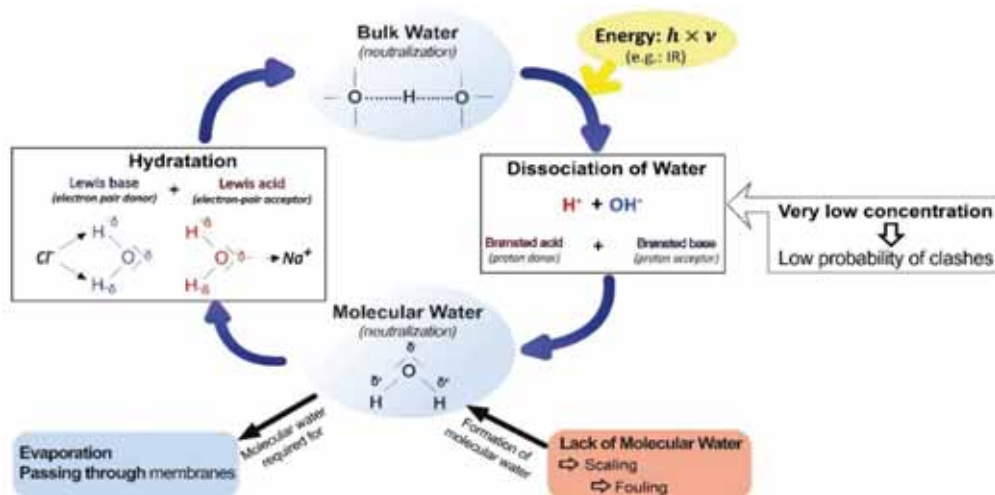


Fig. 2. Cycle of water structures

Table 1. Energetic conditions in the water

Energy of hydrogen bond:	~ 5 kcal/mol
Energy of O-H bond:	~ 100 kcal/mol
Energy of $I^{+/-}$-H₂O bond:	~ 0.005 kcal/mol

In the bulk water, there is an ice-like structure with the well-known "hydrogen bonds". With the addition of melting and dissociation energy, the asymmetric vibrations in the hydrogen bonds increase and dissociation occurs. As a result, there are an acidic proton and a basic hydroxide anion. In the sense of an acid-base reaction, the neutral molecular water is formed. This must be thought of as a single, gaseous molecule.

If ions are present, they interact with the molecular water, resulting in LEWIS acidic and LEWIS basic water molecules. These also react again in the sense of an acid-base reaction and to form hydrogen bonds with the bulk water. Part of the molecular water leaves the system in the form of water vapor depending on the external vapor pressure. For aqueous systems, the following applies (Table 2).

Table 2. Use of supplied energy in aqueous systems

Temperature	Supplied energy is used for
below 0°C	Proton vibration within the hydrogen bond
0 to 100 °C	Dissociation of bulk water / rotational vibration H ₂ O _{molecular} / Proton vibration within the hydrogen bond / partial evaporation

The ion mobility should be thought of as the result of the hydration shell fluctuation, as shown in Table 3.

Table 3. Ion mobility – Theoretical concept

Step	Description
1st	Provision of H ₂ O _{molecular} from the hydrate shell by LEWIS acid-base reaction
2nd	Formation of hydrogen bonds due to neutralization
3rd	Formation of the cluster structure of H ₂ O _{Bulk}
4th	Dissociation of hydrogen bonding
5th	Integration of the newly formed H ₂ O _{molecular} into the hydration shell.

In this case, the ion travels the distance of a water dipole in the millisecond range. When creating an external force field (pressure / temperature / electrical potential), the migration takes place purposefully. During the provision of H₂O-molecular from the hydration shell, the ion is temporarily not completely electrically shielded.

The longer the formation of H₂O-molecular lasts, the greater the risk of precipitation processes. This explains why salts of divalent ions have a lower solubility than salts of monovalent ions. For charge shielding of such ions larger hydration shell is needed, thus the structure takes longer.

3.6. Permeability of membranes

Which molecules fit through a membrane? Looking at the performance of current membranes, it is hard to imagine hydrogen or hydroxide ions migrating through the membrane with their hydration shells. The molecular permeability of today's membranes is in the range between 80 to 150 g/mol (molecular weight of substance). This will also make it too tight for dissolved sodium and calcium ions with their hydration shells. What remains is the molecular water.

As explained above, the molecular water is a gas dissolved in the aqueous system which tends to either evaporate or hydrate (Röntgen, 1892). In order to preferentially direct H₂O-molecules through membranes, a pressure is needed which is above the osmotic pressure. As a result, newly formed H₂O-molecules are deflected from their natural migration to the ions and instead are directed to a membrane surface with higher pressure gradients. Thus, H₂O-molecular disappears from the system and lands on the other side of the membrane. Such deficiencies are associated with the loss of hydration shell water to restore equilibrium. If molecular water/water of hydration decrease, the formation of deposits is accelerated.

The alternatives reduce the loss of hydration shell water are:

1. Accelerated regeneration via supply of external energy (e.g., temperature increase).
2. Catalytic acceleration of the formation of H₂O-molecular.

3.7. Heterogenous catalysts for water treatment

The MOL[®]LIK catalysts consist of a special alloy – made of nickel chromium and iron. The material thickness of the unique metal foils used is about 22 microns. By an activation process, the metal foil is changed as follows (Rastetter et al., 2019):

An approximately 50 nanometre thick mineral layer is formed – with a clear demarcation to the metal structure (half of the volume of the mineral layer absorbs oxygen). As a result, areas with excess electron and electron deficiency are formed within the mineral structure. The electron-rich and electron-poor regions differ significantly in the oxygen concentration and in the number of metal cations. In the electron-rich region, the chromium concentration is higher than in the electron-poor region. In contrast, the iron and nickel concentration in the electron-poor region are higher than in the electron-rich region.

A matter of special importance is the fact that the iron ions in the mineral layer are mainly bound in tetrahedral gaps of a spinel structure. On these lattice

sites they cannot be further oxidized under normal conditions. They have an unpaired electron that is capable of interactions in the sense of a LEWIS base. As the result it comes to the expression of a band edge in the visible light area. In aqueous systems, the following processes take place on the surface of the mineral layer of the MOL[®]LIK catalysts (Table 4).

Table 4. Processes on surface of MOL[®]LIK catalysts in aqueous systems

<i>Option</i>	<i>Description</i>
1a	Reaction of electron-poor water structures (proton excess) with electron-rich regions (BRØNSTED acid-base reaction).
1b	Reaction of electron-rich water structures (hydroxide ion excess) with electron-deficient regions (BRØNSTED-base acid reaction).
2	Reaction of excess hydroxide anions with excess protons Result: formation of H-O-H (molecular water).
3	Adsorption of molecular water at electron-poor regions (LEWIS acid-base reaction). This results in a bond that is more covalent. The unpaired electron on the divalent iron plays a special role here.
4	Further reaction of molecular water with excess protons or excess hydroxide ions by forming cluster structures.

In combination with visible light, the covalent bond of the molecular water is significantly weakened. This leaves the composite accelerated, so that reaction (4) is kinetically disadvantaged. As a result, then molecular water is available faster for other reactions (hydrolysis). Hydrolysis reactions can reduce COD and TOC, especially by degrading free ATP. However, the molecular water ensures a fine dispersion of lime particles, which manifests itself in a lower turbidity and a smaller number of filterable substances. It should be noted that all reactions take place only until the temperature- and pressure-dependent thermodynamic equilibrium position between the individual water structures has re-established itself in the water.



Fig. 3. MOL[®]LIK catalyst module (Product catalog, 2018)
(Capacity: up to 100 m³/h water)

The function of the catalyst lasts for many years. The minimum lifetime is 3 years if properly

handled, with 5 to 10 years possible in technical applications. The prerequisite for optimum functionality is that the catalyst surface is kept free of deposits as far as possible.



Fig. 4. MOL[®]LIK catalyst module
(Capacity: up to 2000 m³/h water)

3.8. Catalytically accelerated membrane filtration – Practical experience

A homogenous catalyst for improving membrane filtration processes. Are there any results available? Effects of catalytic acceleration of the formation of H₂O-molecular where found on RO membranes on a pilot installation within the Belgium CARVE project (Chemicaliënvrije Afvalwater Recuperatie in der VoEdingsindustrie). CARVE is a project in the field of zero liquid discharge (ZLD). Here purified wastewater was fed to RO (reverse osmosis) via an MBR (membrane bioreactor). Two cycles are operated (Table 5):

Table 5. CARVE-Project – Description of operational mode

<i>Cycle</i>	<i>Description of operational mode</i>
A	Conventional operational mode
B	Conventional operational mode with catalyst (MOL [®] LIK) installed (Installation point: piping between the MBR and RO)

Just a few days after the start of the experiment, noticeable differences between catalysed (MOL[®]LIK-treated) and conventionally treated water are evident. The catalysed water stabilizes the flow rate at a higher level than conventional treatment. At the same time, fewer cleaning cycles have been necessary in the catalysed experimental cycle (Reyniers and Depuydt, 2017). This observation fits the described water model. With the following table the effect of catalytically accelerated formation of H₂O-molecular on 1 µm prefilter units in the area of industrial surface water treatment is shown (Table 6). In addition to the description of the development of the filter units, the following effects have been observed on the downstream RO membranes (Table 7).

That means:

- Improved efficiency of chemicals used

- Permanent securing of the flow rate
- Less fouling issues on the membrane surface
- Minimize cleaning efforts by more than 50 percent
- Enlarged membrane lifetime

Table 6. Performance development on 1 µm prefilters in the area of surface water

Operational mode		Conventional	Catalytic (MOL®LIK)
Flow Rate	[m³/h]	40 - 90	65 - 100
Differential pressure	[bar]	0.1 - 0.3	0.05 - 0.1
Cleaning Interval	[days]	2 - 3	30

Table 7. Performance development on membranes in the area of surface water (Catalytic retrofitted since 2016)

	Design capacity	Conventional	Catalytic (MOL®LIK)
Permeate flow rate	120 m³/h (stable)	Decreases to 80 m³/h (in short times)	120 m³/h (stable)
CIP-Cleanings	4 CIP per year	22 till 28 CIP per year	10 CIP per year
Membranes life time	5 years	2 years	> 3 years

The many years of applications with catalytically accelerated membrane filtration in the field of surface water treatment have shown that such catalytic processes run with stable performance over periods of more than 7 years (Koppe et al., 2017). Based on these facts, the use of catalytically accelerated formation of H₂O-molecular causes an increase in plant efficiency.

Adapted from on these approaches, there is a current European project INSPIRE WATER (INnovative Solutions in the Process Industry for next generation Resource Efficient WATER management). The scope of this project is checking out the extent to which a treatment of industrial wastewater - up to the use as fresh water - can be operated economically (Dambmann and Zorn, 2018).

This shows that the use of suitable catalyst technology can reduce the risk of performance-reduced deposits. With catalytic converter technology and optimized use of chemicals, economic and ecological advantages were found compared to conventional driving.

4. Conclusions

Water stress is a topic that our ecosystem has mastered since its creation. The combination of filtration and membrane technologies has been proven to separate high quality water and salts for millions of years. Humans just started using membrane technologies to provide clean water just a few thousand years ago.

The driving force for this were energetic aspects. Only for 60 years membrane technologies are

available, which can provide high-quality water on a technically relevant scale.

Economic aspects are currently the driving force to make these systems more efficient. For these efforts to be targeted, knowledge about the physico-chemical processes on membranes is essential. By using the catalytically accelerated water treatment, the system performance and thus the profitability of reverse osmosis membranes is increased. At the same time, this reduces the negative impact on the ecosystem by reducing the associated use of chemicals.

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SMART WATER AND SOIL-SALINITY MANAGEMENT IN AGRO-WETLANDS

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Abstract

Soil salinization is becoming worldwide one of the most serious land degradation issues. Seawater intrusion in upper aquifers is responsible for the largest proportion of salt-affected agricultural lands in coastal areas. In this study, the impact of different irrigation strategies on the salinity of a maize cultivated field located in the coastal plain of Ravenna, Italy, was simulated with the FAO AquaCrop model. Model calibration was supported by comparison with remote-sensed and field collected crop data. Ten irrigation scenarios were tested by varying the irrigation season length, the soil moisture threshold for irrigation (TI), and the irrigation depth (ID), in presence or absence of flooded pipe drains (FD) to create a fresh-water lens preventing salt rising from brackish groundwater.

FD show to be more effective in countering soil salinization than strategies exclusively based on supplying enough water to obtain salt leaching (SL). The best result, in terms of both fodder maize yield and salinization control, is achieved with the combination: FD immediately after sowing, irrigation inhibited in May, TI set at 50% of soil readily available water (RAW), and ID modulated to exceed field capacity and obtain SL. The worse strategy is revealed to be the non-FD scenario, coupled with no irrigation in May and August, TI ranging between 65 and 80% of soil RAW depletion, and ID set at 50 mm. Even if water-conservative, this approach results in high soil salinization and leads to significant yield decrease.

Key words: AquaCrop, groundwater, irrigation, maize, soil salinity

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1. Introduction

Salinization is the process by which water-soluble salts accumulate in soils. It is a concern because salt excess diminishes crop yields by limiting root water uptake and causes de-flocculation of clay particles decreasing soil porosity, which, in turn, reduces soil permeability, hydraulic conductivity (Crescimanno et al., 1995; Shainberg et al., 1992; Tsanis et al., 2016) and capacity to support equipment. Salts may be present in the soil since its formation or

accumulated by water transport and evaporation. Tóth et al., (2008) show that salinization in Europe is a diffused problem along coastlines, particularly in the semiarid Mediterranean area. Seawater intrusion in river outlets and contamination of shallow coastal aquifers is actually also a worldwide growing issue due to sea level rise and land subsidence and, depending on site-specific topography and geomorphology, the presence of a saline groundwater determines a more or less intense soil salinization threatening coastal ecosystems. The process is

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jeopardizing soil integrity, water quality, vegetation biodiversity, and agricultural productivity of these habitats in the Mediterranean basin.

The Ravenna coastline in the Emilia-Romagna region, Italy, bordered northwards by Comacchio lagoon and southwards by Cervia town, is an area increasingly prone to the occurrence of the mentioned processes. A natural subsidence (up to 1.5 mm/year), due to the compaction of the alluvial deposits, has dropped this territory below the sea-level, requiring the establishment of an artificial drainage system. An extensive hydraulic infrastructure, composed by ditches, canals and pumps, was built between 1920 and 1960 to keep the land dry by continuously extracting soil water. The drainage, however, lowered the hydraulic head below the sea level and generated a hydraulic gradient promoting seawater inflow into the groundwater. In addition, the intense urbanization occurring in this area between 1950 and 1980, combined with water and methane extraction from the subsoil and phreatic aquifer exploitation for agriculture, have heavily intensified the subsidence rate (Teatini et al., 2006). The consequent saline seepage led to an average rising of the never stable freshwater-saltwater interface towards the ground surface in the low part of the area determining a more or less intense soil salinization (Antonellini et al., 2008; Buscaroli and Zannoni, 2010; Felisa et al., 2013; Greggio et al., 2012).

The measures currently applied to reclaim salt-affected sites are categorized into physical, chemical, biological, and hydraulic ones. This last, based on salt leaching (SL) through irrigation, is considered the main salinity control method in irrigated lands (Abrol et al., 1988; Visconti et al., 2011). During the growing season, SL can be accomplished by applying an amount of water exceeding soil water holding capacity. The additional water percolates downwards, moving salts below the root zone. A subsurface draining network is then required to collect and discharge the drained saline water.

To determine time and amount of water at each SL intervention, several factors must be considered, including: irrigation method and water quality, soil physico-chemical and hydraulic properties, initial and final desired level of soil salinity, plant transpiration requirements and crop tolerance to salinity. As water for irrigation becomes increasingly scarce and agriculture is called to make a more environmentally and economically sustainable use of it, an accurate irrigation scheduling is necessary.

The project LIFE AGROWETLANDS II (LIFE15 ENV/IT/000423), in whose framework this work has been conducted, is intended to counteract soil salinization affecting the coastal farmlands through a targeted and efficient management of the water resources. To this aim, the crop water productivity model AquaCrop (Steduto et al., 2009) has been chosen as instrument for irrigation management, due to its relative simplicity coupled with the possibility of dealing with soil and water salinity.

In comparison with other crop yield models as DSSAT (Jones et al., 2003), CropSyst (Stöckle et al., 2003), APSIM (Keating et al., 2003), Hybrid-Maize (Yang et al., 2004), which require a large number of input parameters and detailed information about crop development, AquaCrop needs a relatively small number of explicit parameters (Vanuytrecht et al., 2014), which can be easily obtained. Moreover, AquaCrop can perform a salt balance within soil profile, accounting for the salt derived from the water capillary rise and from the infiltration of saline irrigation water, and can calculate how much salt leaches out of the rooting zone. In particular, it simulates crop response to individual effects of salinity stress, as poor development of canopy cover and stomata closure, in terms of final biomass reduction.

AquaCrop has already been tested on different crops in multiple climatic and agro-ecological conditions. Given the relevance of maize as warm season irrigated crop in many world areas, only this crop is analysed here. Wang et al., (2015) adopted AquaCrop to investigate yield and water productivity for rainfed summer maize in the North China Plain. Salemi et al., (2011) and Oiganji et al., (2016) calibrated and validated the model for simulating maize growth under deficit irrigation in Northwest Iran and Northern Nigeria, respectively. Ahmadi et al., (2015), Kheir and Hassan (2016), Abedinpour et al., (2012), and Katerji et al., (2013) evaluated the effects of different irrigation techniques compared to rainfed regime, on maize yield in semi-arid areas of Iran, Egypt, Northern India, and Southern Italy, respectively. Only one study, however, was conducted on maize cultivated in saline environment (Saad et al., 2014).

This work addresses the calibration of the AquaCrop model for maize irrigation in the above referred coastal plain and its use to evaluate the effects of different irrigation strategies on salt accumulation. Several soil and plant parameters have been assessed during maize growth at field level and from remote sources, to assist in the calibration process. A first round of simulations is reported here and discussed to elucidate results already acquired, provide suggestions for irrigation strategy and highlight points needing additional effort. Section 2 describes the study area and its environmental conditions, the field surveys carried out to document phenological development and to validate and calibrate simulations. Section 3 describes survey results, the numerical model calibration, irrigation scenarios, model simulation results, and draws suggestions from simulations and observations for a proper irrigation scheduling. Section 4 presents a synthesis of major results.

2. Material and methods

2.1. The study area

The area surveyed within the Project is the coastal plain between the estuaries of Lamone (44°

31.661' N) and Reno (44° 35.432' N) rivers, around 7 km North of the port of Ravenna (Fig. 1). The area is comprised between the Adriatic coastline eastwards and the small town of S. Alberto (RA) westwards. The south-north road connecting today Ravenna to S. Alberto follows the coastal dune relief of the classic Etruscan period (500-300 b.C.); the entire study area is therefore part of the relatively recent deposits and is marked by intersecting ridges along the Etruscan, the medieval (followed by the Romea road connecting Venice to Rome), and the present littoral dunes, combined with riverine levee systems along the present Lamone, Destra Reno (ancient Lamone) and the present Reno (ancient Po di Primaro) river paths. The low lands between ridges were covered by brackish waters until the reclamation carried out in the last two centuries and finished in 1962. North of the Reno river and South of the present Lamone channel, coastal lagoons are still present (Lamberti et al., 2018). The agricultural study area, excluding dunes and levees, is therefore a low one with elevation ranging from -2 to +1 m a.s.l., and suffers from salt intrusion through the under-laying sandy aquifer. The area is artificially drained and can be irrigated with good quality water derived from the Reno river upstream the Voltascirocco barrage, as well as with low quality drainage water of the Destra Reno channel.

A field was selected (Fig.1) to test the model in the Biomarcabò unit of the Agrisfera Cooperative, which is involved as project partner.

2.2. Environmental conditions

The climate in the area is classified as Mediterranean North (Metzger et al., 2005), i.e. featuring a wet winter followed by a long growing season (7 months/yr, Apr. to Oct., of mean temperatures $> 10^{\circ}\text{C}$), but frequent and sometimes prolonged drought periods occurring in the summer time (July most critical), often requiring irrigation. The climate in the area features an average precipitation quite constant around the year (ca. 50 mm/month), whereas potential evapotranspiration is characterized by monthly values below 20 mm/month in Dec. and Jan., and a peak in July around 160 mm/month (Fig. 2).

Soil properties in the area are extremely variable, reflecting the different deposition processes. The field focused in this analysis is placed in the lowest part of the area that was seat for a lagoonal marsh in the 19th century; the soil texture is mainly silty-clayey or clayey. The readily available water (RAW) holding capacity of these soils is normally good, exceeding 100 mm, except for saline clays due to the high-water content at wilting point (WP).

Crops cultivated in the area are: winter cereals (wheat, barley) sown in October and harvested in June, the pluriannual forage lucerne, and summer crops as maize, sorghum, sunflower and soybean, sown in spring and harvested in the summer time. Only maize is regularly irrigated in the area.

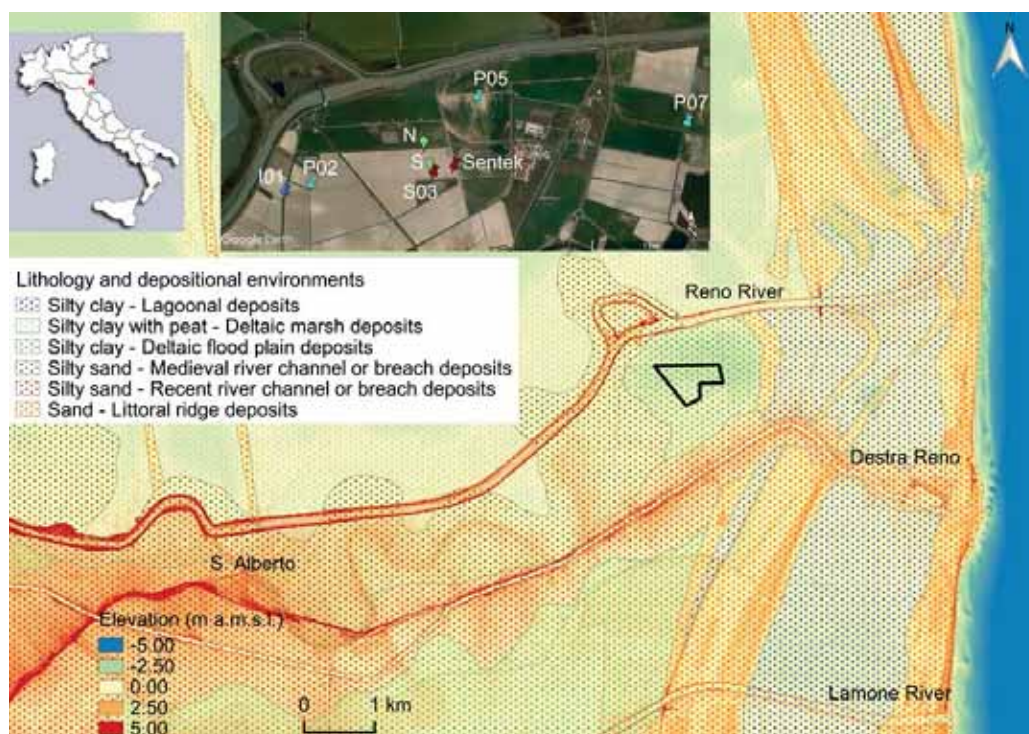


Fig. 1. The study area showing elevation, soil texture and depositional origin. The P02 and P07 marks indicate the meteorological stations provided with piezometers and soil sensors. The S03 and Sentek marks indicate the soil moisture and salinity sensors installed in the experimental field. The P05 mark indicates a further piezometer and soil sensor. N and S marks indicate the survey points in the field. The field involved in this paper is located in the lowest part of the area and its border is pointed out in Figure

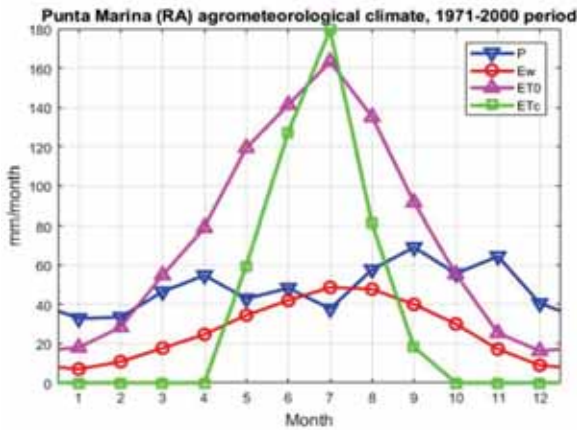


Fig. 2. Climatic conditions of the area

2.3. Groundwater and drainage

The area is artificially drained by two channel networks contained within major levees and ending with pumping stations pouring water into the Destra Reno channel, raising it from -3 m a.s.l to approximately the sea level. Agricultural fields were originally drained by ditches dug at a short spacing (around 25 m), reflecting the low permeability of soils. In order to increase the size of fields, in most part of the area ditches are now substituted by draining pipes, placed usually 90 cm below soil surface at 10 m spacing, with slope 1:1000 towards the drainage channel.

The drainage network depresses the water table well below sea level. Drainage pipes actually separate the upper agricultural soil, where water exceeding field capacity can percolate down draining salt and soluble nutrients, from lower layers, where saline or brackish water is normally present. In dry periods and non-irrigated fields, salt moves upwards by capillary rise and is concentrated at the soil surface in some areas.

Groundwater level and salinity were monitored in eight piezometers (denoted Pnn) of which three are visible in Fig. 1 (see also Cipolla et al., 2019); soil water content and bulk conductivity were monitored at 50 cm depth at piezometer stations and three further soil stations (e.g., S03 in Fig. 1). In the field object of this study, a temporary Sentek sensor measuring soil moisture and salinity down to 60 cm depth with 10 cm resolution was placed around start-up of the irrigation period to provide better information on water percolation in the soil (Fig. 3).

2.4. Maize cultivation

The experimental field was managed according to the principles of organic farming which are adopted in the specific unit of the Cooperative. The maize hybrid Krups (FAO 600), supplied by SIS (S. Lazzaro, Bologna, Italy), was sown on May 10, 2018, with a seed spacing of 15 cm on the row and 75 cm between rows. An average crop density of 7.8 plants m⁻² was achieved after seedling emergence.

Soil was ploughed at 35 cm depth, harrowed and fertilized with the liquid fraction of digestate from cattle slurry before sowing. A second dose of liquid digestate was supplied during vegetative growth.

The crop was harvested on Aug 11, 2018, at the early dough stage. The whole plant was cut and chopped to produce maize silage.

The amount of water, supplied during maize growth by sprinkler irrigation operated with travelling gun, was determined by the farmer with the IRRINET software (<http://www.consorziocer.it>), a decision support system widely used in the area. This tool operating on an online platform calculates the real time water budget based on a crop growth model coupled with precipitation, the Hargreaves ET₀ (Hargreaves and Samani, 1985), and potentially the contribution of the shallow water table. Soil salinity is not accounted for.

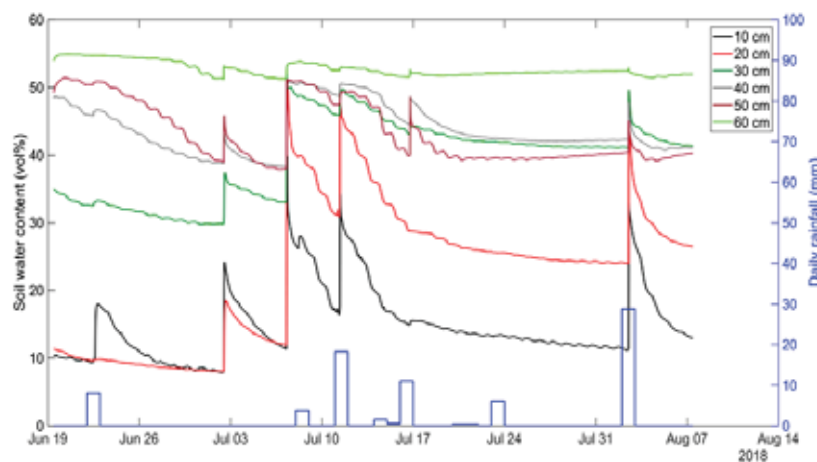


Fig. 3. Soil water content at different depths and rainfall recorded at stations Sentek and P02; with increasing depth average humidity as a consequence of rainfall or irrigation events normally increases while its time variation decreases; exceptions are present 1) due to cracks in the clayey soil, through which water can percolate to the deeper layers without increasing humidity of upper layers, and 2) superficial irrigation and rainfall in July are absorbed in the upper layers generating an inversion of the usual humidity gradient: in July humidity of the 40-50 cm deep layer is lower than in the 20-40 cm deep layers

2.5. Meteorological data

The climatic parameters fed to the AquaCrop model are: daily maximum and minimum air temperature ($^{\circ}\text{C}$), relative humidity (%), average wind speed (km day^{-1}), precipitation (mm), global solar radiation ($\text{MJ m}^{-2} \text{day}^{-1}$) and average annual value of atmospheric CO_2 concentration. These data were derived from the measurements of the P02 meteorological station located near the field (Fig. 1). Missing data were replaced with data from the near P07 meteorological station.

The CO_2 concentration data, not recorded by the installed stations, were obtained from the Italian Air Force meteorological station (<http://www.meteoam.it/pubpage/3/9>) at Monte Cimone. A model integrated tool calculates the reference evapotranspiration ET_0 (mm day^{-1}) with the FAO Penman–Monteith equation (Allen et al., 1998).

2.6. Field surveys

Crop data. In order to verify information provided by the seed supplier, document crop development and check satellite information, a field survey of crop phenology, canopy development, root density distribution and biomass accumulation over time was carried out from May 9 to August 30 with weekly observations. Several crops were surveyed including maize in several fields. In the experimental field object of the present analysis, two monitoring points were established, one at the North and the other at the South side of the field (Fig. 1). Collected information include:

1. The principal maize growth stages were identified following the BBCH scale (Weber and Bleiholder, 1990; Lancashire et al., 1991) criteria. To achieve that, the plant height, number of leaves and nodes, start and end of the flowering period and fruit development were measured and recorded on a sample of 20 plants in each point. Average value was then calculated for each item.

2. Canopy Cover (CC, %) was derived from digital images produced by mobile devices in the field and subsequently processed using the Canopeo app for Matlab developed by Patrignani and Ochsner (2015). Ten photos were taken at each sampling point, five on the row and five on the inter-row. Average values were then calculated. Leaf area index (LAI) and Normalized Difference Vegetation index (NDVI) were also assessed, the first by sampling five random plants and measuring leave surface, the second systematically using the instrument GreenSeeker (Trimble Ag Field Solutions, Sunnyvale, CA, USA).

3. Plant biomass was measured with a 3-week interval, starting from the stem elongation phase. The plants on two adjacent rows per 2 m length were cut from the base and weighed with a dynamometer to determine the fresh weight (kg m^{-2}). A sub-sample was oven-dried at 105°C to determine the dry weight (kg m^{-2}).

4. Root density was measured at flowering end, when root growth is considered completed. Root samples were taken by coring the soil with a one-meter deep steel cylinder, 8 cm in diameter. The extracted soil column was divided in 20 cm long portions. Soil was washed-out and the remaining roots were weighed with a precision scale. Root volume density was calculated according with Wu et al., (2017).

5. The field received a total 135 mm irrigation in three interventions (June 20, July 10 and 19; exact dates slightly vary due to sprinkler displacement to cover the large field).

2.7. Model criteria

AquaCrop model version 6 was used. It was tested first by comparison with observed data, and then used to simulate irrigation management scenarios. For model calibration the following data were used.

- a. Crop data. The crop module was implemented in Growing Degree Days (GDD). The default values provided by the FAO AquaCrop model for the maize crop parameters were modified according to the specific characteristics of the hybrid used. The GDD data provided by the seed supplier were used for this purpose. FAO default stress-coefficients were left unchanged.

- b. Field management. Soil fertility and weed management were set as optimal in the model. The soil Curve Number (CN) based on soil textural class was reduced by 5% to account for soil tillage effect on soil permeability and water infiltration rate.

- c. Soil characteristics. Soil moisture and salinity were monitored throughout the crop growing season using two sensors (GS3-Meter and Sentek) located at the South and East side of the field (Fig. 1). Four soil layers of variable depth were considered in the model: 0-10, 10-30, 30-60, 60-120 cm depth (Table 1). Soil hydraulic properties, water content at saturation (SAT), field capacity (FC), permanent wilting point (PWP) and saturated hydraulic conductivity (K_{sat}), were calculated by means of the Soil Water Characteristic software (Saxton and Rawls, 2006) based on soil texture, compaction, organic matter and salinity. Calculated parameters were further adjusted based on the Sentek moisture readings at 10 cm depth intervals, to better represent tillage driven porosity variation along depth and its influence on water infiltration and holding capacity.

- d. Groundwater. Groundwater level and salinity were monitored by means of the P02 and P05 piezometers installed in the pilot area close to the experimental field (Fig. 1). Groundwater level and salinity observed at P02 were therefore imposed to this module. Starting from June 19, the above described network of plastic pipes located below field surface was flooded with fresh water by raising water level in the discharging ditch at the north boundary of the field; however, the fresh water lens due to pipe slope did not extend to the whole field, but only to around

half the field, i.e. the northern part near the ditch. Owing to this, the following two conditions were represented in simulations as groundwater boundary conditions:

- P02 observed groundwater level and salinity (brackish groundwater, BG);
- constant fresh water lens of good quality at 80 cm depth (flooded drains, FD).

e. Initial conditions. The simulation was run starting on April 1st using: 1) for water content and salinity of the 3rd layer, values obtained from records of the GS03 sensor installed in the field, and 2) a regular gradient of water content and salinity along the vertical. The 40-day long simulation before actual sowing (May 10) reduces the effect of the initial gradient once the relevant simulation phase starts.

f. Irrigation. Irrigation water quality was set as good, according to the results of the monitoring done during the growing season. The real irrigation events are reproduced in the calibration phase.

3. Results and discussion

3.1. Crop development

The development of the crop followed almost exactly the anticipated schedule based on the pattern indicated by the seed supplier. Ground observed CC

and NDVI values matched quite well values obtained from Sentinel 2 and Landsat 8 satellite observations, respectively with 10 and 30 m resolution. Until flowering, NDVI and CC showed practically equal numerical values, whereas after flowering both remained almost constant and therefore could not describe the final reproductive and crop ripening phase.

Fig. 4 presents NDVI values at the two survey points in the field. Growth in space and time was fairly consistent; the spatial distribution at the end of the vegetative stage can be observed from the NDVI satellite image at June 30 (just before flowering) presented in Fig. 5a.

As a partial conclusion, the space-time pattern of the crop vegetative development is coherently described by ground and satellite observations of the NDVI parameter.

3.2. Final crop yield

At harvesting time, a big cutter-shredder machine was used equipped with a crop mass and humidity measuring system and GPS positioning, so that the detailed yield map for the field was produced that is shown in Fig. 5b. Point measurements and the continuous map obtained by kriging are shown in the Figure.

Table 1. Physical and hydraulic properties at increasing soil depths at the Biomarcabò field

Horizon	Depth (cm)	Textural Class	SAT (% vol)	FC (% vol)	PWP (% vol)	K_{sat} ($mm \cdot day^{-1}$)	Compaction
1	0-10	Silty clay	58.5	41.5	27.0	283	loose
2	10-30	Silty clay	58.5	41.5	27.0	283	loose
3	30-60	Silty clay	53.9	40.6	26.9	138	normal
4	60-120	Silty clay	47.0	39.2	26.8	31	dense-hard

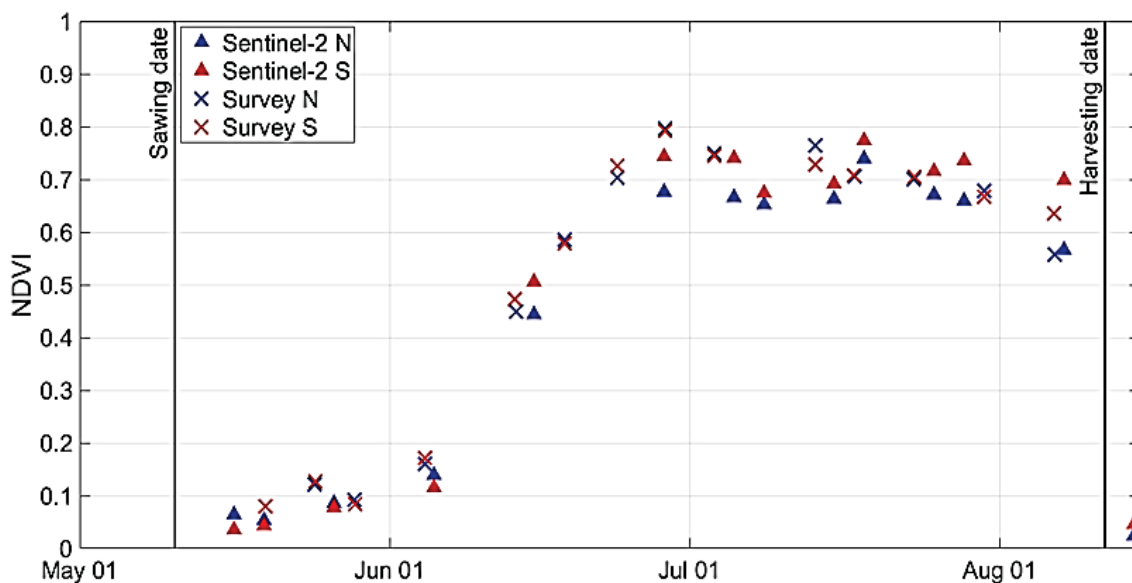


Fig. 4. Time evolution of NDVI at the survey points according to different sources: ground observations at positions N/S, and Sentinel 2 observations at the same positions, obtained from the EO Data Service (<http://www.eodataservice.org/>).

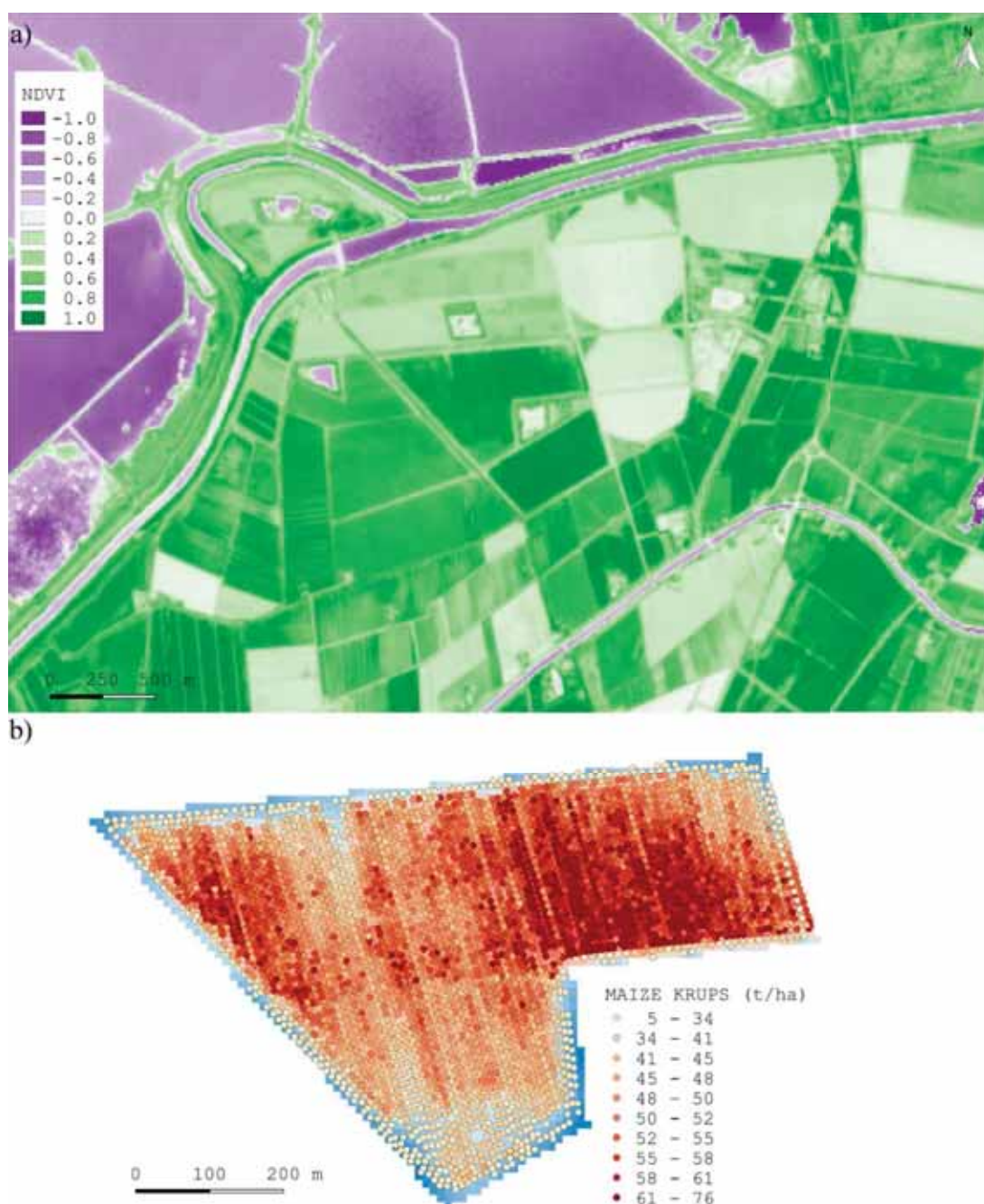


Fig. 5. NDVI distribution over the area on June 30, 2018, i.e. after the first irrigation and just before flowering-a).
Yield map at harvesting time in the surveyed field -b)

Some features of maps in Fig. 5 are common, and in fact their correlation coefficient is high (0.80), showing that most processes that limit yield in the field are not varying in time and are presumably related to soil conditions.

3.3. Model calibration

The agreement between model results and field observations was already good with the standard parameters. Only a minor adjustment to initial leaf area was necessary to catch the initial growth speed of

the crop. Fig. 6a reports the results of a preliminary simulation in the calibration phase. Based on field observations, the calibrated parameters initial leaf area and initial root depth were finally assumed equal to 8 cm²/plant and 25 cm. Model results can be compared with observations as far as canopy cover and dry crop mass per unit surface are concerned; Fig. 6b shows the excellent results obtained.

The model, therefore, can be considered successfully calibrated, so that reliable conclusions can be drawn from the simulation of different irrigation strategies.

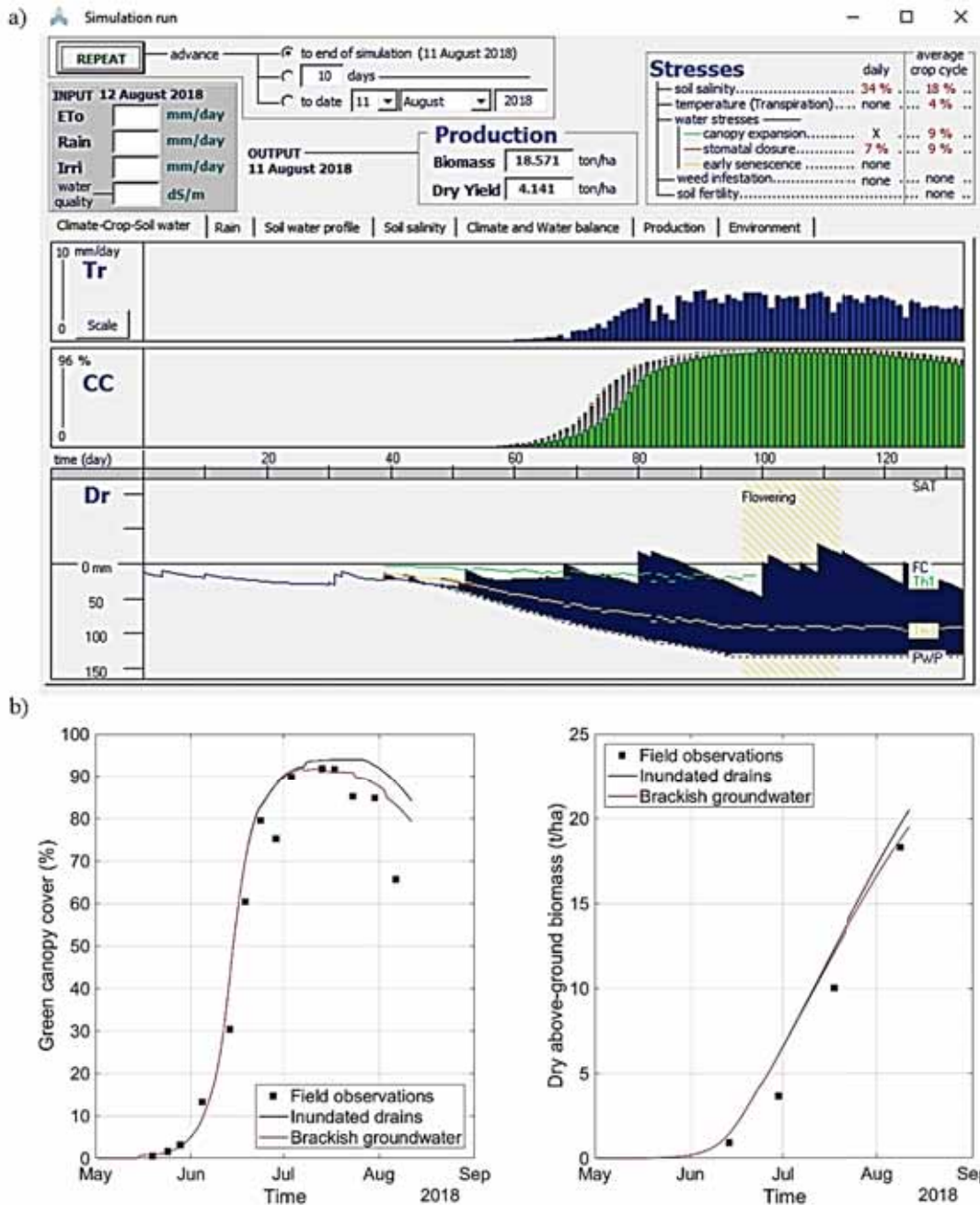


Fig. 6. Time evolution of soil water content during a preliminary simulation in calibration phase, with the standard initial leaf area of 6.50 cm²/plant and 15 cm initial root depth, AquaCrop graphical output; the combination of a rather dry May with the adopted parameters results in a delayed simulated crop development (the green CC graph) compared to the optimal one (the grey one)-a). Comparison between final calibrated model results and field observations at survey point N (Fig. 1) for the two parameters that are simulated and observed-b)

3.4. Implemented irrigation criteria

The irrigation module was set on “generation of an irrigation schedule” mode, and the sprinkler irrigation method, wetting 100% of the soil surface, was selected. The model was asked to establish an irrigation event whenever the Readily Available Water (RAW) was depleted below a given level M_{min} while the irrigation volume is set indirectly fixing an upper level M_{max} to be reached, which is not far from field capacity. The two levels may vary in time to

follow crop sensitivity to water stress during its development. Due to practical reasons and following regional suggestions, that aim at preserving the water resource, a maximum 50 mm per irrigation event is normally proposed. Since this amount is significantly lower than RAW in soils of the area, irrigation may be scheduled keeping soil humidity around the lower RAW limit in order to reduce leaching due to unforeseen rainfall events, or around the upper limit aiming to produce some leaching; the following criteria are in fact tested.

The 1st criterion is similar to the standard suggested for Maize (<http://www.fao.org/land-water/databases-and-software/crop-information/maize/en/>) by FAO Land & Water: “to obtain a good stand and rapid root development the root zone should be wetted at or soon after sowing”; the acceptable water depletion level is about 40% in the establishment period, between 55 and 65% during the vegetative, flowering and yield formation periods, and 80% during the ripening period. Here and in what follows, percent figures refer always to RAW depletion level, as used in AquaCrop.

In the surveyed Region, irrigation is usually discouraged until June since in April and May soil water content accumulated in winter time is usually sufficient; therefore a 2nd criterion is simulated representing the local irrigation strategy. Irrigation is proscribed until June; M_{min} is then set at 80% of RAW depletion; starting from stem elongation, M_{min} is gradually raised to reach 65% at flowering start, when the maize plants are most sensitive to water stress. From the flowering end, M_{min} is gradually lowered attaining the initial 80% at the end of the ripening phase, i.e. at the dough stage, and irrigation is inhibited in August, since harvesting in the given circumstances (sowing date, seed hybrid and accumulated GDD) is scheduled no later than August 15. Irrigation depth is fixed at 50 mm (M_{max} adapted to obtain this depth). This criterion results in a rather

water conservative one, based on most common conditions in the region.

In the 3rd criterion, M_{min} is kept constant at 50% of RAW and irrigation volume fixed as the amount restoring water content at field capacity (M_{max}). This criterion aims at maintaining yield potential at the expense of some inefficient water utilisation when a heavy rainfall occurs just after an irrigation event; in any case, since FC is not intentionally exceeded, leaching is not intentionally produced.

Since none of the previous criteria represents an intentional leaching strategy, a further criterion, the 4th, is simulated where M_{min} is fixed at 40% and irrigation amount is controlled to produce a 5 mm excess above FC. Finally, since several of the previous irrigation criteria cause a number of irrigation events greater than usual and might result non sustainable from an economic point of view, a 5th scenario is introduced where irrigation is inhibited in May and August, the lower and upper levels for irrigation are set at 50% of RAW and FC+10 mm respectively.

Actual irrigation and the 2nd criterion result in salinization prone strategies, whereas all the others criteria aim at controlling salt accumulation in soil. All irrigation criteria are simulated under BG conditions as well as under FD conditions. The main results obtained by AquaCrop (Fig. 7 refers for example to the 1st criterion with brackish groundwater) are presented in Table 2.

Table 2. Synthesis of results of the tested scenarios for the simulation period April 1 – August 11, 2018.

<i>Irrigation criterion</i>	<i>Crop yield, dry above-ground biomass (t/ha)</i>	<i>Irrigation depth (mm) and events</i>	<i>Final salt content in the root zone (t/ha)</i>	<i>Crop transpiration (mm)</i>	<i>Runoff (mm)</i>
2018, FD	20.524 (94%)	135 (3)	8.436	266.4	6.7
2018, BG	19.524 (90%)	135 (3)	17.404	253.4	6.3
1, FD	21.050 (97%)	134.7 (4)	8.440	273.1	4.0
1, BG	20.032 (92%)	123.8 (3)	17.398	259.9	7.0
2, FD	20.508 (94%)	147.8 (4)	8.411	266.2	4.1
2, BG	19.513 (90%)	135.2 (3)	17.400	253.3	7.3
3, FD	21.053 (97%)	152.7 (4)	8.540	273.1	2.4
3, BG	20.029 (92%)	168.3 (4)	17.550	259.9	4.6
4, FD	21.061 (97%)	184.4 (5)	8.680	273.2	7.0
4, BG	20.025 (92%)	154.9 (4)	17.506	259.8	2.5
5, FD	20.524 (94%)	155.5 (3)	8.501	266.4	2.9
5, BG	19.524 (90%)	105.7 (2)	17.348	253.4	3.1
5, FD since 11/5	21.179 (97%)	154.1 (3)	7.620	274.9	2.6

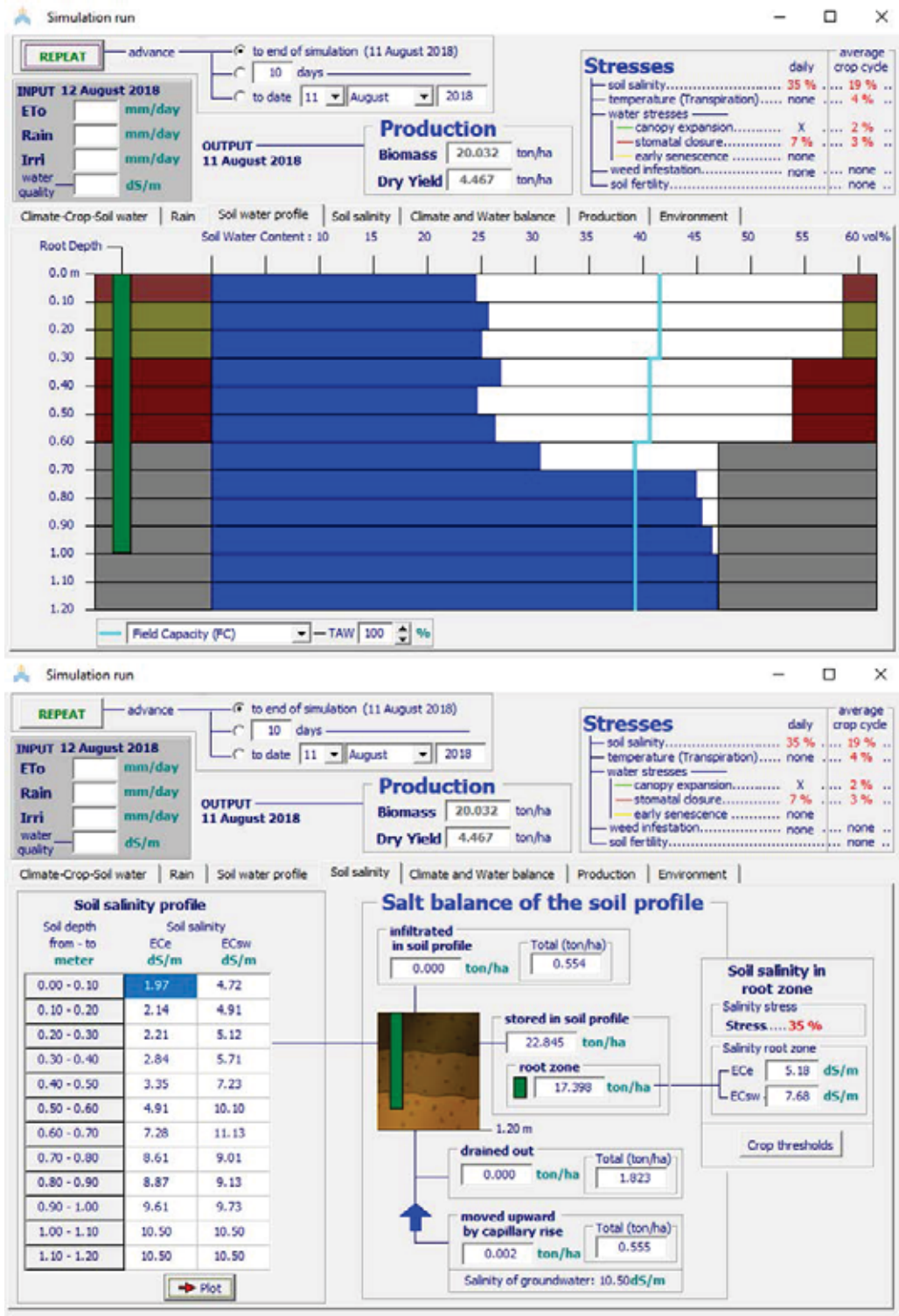


Fig. 7. Vertical distribution of water and salt content in the soil at harvesting time for scenario 1 with brackish groundwater, AquaCrop graphical output.

Crop yield, i.e. dry above-ground biomass, is presented also as percentage of the potential yield under no stress conditions (21.763 t/ha). The salt content at sowing time within the final root depth is around 9.0 t/ha; therefore, all the FD scenarios represent a moderate reduction of salt content in soil, whereas all BG scenarios represent a significant increase of soil salt content.

3.5. Effects of irrigation strategy on crop yield and soil salinization

It must be focused that the 2018 summer season was not an arid one: 190.1 mm rainfall occurred in the simulation period, most of which in June and July. Irrigation criteria are all based on the current soil water content and no rainfall forecast is accounted for. Therefore, when simulation results are

analysed the reader must be aware of 1) the weak relevance of irrigation in the period, and 2) the uncontrolled occurrence of rainfall after irrigation events, a circumstance that introduces some random behaviour, obscuring the systematic average effect of the criterion.

In any case, a major systematic effect is due to inundated drains. It decreases significantly the accumulation of salt in the agricultural soil originating from groundwater by capillary rise under a water content gradient, and the consequent salinity stress. Crop yield and transpiration are increased, and this forced sometimes an additional irrigation event (40-50 mm) by the end of July, producing a weak benefit since a significant rainfall event occurred at the beginning of August.

The best results, both for crop yield and salinization control, were obtained by inundating drains just after sowing. The depth-time evolution of soil water and salinity content during the growing season is presented in Fig. 8. The general behaviour of salinization prone scenarios is represented in Fig. 8a-b, where salt rising is apparent in absence of flooded drains. All the other scenarios are qualitatively represented in Fig. 8c-d, where the earliest drain inundation scenario (5th - FD since 11/5) and one of the most water consuming scenarios (3rd - BG) over brackish groundwater are represented.

The former is able to contrast salt rising-up, whereas the latter does not involve a sufficient amount of irrigation water to produce a complete soil leaching; percolation is limited to the first superficial layers and salt is not removed from the whole soil column.

3.6. Discussion and suggestions for irrigation scheduling

In the absence of inundated drains, the agricultural soil is dried up in late spring and summer and salt rises regularly towards the surface. In summertime, irrigation and rainfall events do not usually compensate the prevailing transpiration, but cause fluctuations of water content in the shallow layers. This is what was observed by the Sentek sensor, placed in the upper part of the field beyond the limits of inundated drains action. When and where drains are inundated (in 2018 drains were inundated at the first irrigation time), capillary rise, that is particularly active where humidity gradient is intense, draws fresh water up, whereas brackish water is not subject to relevant humidity gradient and does not rise. If drains are inundated before the formation of the drying front in the soil, i.e. in May, salt accumulation in soil is almost completely avoided.

The irrigation practice in the area and the regional government suggestions are conditioned by experience in the prevalent environmental conditions, where groundwater is several meters below the soil surface and soil surface has a non-vanishing slope. Therefore, water percolating from the top soil is assumed to be lost for agriculture and irrigation is not practiced by flooding that involves low energy consumption (less than 1 m head drop), need of manpower and water use efficiency (50%), but mainly with sprinkler that requires energy (several 10 m head) and significant amount of manpower to obtain a high water use efficiency.

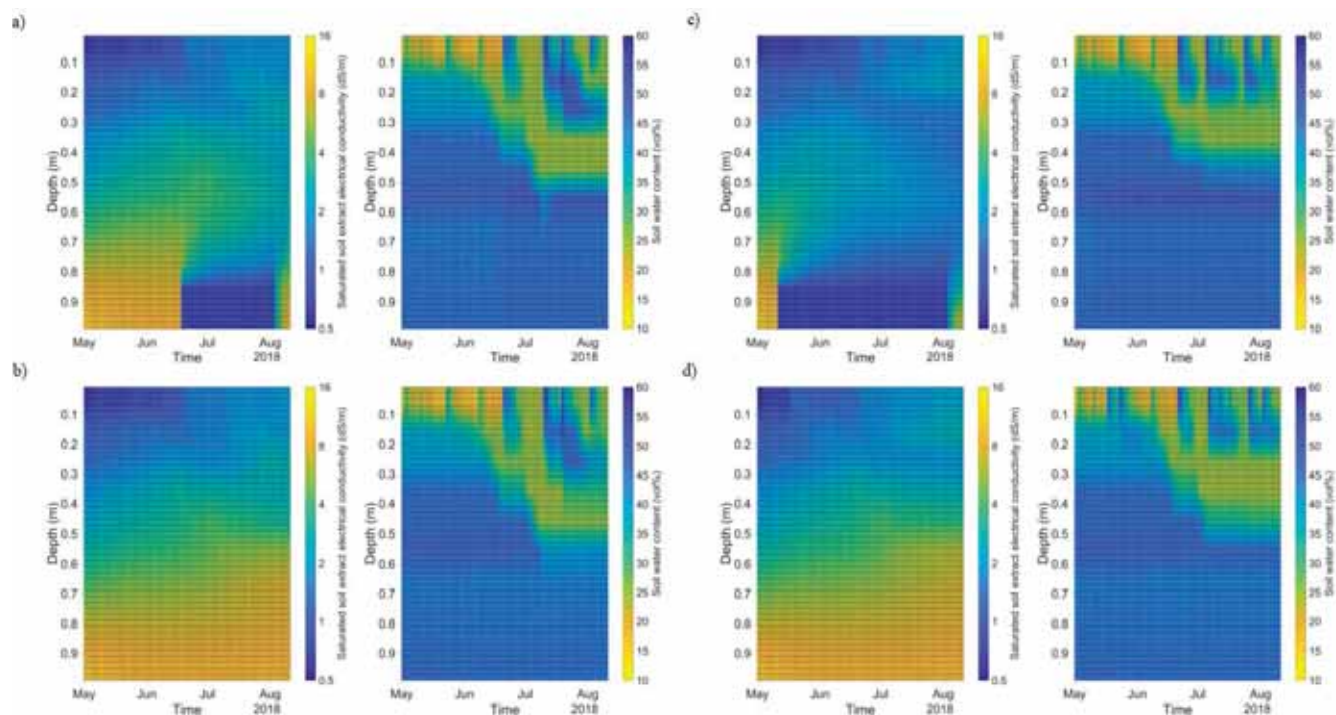


Fig. 8. Depth-time distribution of salinity and water content in the case of inundated drains (a) and brackish groundwater (b) considering the actual 2018 irrigation scenario. Depth-time distribution of salinity and water content in the case of irrigation scenario 5 with inundated drains just after sowing (c) and irrigation scenario 3 with brackish groundwater (d)

This water saving strategy is not justified in the lowest areas of the basin, where water flowing in rivers if not used is poured into the sea, or if flowing in the drainage channels and not used must be pumped up to the sea level.

Drainage pipes are very useful to keep the soil dry without sub-dividing large agricultural areas in many small fields divided by ditches, but can also be profitably used for subsoil irrigation preventing salt rising from brackish groundwater. Some minor adaptation can be suggested for the double use as reducing the slope (from 1 m/km to 0.5 m/km) and/or placing pipes with a double slope from the field centre to the surrounding ditches, so that with 0.1 m submergence at drain outlet the served length of the field may increase from 100 m to 200 or 400 m.

4. Conclusions

Field survey and satellite images provide an adequate description of crop growth and development.

AquaCrop is a reliable model to anticipate crop development and yield also in areas exposed to salinization, as well as to simulate effects of irrigation strategy on salt accumulation in soils.

Subsoil pipe drainage was introduced in the area to extend field surfaces and use large capacity field equipment; it has shown to be also an efficient tool to contrast soil salinization where a humid season, as winter is in the study area, causes some natural leaching.

The utilization of subsoil drains for irrigation generates a fresh water lens quite effective as a barrier against capillary rise of brackish water and salt accumulation in the agricultural soil.

The survey year (2018) was not an arid one, therefore crop yield and irrigation resulted weakly sensitive to the adopted irrigation strategy.

All the tested leaching strategies were not completely effective in the simulated period (spring and summer), and presumably need a humid climate for the remaining seasons to remove salt in a year cycle.

The irrigation practice most diffused in the Emilia-Romagna region is probably not appropriate for the lowest areas near the sea, where reducing the consumption of fresh flowing water, when available, is nonsense.

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COMBINED ASSESSMENT OF CHEMICAL AND ECOTOXICOLOGICAL DATA FOR THE MANAGEMENT OF CONTAMINATED MARINE SEDIMENTS

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Abstract

Sediments in coastal areas can accumulate a variety of contaminants, acting as both carriers and long-term secondary sources of contamination for aquatic ecosystems. Nowadays, there is a growing interest on developing new assessment criteria of sediment ecological quality for setting priorities and management strategies of contaminated materials. According to the literature, the weight of evidence (WOE) approach has been developed to provide a multidisciplinary characterization which combines different studies such as chemical analyses, laboratory and field-based studies to assess the bioavailability of pollutants and ecotoxicological assays. However, applications on complex case studies are limited. In order to strengthen the current literature, this study presents the first results of the application of a WOE model (Sediqualsoft) to a marine chronically polluted area. To this aim, a laboratory experimental investigation was carried out on the polluted sediments of the Mar Piccolo in Taranto (Southern Italy). The combination of chemical and ecotoxicological data confirmed the results obtained with the conventional approaches, highlighting sediment contamination. Even more, the obtained biological responses highlighted an unexpected toxic effect not revealed by conventional approaches: the level of contamination did not seem to be proportional to the ecotoxicological assessment. All these observations have raised numerous questions about the potential mobility of pollutants and additional risks to the environment. The Sediqualsoft model has proven to be a useful tool for processing complex scientific data, playing an important role in contaminated sediment risk assessment supporting stakeholders and decision makers.

Keywords: chemical/ecotoxicological data, contaminated sediments, Mar Piccolo, Sediqualsoft

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1. Introduction

Marine sediments can accumulate a variety of heavy metals (i.e. As, Cr, Hg, Pb, Zn) as well as organic pollutants, including tributyltin (TBT), polychlorinated biphenyls (PCB), pesticides and polycyclic aromatic hydrocarbons (PAHs), acting as both carriers and long-term secondary sources of contamination to aquatic ecosystems (De Gisi et al., 2017a; Lofrano et al., 2016). Contaminated sediments pose major concerns for human health and the environment, because the pollutants can re-enter the overlying water and become available to benthic

organisms and subsequently pass into aquatic food chains (Todaro et al., 2018).

The assessment of contamination level of marine sediments is usually done by comparing the chemical concentrations of samples to reference values (e.g. law limits). However, this approach does not account that contaminant behavior (i.e. remobilize or desorption) in sediments is a dynamic process and bioavailability of contaminants is regulated by physical, chemical and biological properties of marine environment (e.g. Eh, pH).

Chemical analyses alone do not necessarily reflect the bioavailability and the toxic action of

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measured contaminants; multidisciplinary approaches are required to assess the chemical, biological and toxicological impact of complex mixtures of contaminants. In this respect, ecotoxicological bioassays are important complementary tools to evaluate synergistic effects of contaminant mixtures in sediments. Therefore, chemical analyses and ecotoxicological bioassays are important components to assess the environmental quality of contaminated marine areas and to establish the link between contaminants and ecological responses. In this respect, there is a growing interest on developing new assessment criteria of sediment ecological quality for setting priorities and management strategies of contaminated materials. Different sediment quality guidelines (SQGs) have since been developed for use in assessing sediment quality, meaning contaminant concentrations that cause adverse effects (Christopher et al., 2002). As shown in Tables 1, there are several sediment quality guidelines (SQGs) in use today. Additional information on each guideline are available in the papers reported in the references.

In particular, the Weight Of Evidence (WOE) approach has been developed to provide a multidisciplinary characterization which combine different studies, or Lines Of Evidence (LOEs) (i.e. chemical analyses, laboratory and field-based studies to assess the bioavailability of pollutants and ecotoxicological assays) (Benedetti et al., 2012; Chapman, 2007; Dagnino et al., 2008). Chemical and biological data can be elaborated within a quantitative model (Sediquasoft) which combines various typologies of studies, including sediment chemistry, ecotoxicological bioassays, bioaccumulation and biomarker results. WOE methods are often key components for evaluate and classify the ecological status of water bodies (WFD, 2000) and for Ecological Risk Assessment (ERA) (Bebianno et al., 2015).

The aim of the study was the application of Sediquasoft model, based on the combination of

chemical and ecotoxicological data, to assess the environmental quality in a marine chronically polluted area. For the purpose, thirty-eight sediment samples (taken from 19 boreholes) were tested in the laboratory, and the chemical characterization was integrated with acutely toxic effects by Sediquasoft. As a consequence, the following subgoals have been investigated: (i) to examine the spatial variation of heavy metals and organic compounds contaminants, (ii) to assess the contamination and the sediment quality with multidisciplinary approach and (iii) to investigate the link between contaminants and ecological responses.

In order to apply Sediquasoft, the case study of the Mar Piccolo in Taranto (Southern Italy) was addressed (Fig. 1). The Mar Piccolo is an inner sea located on the North of the city of Taranto (south of Italy), with a surface area of 20.72 km². Several researchers (Cardellicchio et al., 2007; De Gisi et al., 2017; Di Leo et al., 2014; Mali et al., 2017; Petronio et al., 2012; Vitone et al., 2016) have shown that also the submarine sediments in the Mar Piccolo contain high concentrations of heavy metals (Hg, Pb, Cd, Cu and Zn) and organic pollutants (PCBs and PAHs). The Mar Piccolo basin is part of a large area that has been declared “at high risk of environmental crisis” and included into the list of the Sites of National Interest (SIN Site) (Italian Law, 1998).

2. Material and methods

2.1. Sediment chemical analyses

Sediment samples were collected to depths of about 3.0 m from the seafloor, transported to the laboratory at 4 °C and stored for subsequent use for bioassays and for the determination of contaminant concentrations. In particular, chemical analyses to determine the concentrations of metals, PAHs and PCBs in the sediments were carried out.

Table 1. Types of SQGs in use

<i>SQG Category</i>	<i>Approach</i>	<i>References</i>
Theoretical	Equilibrium Partitioning	Di Toro and McGrath (2000)
	Spiked Sediment Toxicity Test	Simpson et al., (2004)
	Porewater Effect Concentration	Hübner et al., (2009)
	Equilibrium Partitioning	Yun-Zeng et al., (2007)
	Acid Volatile Sulfides	De Jonge et al., (2009)
Empirical	Tissue Residue Approach	Meador (2006)
	Screening-Level Concentration	Von Stackelberg and Menzie (2002)
	Effects Range-Low and Effects Range-Median	US EPA (1997)
	Threshold-Effects Level and Probable-Effects Level	US EPA (1997)
	Apparent-Effects Threshold	Cabbage et al., (1997)
	Consensus-Based Evaluation	MacDonald, Ingersoll et al., (2000)
	Logistic Regression Modeling	Field et al., (2002)



Fig. 1. (a) Gulf of Taranto; (b) location of the samples taken within the First Bay of Mar Piccolo

The pH, Eh and conductivity were measured with electrodes in a Multi-Liner instrument (CyberScan PC5000, Eutech Instruments). The grain size distribution of the sediments has been estimated through sieving for particles larger than 75 μm (i.e. retained on the N. 200 sieve) and by sedimentation process (i.e. hydrometer analysis) for the finer ones. The grain size distribution was then obtained either by including or excluding the gravel fraction, mainly represented by mussels, shells and fossils. In the latter case, the weight of the material passing the 1 mm (No. 18) sieve was set to be equal to the total soil weight. Organic content was determined from each sediment samples as the percentage of weight loss by drying at 100 °C for 24 h and combustion at 450 °C for 5 h. Total metal content was determined in the < 63 μm fraction after wet digestion with HNO_3 and ultrapure water. The concentrations of metals were obtained by ICP-OES, Inductively Coupled Plasma - Optical Emission Spectrometry (iCAP 7000 Series - Thermo Scientific). Polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) in sediments were determined by a Gas Chromatograph - Mass Spectrometer (GC-MS, Thermo Scientific TRACE 1300).

2. 2. Sediment toxicity bioassays

Toxicity bioassays were performed according to standardized protocols, in terms of exposure conditions, matrix and biological endpoint, with bioindicator species that are widely applied and recognized to assess potential ecotoxicological effects to marine biota. Selected species were the bacterium *Vibrio fischeri*, the algae *Phaeodactylum tricorutum*, and the shellfish *Mytilus galloprovincialis*. These were selected considering phylogenetic diversity and within the most widespread organisms already used in the scientific literature for marine sediments monitoring (Simpson et al., 2017) as well as required by Italian legislations (MD, 2016).

The sediment samples were mixed in a 1:4 (v/v) ratio of dry material to seawater and placed on a jar test for 24 h, at a speed of 200 rpm. After mixing the samples were centrifuged at 3800 rpm for 15 min,

to separate the water from the sediment. The aqueous fraction (elutriate sample) was poured off and stored at 20 °C for use. Prior to use, seawater was filtered (0.45 μm membrane filters), analysed and stored in the refrigerator at +4 °C.

The acute toxicity tests on the sediments were performed according to the ISO 11348-3 (2007). This bioassay exposes luminescent bacteria (*V. fischeri*) to aqueous samples and measures the increase or decrease in light output by the test organisms. Bioluminescence at 15 °C was measured with a Microtox luminescence meter after 30 min of incubation.

Toxicity tests with *P. tricorutum* were carried out according to the ISO 10253 (2016) determining the growth inhibition (or biostimulation) of microalgae exposed to sediment elutriate samples. The algal culture was exposed for 72 ± 2 h at 20 ± 1 °C and 6000–10,000 lux and cellular density was evaluated by spectrophotometer. Toxicity and stimulation effect data were determined as percentages of growth inhibition or growth stimulation (biostimulation) in the observed population.

The embryotoxicity test with *Mytilus galloprovincialis* was performed according to ASTM (2004). Thermal shock technique (18 ± 1 °C and 28 ± 1 °C) was then employed to induce the mussels to spawn. At the end of the spawning, gametes derived from a batch of three males and three females have been removed by filtration at 32 μm (sperm cells) and 100 μm (eggs) to remove impurities. Eggs suspended in 500 mL seawater were fertilized and fertilization success was qualitatively checked by microscopy. At the end of the test, fertilized eggs were fixed in a 4% NBF (Neutral Buffered Formalin) and 100 larvae were observed with a microscope to verify the presence of abnormalities (e.g. malformed or damaged shell). Toxicity effect data were calculated as the ration of the number of retarded and malformed larvae to the 100 counts (percentages of abnormal larvae).

Toxicity tests were performed in triplicate and negative (test without contaminants) and positive (test with reference toxicant) controls were included in each experiment (ASTM, 2004).

2.3. Weight of evidence elaboration

Data on sediment chemistry and toxicity bioassays measured were elaborated within the quantitative WOE model, Sediqualsoft. Conceptual elaborations of the model, whole calculations, detailed flow-charts, rationale for weights, thresholds and expert judgments have been fully given elsewhere (Benedetti et al., 2012, 2014; Piva et al., 2011; Regoli et al., 2014).

The evaluation of hazard from sediment chemistry is based on the calculation for each pollutant of Ratio to Reference (RTR), i.e., the ratio between measured concentrations and those indicated by various SQGs; depending to the typology of chemicals (i.e. “non-priority”, “priority” or “priority and hazardous” pollutant according to EC Directive 2008/105) this value is further corrected (RTR_w). In this study, the considered SQGs derived from a database of marine sediment chemistry, provide values indicative of concentrations below which adverse effects are rarely observed.

The HQ_C (Hazard Quotient for chemistry) is calculated with Eq. 1 (Piva et al., (2011). RTR_w is the sum of the parameters with RTR < 1 (i.e. values below the sediment normative limit), while for those with RTR > 1, the RTR_w are individually added into the summation (Eq. 1):

$$HQ_C = (\sum RTR_{W^*(RTR < 1)} / N + \sum RTR_{W^*(RTR > 1)}) \quad (1)$$

where N is the number of parameters with RTR < 1.

HQ_C increases according to both the number and the magnitude of the exceeding parameters. Based on expert judgment, the values of HQ_C were assigned to one of six classes of chemical hazard identified according to different colours: absent (white) < 0.7; negligible (green) 0.7 ÷ 1.3; slight (azure) 1.3 ÷ 2.6; moderate (yellow) 2.6 ÷ 6.5; major (red) 6.5 ÷ 13; severe (black) > 13.

For ecotoxicological bioassays, the cumulative hazard quotient (HQ_{Battery}) is calculated by the summation of the weighted effects obtained by the summation of the weighted effects (Effect_w). The biological importance of the endpoint of each test and the exposure conditions (w₂) (Eq. 2):

$$HQ_{Battery} = \sum Effect_w(k) \cdot w_2$$

The HQ_{Battery} is normalized to a scale ranging from Absent (i.e. HQ_{Battery} < 1) to severe (i.e. HQ_{Battery} > 10) when all the assays exhibit 100% of effect; the HQ_{Battery} is then assigned to one of five classes: i) absent; ii) slight; iii) moderate; iv) major; v) severe.

Results from individual LOEs are finally elaborated within a classical weight of evidence approach which, after normalization of indices to a common scale, integrates and gives a different weight to various lines of evidence. An overall WOE level of risk, named Integrated Hazard Index (IHI), is thus calculated and associated to 1 of 5 classes (absent,

slight, moderate, major, severe) (Piva et al., 2011).

3. Results and discussion

3.1. Sediment chemical characterization

Sediment samples are essentially fine-grained soils, for which the clay fraction, CF, varies between 37.21% (S17 sample) and 56.98% (S9 sample), sand fraction, SF, between 3.35% (S8 sample) and 14.84% (S14 sample) and the silt fraction, MF, ranges from 39.04% of the S9 sample to 55.33% of the S11 sample (Todaro et al., 2018).

The sediments used for the tests were characterised by the physical-chemical properties reported in Table 2. pH ranged from 8.6 (minimum at site 4) to 9.9 (maximum on the site 18); while Eh ranged from -86.8 (site 4) to -174.6 (site 1). The parameters have a similar trend; they increase with depth. Moisture was ranging from 37.8% (sites 9) to 53.9 (site 17) while the percentage of organic matter (OM) was minimum at site 1 (10.9%) and maximum at site 4 (21.1%). Even if few data were available, it should be noticed a tendency of OM to decrease with depth; if the data detected are compared, it could be noticed that Mar Piccolo basin has similar or slight higher organic matter values then other sites (Sollecito et al., 2019). pH values and organic matter concentration in sediments influence the mobility of metals in the sediments, and consequently their distribution (Calace et al., 2008).

Analyses included priority and specific compounds (WDF, 2000) such as metals (As, Cd, Cu, Cr, Hg, Ni, Pb, V and Zn) and organic chemicals (PAHs and PCBs). Levels of these compounds were integrated with ecotoxicological bioassays to better assess sediment quality at different sites of Mar Piccolo. The contents of contaminants were better discriminated in comparison to the limits of site-specific law (ICRAM, 2004), highlighting differences between various sediment samples.

Concentrations of metals in sediments are shown in Fig. 2. In general, metal concentrations decrease with sampling depth. A different pattern was observed for site 1 (i.e., As, Cd, Cu, Hg, Ni, V and Zn concentration increases with depth) and site 9 (i.e. As, Cd, Ni and Zn concentration increases with depth). Nevertheless, As, Cd, Cu, Hg, Pb and Zn presented higher concentrations in the south of the Mar Piccolo (from sites 4, 6 and 7). With the exception of Ni, Cu and V, all the other metal levels (in at least one site) were higher than limits of site-specific law.

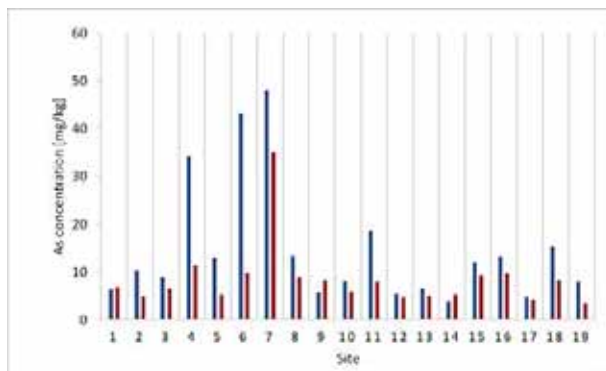
PAHs' and PCBs' concentrations in sediments are in Fig. 3. Levels were higher the detection limit in sites 1, 7, 8, 10, 13, 14, 15, 16, 17, 18 and 19 for PAHs and in sites 2 and 6 for PCBs; organic contaminants followed a decreasing trend with sampling depth, excluding the site 7. The highest PAHs from site 1 (9391 µg/g d.w.) was especially due to fluoranthene (624 µg/kg d.w.), anthracene (268 µg/kg d.w.), pyrene (562 µg/kg d.w.), chrysene (311 µg/kg d.w.), benzo[k]fluoranthene (408 µg/kg d.w.),

indeno[a,h]pyrene (280 µg/kg d.w.) among others. The sum of carcinogenic PAHs was lower than the non-carcinogenic; although lower PAHs' concentrations were measured at sites 15 and 16, carcinogenic PAHs were comparatively higher to non-carcinogenic. However, PAHs' concentrations are

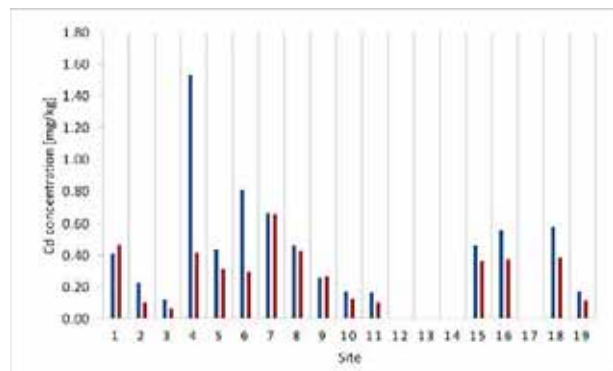
always lower to limits of site-specific law (4000 µg/kg). The knowledge of the concentrations of these compounds in sediments is important to evaluate the contamination of a marine ecosystem but alone, they do not provide information on potential toxicity for those organisms exposed to such chemical mixtures.

Table 2. Chemical parameters in sediments measured in different sampling site

Site	Depth from seafloor (m)	pH	Eh	Moisture (%)	Organic Matter (%)
1	0.0-1.5	8.8	-161.1	40.3	10.9
	1.5-3.0	9.1	-174.6	44.6	17.1
2	0.0-1.5	9.2	-117.3	48.3	15.5
	1.5-3.0	9.4	-125.2	45.9	16.9
3	0.0-1.5	9.3	-122.0	46.0	16.1
	1.5-3.0	9.2	-114.1	41.8	15.1
4	0.0-1.5	8.6	-86.8	47.7	21.1
	1.5-3.0	9.3	-120.2	39.7	19.6
5	0.0-1.5	9.2	-115.6	49.6	20.0
	1.5-3.0	9.1	-109.8	48.1	16.8
6	0.0-1.5	9.1	-110.8	46.1	15.3
	1.5-3.0	9.3	-126.6	40.3	10.7
7	0.0-1.5	9.4	-127.1	45.7	15.5
	1.5-3.0	9.4	-122.4	48.2	19.7
8	0.0-1.5	9.2	-116.2	50.2	15.3
	1.5-3.0	9.2	-108.7	52.0	15.5
9	0.0-1.5	9.4	-124.9	39.8	18.6
	1.5-3.0	9.3	-124.8	37.8	16.2
10	0.0-1.5	9.3	-121.6	49.4	17.2
	1.5-3.0	9.2	-119.4	46.7	16.9
11	0.0-1.5	9.2	-119.3	51.2	20.2
	1.5-3.0	9.3	-119.6	49.1	19.5
12	0.0-1.5	9.4	-131.1	48.3	16.3
	1.5-3.0	9.6	-137.1	36.4	11.1
13	0.0-1.5	9.3	-123.9	50.3	17.1
	1.5-3.0	9.5	-132.4	47.5	16.8
14	0.0-1.5	9.3	-120.9	50.7	17.0
	1.5-3.0	9.5	-132.2	43.1	15.1
15	0.0-1.5	9.4	-126.5	50.9	16.1
	1.5-3.0	9.5	-132.5	49.5	16.6
16	0.0-1.5	9.3	-122.3	48.1	19.8
	1.5-3.0	9.4	-126.0	43.6	17.5
17	0.0-1.5	9.2	-120.0	53.9	16.3
	1.5-3.0	9.3	-123.7	48.3	17.1
18	0.0-1.5	8.9	-101.3	52.6	18.3
	1.5-3.0	9.9	-104.5	48.7	15.2
19	0.0-1.5	9.4	-126.9	49.5	14.5
	1.5-3.0	9.1	-113.1	47.9	17.7



(a)



(b)

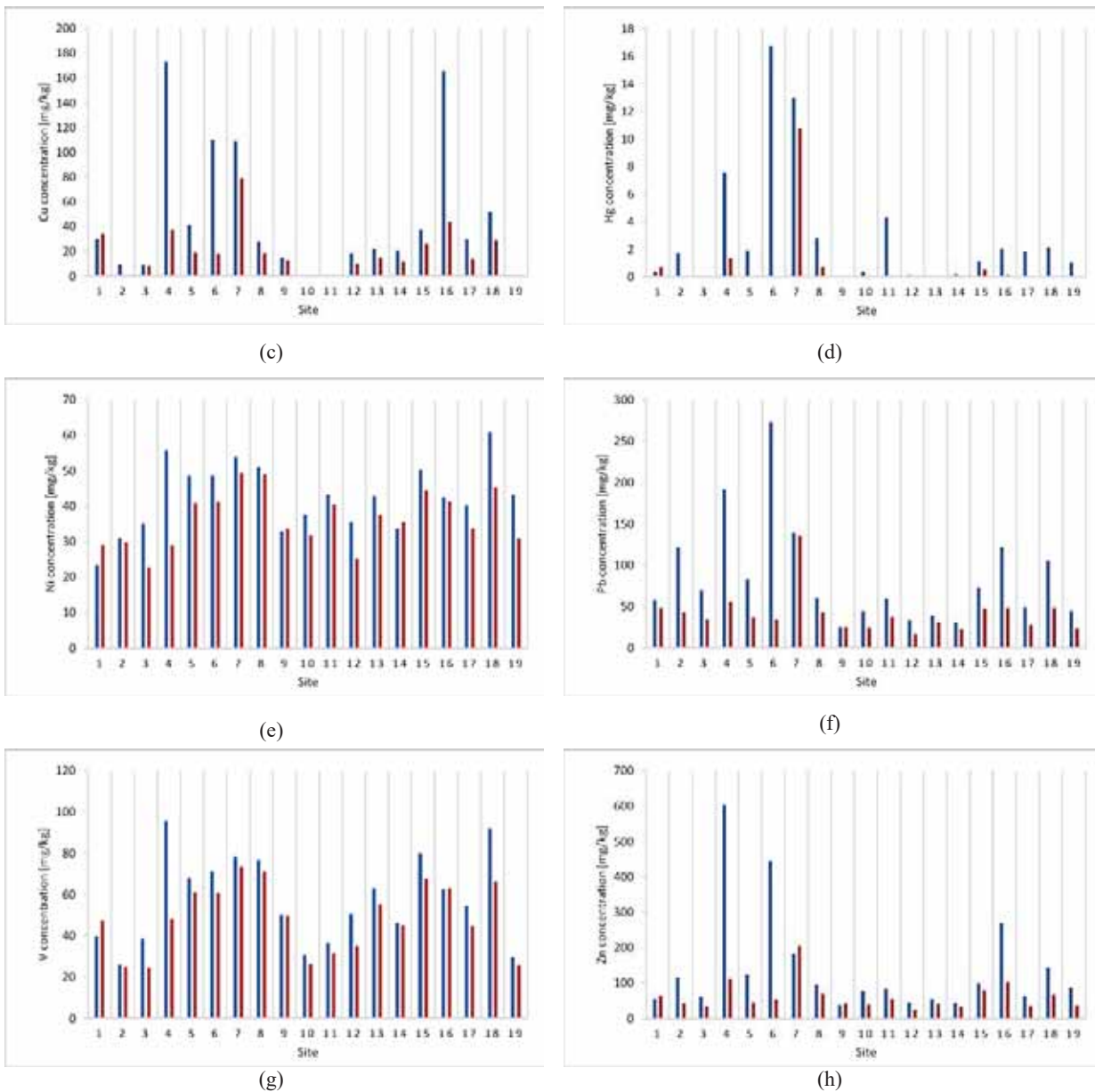


Fig. 2. Metal concentrations in sediments: (a) As; (b) Cd; (c) Cu; (d) Hg; (e) Ni; (f) Pb; (g) V; (h) Zn. (Blue bars are used for samples taken at 0.0-1.5 m below seafloor and red bars for samples taken at 1.5 - 3.0 m below seafloor. The dashed lines indicate the site-specific law (ICRAM, 2004))

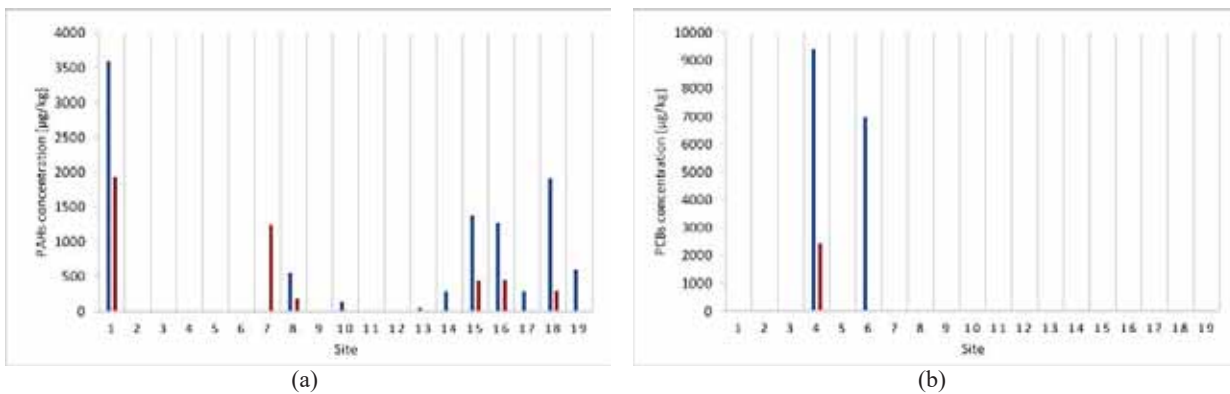


Fig. 3. Organic contaminants concentrations in sediments: (a) PAHs; (b) PCBs. (Blue bars are used for samples taken at 0.0-1.5 m below seafloor and red bars for samples taken at 1.5 - 3.0 m below seafloor. The dashed lines indicate the site-specific law (ICRAM, 2004))

3.2. Sediment ecotoxicological characterization

Toxicity results are shown in Table 3 for both bioassays, as well as integrated toxicity judgements according to Ministerial Decree (2016). The toxicity score was based on five toxicity classes: absent, low, medium, high and very high. Only the bioassay measuring the inhibition of larval development in *M. Galloprovincialis* was markedly affected by all the sediment samples, while more heterogeneous responses were obtained for other bioassays. Several elutriates were stimulatory relative to the negative control (i.e. test that is essentially free of contaminated sediment), presumably because of the release of nutrients such as ammonia.

The possible contribution of ammonia, as confounding factor, was investigated according to Losso et al., (2007). Total ammonia values in elutriates were under the no observed effect concentration (NOEC = 5 mg/l), which is considered as the sensitivity threshold limit value of the test towards ammonia. They were therefore not considered as confounding factors.

The elaboration by Italian Law summarized the hazard indicated by the whole battery of tests as “Slight” for sediments S1 (0.0-1.5 m), S9 (0.0-1.5 m), S5 (0.0-1.5 m) and Moderate for the other samples (see Table 3). The ranking indicated that all leachate samples could be deemed as moderate toxic.

The results have put in evidence that not always the presence of contaminants in sediments is a toxicity index. In the case of S1 also having found high concentration of PAHs, they are not found to be the main responsible of the toxicity of sediments.

3.3. WOE combined assessment

Data obtained from sediment chemistry and bioassays were finally integrated within a WOE model which elaborates specific hazard indices for each typology of data, before their differential weighting in an overall quantitative risk assessment. An example of the model output for elaboration of different LOEs is given in Fig. 4. The chemical characterization of sediments (LOE1) is typically summarized toward various SQG, providing the quantitative value of

chemical hazard quotient (HQ_C), the parameter which gives the highest contribution (in %) to the HQ_C, the number of exceeding parameters, the number of parameters (among those analysed) which are considered in that SQG, the total number of analysed parameters and the level (or class) of hazard assigned to HQ_C (from Absent to Severe).

The module on ecotoxicological bioassays (LOE4) summarizes results for both individual bioassays and for the integrated battery, including number of tests, threshold of the battery, value of the HQ_{Battery} and class of hazard for bioassays.

The results of analysis were compared by Sediqualsoft to baseline chemical levels (LCL) from the Italian law for the management of dredged sediments, that are respectively values typical for national coastal sediments potentially causing negative effects on aquatic communities (MD, 2016). In the top layer (0.0-1.5 m), the concentration of several heavy metals (i.e. Cd, Cr, Cu, Hg, Ni, Pb and Zi) was exceeding than LCL. PCBs showed a very high concentration in the south area (with maximum concentrations of 9300 µg/kg d.w. in station S04 and 6900 µg/kg d.w. in station S06). The PHAs exceed limits in several areas (i.e. S01, S02, S04, S05, S06, S07, S015, S016 and S18), reaching maximum value of 18000 µg/kg d.w in area S06.

Sediments from the superficial layers result more contaminated than those from deeper layer. Biological responses highlight an unexpected low toxic effect for model organisms exposed; Hazard Index for biomarkers range from “absent” (north area) to “moderate” (south area). Fig. 5 show the distributions of the Integrated Hazard Index (IHI) at various depths below the seafloor (i.e. 0.0-1.5 m in the Fig. 5a and 1.5-3.0 m in the Fig. 5b). In the top layer, the integration of data identified the higher risk at areas influenced by industrial activity (i.e. sites 4, 6, 7 near naval arsenal); while only site 9 appeared as not impacted. To some extent, the polluted areas (i.e., sites 10, 12, 14 and 19) are close to the areas of mussel-culture activity and fishery areas, raising concerns with respect to environmental and health issues. In general, the class of hazard tend to decrease with depth; only in the stations S03 and S09 the index is lower in the top layer (Fig. 5).

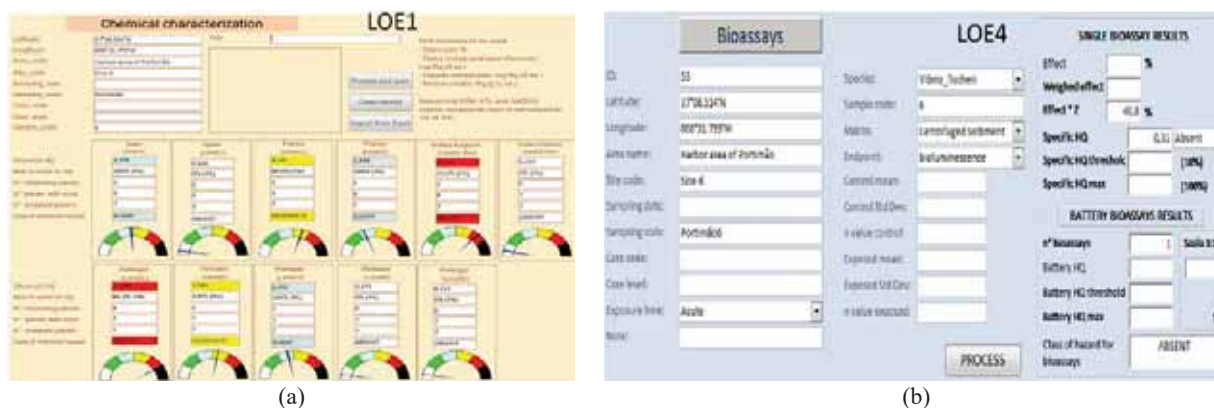


Fig. 4. Representative model output of hazard elaborations from various Lines of Evidence: (a): LOE1, sediment chemistry; (b): LOE4, bioassays

Table 3. Toxicity effect (in percentage, %) resulting from leachate samples prepared in seawater (positive values: toxic effects; negative values: stimulation effects)

Site	Depth from seafloor (m)	<i>Vibrio fischeri</i>	<i>Phaeodactylum tricornutum</i>	<i>Mtilus Galloprovincialis</i>	Toxicity data integration
1	0.0 – 1.5	-21.7	-27.6	2	Slight Toxicity
	1.5 – 3.0	-23.3	-31.5	60	Moderate Toxicity
2	0.0 – 1.5	14.5	18.6	100	Moderate Toxicity
	1.5 – 3.0	5.0	16.2	100	Moderate Toxicity
3	0.0 – 1.5	17.0	18.3	100	Moderate Toxicity
	1.5 – 3.0	37.5	17.3	100	Moderate Toxicity
4	0.0 – 1.5	21.7	-55.7	100	Moderate Toxicity
	1.5 – 3.0	11.3	-38.2	100	Moderate Toxicity
5	0.0 – 1.5	18.0	-32.3	100	Moderate Toxicity
	1.5 – 3.0	26.0	-39.8	100	Moderate Toxicity
6	0.0 – 1.5	7.0	-42.4	100	Moderate Toxicity
	1.5 – 3.0	25.0	-32.0	100	Moderate Toxicity
7	0.0 – 1.5	-12.7	-45.0	100	Moderate Toxicity
	1.5 – 3.0	-22.0	-38.5	100	Moderate Toxicity
8	0.0 – 1.5	-38.7	-46.7	100	Moderate Toxicity
	1.5 – 3.0	-15.0	-44.4	100	Moderate Toxicity
9	0.0 – 1.5	4.5	6.0	28	Slight Toxicity
	1.5 – 3.0	-4.0	17.2	73	Moderate Toxicity
10	0.0 – 1.5	11.0	16.7	87	Moderate Toxicity
	1.5 – 3.0	-7.0	16.3	100	Moderate Toxicity
11	0.0 – 1.5	46.0	16.8	100	Moderate Toxicity
	1.5 – 3.0	21.5	15.8	100	Moderate Toxicity
12	0.0 – 1.5	1.5	16.4	100	Moderate Toxicity
	1.5 – 3.0	9.5	15.6	100	Moderate Toxicity
13	0.0 – 1.5	7.0	17.1	100	Moderate Toxicity
	1.5 – 3.0	5.0	17.6	63	Moderate Toxicity
14	0.0 – 1.5	27.0	18.5	100	Moderate Toxicity
	1.5 – 3.0	40.5	15.9	100	Moderate Toxicity
15	0.0 – 1.5	8.0	17.2	33	Slight Toxicity
	1.5 – 3.0	-15.5	12.3	59	Moderate Toxicity
16	0.0 – 1.5	21.3	-27.9	87	Moderate Toxicity
	1.5 – 3.0	19.0	-20.7	84	Moderate Toxicity
17	0.0 – 1.5	3.50	16.4	100	Moderate Toxicity
	1.5 – 3.0	17.0	14.8	100	Moderate Toxicity
18	0.0 – 1.5	18.0	-44.5	100	Moderate Toxicity
	1.5 – 3.0	23.7	-41.9	100	Moderate Toxicity
19	0.0 – 1.5	37.5	18.9	100	Moderate Toxicity
	1.5 – 3.0	38.0	15.8	100	Moderate Toxicity



(a)



(b)

Fig. 5. Spatial distributions of the Integrated Hazard Index: (a) 0.0-1.5 m below seafloor; (b) 1.5-3.0 m below seafloor. (Class of hazard: ● Absent; ● Slight; ● Moderate; ● Major and ● Severe. In red the mussel-culture areas)

One reason can be the dynamic conditions (site 3 is in the direction of Navigable Channel and site 9 is near submarine freshwater springs, called “Citro Citrello”) that might promote a mixing of sediments. The IHI confirmed the critical situations in the Mar Piccolo; the level typically ranged from Slight to Major.

4. Conclusions

The assessment of the health status of sediment of Mar Piccolo highlighted the importance of combining sediment chemistry and ecotoxicological assays and revealed the existence in the marine basin of areas strongly compromised (sites at higher risk) due to industrial and harbor activities.

Results confirmed that sediments are strongly contaminated; nevertheless, the obtained biological responses highlighted an unexpected toxic effect: level of contamination seems to not affect in a proportional manner the biological compartment. All these observations raise numerous questions about the potential mobility of pollutants and further risks for the surrounding environment, making this site a model environment to study.

SediquaSoft model was confirmed a useful tool to elaborate complex scientific data in integrative indices for stakeholders and decision makers, supporting a more comprehensive process of site-oriented management decisions.

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CIRCULAR ECONOMY AND UPCYCLING OF WASTE AND PRE-CONSUMER SCRAPS IN CONSTRUCTION SECTOR. THE ROLE OF INFORMATION TO FACILITATE THE EXCHANGE OF RESOURCES THROUGH A VIRTUAL MARKETPLACE

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Abstract

The activities connected with the construction sector are responsible for several environmental impacts, both in the construction sector, and in the many manufacturing sectors involved in the supply chain of materials and products (mining sector, manufacturing sector, waste treatment etc.). The building products have a marked cross-sectorial connotation, according to the ANCE 2016 report, 31 economic sectors out of 36 are suppliers in the construction sector. One of the possibilities to reduce the environmental impacts of this sector is the limitation of the impacts of extraction, supply and production of materials, enhancing the possibility of using secondary raw materials from various sectors. From a circular economy perspective, the possibility of exchanging recyclable waste materials is crucial. In this regard, the paper presented, deals with the theme of strategies for the activation of waste inter-sectorial recycling scenarios. The hypothesis is the creation of a virtual marketplace, structured in an organized network, where the different users (producers or potential users of scraps/waste, industrial process planners, territorial administrators, etc.), can identify and locate scraps/waste usable for recycling. The use of the marketplace requires the profiling of companies that can offer resources and/or search for them, using specific search keys. The research can be conducted identifying: the secondary raw material obtainable (through the CER code and/or Omniclass 41), the origin or destination supply chains (through the NACE, UNI8290 and Omniclass 21 coding) and the georeferencing (through GIS). The work presented is the result of a post-doctoral research project funded by the Fondazione Fratelli Confalonieri of Milan, which had as its central theme, the systems for the exchange of recyclable waste and scraps.

Keywords: waste, recycle, circular economy, environment

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1. Introduction

The global environmental problems that humanity has to face today are largely due to the excessive exploitation of natural resources. This approach represents a serious problem for the planet, which face the exponential growth of its population (with a consequent request for a greater quantity of resources) and does not assist to a radical transformation of the actual trend on the resource management (in order to achieve a better control, a reduction, etc.). The Global Footprint Network

annually estimates the date on which the Earth Overshoot Day occurs (the day in which humanity depletes the resources that the planet produces in a year), and we can observe that in the most industrialized countries, this day arrives always in advance on the calendar. In 2018, Earth Overshoot Day for the planet occurred on August 1, earlier than the last year. This is caused by the most widespread model of economic development at a global level (linear model with high production of waste), which does not provide for a strict control of the use of resources, and does not consider the need to transform

all productions into sustainable production systems for the environment and the planet. Despite numerous initiatives aimed to change the production trends, Europe is still closely linked to the old production systems. The European Environment Agency (EEA, 2018) reported that Europe currently consumes almost twice the resources it can produce. This modus operandi is no longer sustainable and increasingly requires radical and widespread initiatives, aiming to a radical change in production systems and consumption models.

Among the main pressures inflicted on the environment by humanity, there are, with no doubt, waste. Waste are a manifold problem: their disposal is expensive both economically than environmentally, they can create problems to human health, they are not a valued resource etc. All the aspects listed are able to influence the quality of human life, turning waste into a serious problem, which can be perceived both at the local level and at the global level. Waste is also a problem for many companies, because they affect production both in economic terms (disposal costs) than in environmental terms (environmental impact of production). Turning waste into a resource, is therefore an important change, in order to make the industrial production more sustainable (Blengini et al., 2017; EC Communications, 2011a, 2011b, 2014a, 2014b, 2015). According to studies conducted in Europe, we can affirm that the construction sector, together with the use phase of the buildings (EC Communications, 2014b), are able to use about half of the materials extracted and to produce about a third of all waste. This sector generates environmental pressures at each stage of its life cycle, but it is possible to highlight the greater consumption of resources both in the production phase of construction products and in the phase of disposal. In the construction phase, the problem is connected to the use of resources (in many cases virgin materials) and to the production of waste (both during production than during assembly of material or component). In the phase of decommissioning, the main problem is the tendency to not recover waste from C&D, and to not separate them in an appropriate way to recycle etc.

The main theme is the average lifetime of materials or components, which is never long enough to compensate the impacts produced. This causes a great loss to the economy, because typically they are either disposed in landfills (finally ending the useful life cycle of the material) or are recycled according to the principles of downcycling (recycle that distorts the material as such, transforming it into something less noble and of a lower quality). Being able to make resource consumption more efficient is therefore one of the few viable strategies (EEA, 2017, UNEP, 2017) to improve the environmental situation. This represents a real change for the lifestyle of people and in production for the companies, and these changes must be encouraged and promoted in all possible ways. The information sharing is at the center of this research contribution (outcome of a post-doctoral research project funded by the Fratelli Confalonieri

Foundation of Milan), which started with the aim of making productions processes more sustainable, through the contribution of sharing information and the creation of intelligent networks able to trade in resources.

The study is focused on the construction sector, because it is extremely representative of the waste management problem: too many sectors, large quantities of materials and countless companies involved (ANCE, 2016; Migliore et al., 2018). Moreover, the economic importance of construction sector should not be neglected, because it generates almost 10% of the GDP of the European economy and represents about 20 million jobs (EC Communication, 2012). Therefore, starting from the assumption that waste represents a resource and not a problem, the intention of the work is to make possible the tracing of waste and their cataloging and sorting through a virtual marketplace, in which different actors/users can interact tracing resources or tracking down possible partners to which allocate their waste.

2. Reduction and reuse of resources in the construction sector

The construction sector is currently object of numerous initiatives aimed at improving the management of virgin natural resources and the reuse/recycling of scraps/waste. Both in European than in Italian contexts, various initiatives have been launched to improve the current situation. The political interest on this issue is very high and many tools have been made available to support initiatives aimed at improving production systems by controlling and reducing the use of resources. In the European context, the 2014 COM 445 (EC Communication, 2014b) highlighted the actions that can be activated to make the construction sector more sustainable and the results which can be obtained. Among the possible actions listed (EC Communication, 2014b) emerge: the promotion of a design that can calibrate the use of resources with respect to the needs and functionality of the building and promotes the selective demolition scenarios; the planning of building site activities in order to improve a better energy-efficient use of resources and products; the promotion and the production of more resource-efficient construction products (use of recycled materials, reuse of existing materials, use of waste as fuel etc.); the spread of resource-efficient buildings and renovations, where construction waste is reduced and materials and products are recycled/reused in order to dispose a smaller quantity of waste in landfills. Obviously, some key aspects are fundamental for the success of long-term initiatives, such as the existence of an efficient recycling system at local, regional or national level which represents an attractive and cost-effective alternative to the landfill. It should be noted that the interest in recycling is often determined by specific factors: the transport distance to the recycling sites (positive balance between induced impacts and avoided impacts, economic compensation between

disposal costs and transport costs to recovery sites, etc.), and the possibility of being able to guarantee a level of purity and standardization of the recovered materials that should be reused. The standardization factor will become one of the key aspects in the marketplace, because if the characteristics of scraps/waste material, (intended to direct or indirect recovery) are not clarified, it will be difficult to find potential users. The COM 455 also speaks about the importance of the dissemination of standardized and comparable information, a fundamental tool to allow anyone to make choices that are not only ethical but also, and especially, professional and technical. Indeed, speaking about the great quantity of tools and supports for environmental improvement, means to acknowledge that often there is no linearity between information, especially between global scale and national scale.

Another aspect that emerges, related to products or building environmental certifications, is that they are predominantly voluntary, and the percentage of certified buildings is currently very low, about 1% (EC Communication, 2014a, Ecorys, 2014). This aspect, if supported at national and EU level, could encourage greater interest from companies to do better under the environmental point of view and to highlight the certification as a factor of competitiveness. Having available helpful and usable information, could in fact facilitate decision-making processes and launch industrial ecology scenarios at different scales.

3. The virtual marketplace for the exchange of resources

The purpose of this work is to demonstrate the importance of the role of information in promoting the recovery and recycling of resources, focusing on the construction sector. The actions carried out to structure the marketplace were: analysis of good practices (replicable and perfectible in other production contexts), taxonomy of waste (identification of fundamental characteristics that can allow the traceability of waste for a possible cross-sectorial transfer), contextualization on the territory of resources (geo-localization), creation of synergies in the marketplace.

3.1. Analysis of best practices in the European context

A cognitive action carried out with the aim of identifying some possible best practices, already experimented and therefore replicable, was the one related to the study of research projects concerning the valorisation of waste in the European context. Nowadays, the experiences of recovery and enhancement are many, and they are rapidly spreading. However, research and documenting these experiences in an appropriate way is not easy; if the initiatives are conducted privately by the companies the modus operandi is never disclosed (for obvious reasons of industrial policies), but when these eco-

innovation processes take place through public funding, we can rely on a lot of information.

For this reason, the study focused on research projects funded within the European Union on issues that concern the protection of the environment and more specifically the valorization, recycling or reuse of waste. These projects respond to the main macro themes (EC Communication, 2011a, 2011b, 2014a, 2014b, 2015) promoted by the EU on environmental issues, and the most analyzed were: Life projects (EEC, 1973), Cip projects (EU, 2006) and Horizon 2020 projects (EU, 2013), which represent the most successful initiatives found on the national and international scene. The study conducted on projects belonging to these investment programs, is divided into two phases. The first phase was aimed at identifying relevant projects in the construction sector that operate on the theme of recovery and valorisation of scrap/waste. The survey was conducted using the IT support provided by the European Union.

Concerning to recycling, the elimination of waste and its reduction, emerges that there is a marked interest in recycling, but the elimination and reduction of waste are actions that are difficult to implement on their own. However, in many cases it has emerged that some initiatives are addressed to combine both reduction, elimination and recycling.

It is clear that, in order to activate circular economy scenarios, it is not possible to limit the study to a single perspective. If the scope of the initiative is wider, more results will be achieved (it is necessary to overcome the "company boundary" and think about systemic solutions that can involve different subjects, even belonging to different sectors of goods). The second phase of the study was aimed to summarize the results of the analysis carried out on the projects (Fig. 1), and to categorize them according to different research keys (NACE code, CER code, Omniclass 21) that will be useful to filter and identify that projects which meet the needs of the users of the marketplace.

The categorization system used (Migliore et al., 2014, 2015, 2016) starts from the examination of the projects, identifies the type of innovation implemented (new production process, innovative production process, new product with recycled content, innovative product with recycled content, service) and categorizes the type of impact expected from the project (recycle, reduction or zero waste). Then, it tabs the projects using different types of codes that will allow the tracing (CER code for the type of waste, NACE code for the type of the product sector involved in entry and exit, Omniclass code to identify the type of product for the building involved etc.).

3.2. Proposal of a waste taxonomy

A very important action carried out with the aim of making fluid the exchange of resources, is tracing the waste with a unique code that makes its identification simple and immediate. The knowledge of scrap/waste is essential to be able the transfer and/or the reuse it in other production contexts.



Fig. 1. System of analysis and categorization of research projects identified as best practices

One of the problems that currently restricts the use of waste is a lack of knowledge of how and what is produced in terms of scrap/waste. Many waste materials are potentially reusable, but without a system that can trace and identify their position, quantity and quality it is difficult to implement any type of recovery. A valid strategy to facilitate the reuse and enhancement of pre-consumer material waste, could be the implementation of an easier access to information from potential stakeholders (end users). The activation of industrial synergies based on recycling, depends on the possibility of tracing waste that can be valorized.

The characteristics that distinguish them are not always compatible with the possible reuse or re-insertion in the supply chain of origin or in other

supply chains. To do this, it is essential to share valid information about the characteristics of potentially exploitable waste. The information has to be well organized and related to the various assessment purposes, that may be necessary to implement continuous recycling plans. It should be emphasized that waste is often not considered in its potential as a second raw material, due to insufficient information associated with it (spatial location, quantity, morphology, technical characteristics, availability over time etc.). Currently, the waste is identified with the CER code (EC Communication, 2014c), a unified system at European level that allows to recognize the waste compared to the sector and the production process that generates them, as well as for their hazardous or non-hazardous.



Fig. 2. System for the construction of the waste identification code

However, in some cases this code is rather generic (usefulness for a simple disposal), and useless for the purpose of a possible recovery/enhancement. The CER code is not created for this purpose and it fails to transmit information relating to the quality and characteristics of the waste. However, it represents a good starting point for being able to construct a speaking code that identifies a refuse in a univocal way by associating it with a series of useful information for the purpose described.

The proposal presented in this research project aims to associate different information through the combined use of the NACE code (Eurostat, 2007), the CER code (EC Communication, 2014c) and the Abaco code (Fig. 2). The proposed code is divided into 4 parts: the first part groups all the processes that compose it under the main production chain (it cannot be codified and can only be referred to as a trace for specific research on the supply chain); the second part of the code derives from the NACE database (Eurostat, 2007) and identifies the processes that mainly make up the supply chain; the third part identifies the scrap/waste according to the CER code (EC Communication, 2014c); the fourth and last part identifies the waste according to the "Abacus code". The Abaco code object of the research consists of three fields separated by points that identify three main peculiarities of the waste useful for the identification for recovery purposes.

Figure 2 shows the trace followed to build the Abacus code which is considered valid for recycling: the type (pre-scrap, byproduct, sludge, powder, post-scrap), the size (small, medium, big) and the purity of the waste (pure, mixed, scrap). Obviously will be also necessary to draw up a technical sheet that will contain specifications of mechanical, physical and chemical nature.

3.3. Contextualization on the territory of the materials

The last action carried out, to build the necessary information for the recovery and exploitation of waste, concerns the knowledge of quantity and location. This operation can be done through the use of GIS (Geographic Information System) applications. The information implemented with this logic can also contribute to evaluating other details, such as transport network, hydrography, population characteristics, economic activities, jurisdiction and other characteristics of the social and natural environment. In this study, these variables were not considered, but it is clear that they can be implemented and become useful data to provide other studies and other views of the territory.

With reference to the project conducted, the goal is to be able to place on the territory the quantities of by-products, waste and exploitable waste and to develop monitoring and analysis of possible scenarios. For example, through the Geoportal of the Lombardy Region, using the GIS set up for the land register, it

can be shown how an instrument already in use at the regional level can also become useful for recycling and for the circular economy.

Figure 3 shows an extract of the elaborations that can be done through the Lombardy Region Geoportal and shows that this tool already provides an "identity card" of the quarries, information that can be implemented and made even more useful under other aspects. For example, by entering the quantities of materials extracted and the quantities of scrap/waste not recovered, it will be possible to provide useful data to the planners in order to activate management plans optimized from the environmental point of view. Similarly, the industries present on the territory could be mapped and with the same logic of the quarries, a series of information on the production of waste could be referred to them. All this information is part of the network of facilities made available in the marketplace to track industrial ecology scenarios.

3.4. Structure of the marketplace

The marketplace proposed by this research is configured as a space in which heterogeneous users with different objectives can relate in order to identify possible forms of collaboration. The common aim is to extend the useful life of resources, through exchanges of waste materials, which can turn into resources for other productions. The marketplace is therefore configured as a real network of companies and actors with decision-making power on the production chains. The expectations arising from the use of the marketplace are to encourage the exchange of scrap/waste through their technical profiling. This operation becomes possible according to the connections established between the needs of the different "users" (Fig. 3). The first users are the scrap/waste producers who want to offer it, the second users are the «users» looking for the secondary raw material, the other hypothetical «users» are professionals looking for materials with recycled content or «stakeholders» who intend to start up innovative recovery processes within their production chains.

The different users, profiling themselves, will have to define their scope of operation (NACE sector). This initial information will allow us to identify companies that belong to the same sector and which, having common interests, can establish profitable industrial connections for environmental improvement. Beyond this primary information, users will provide specifications to implement four macro-areas, sufficient to be able to track down resources (primary, secondary) and output products from production processes. The macroareas identified are raw materials (which are used to produce the main product of the supply chain), products, by-products (if any) and waste. Each user can fill out this information according to what he is offering and what he is looking for.

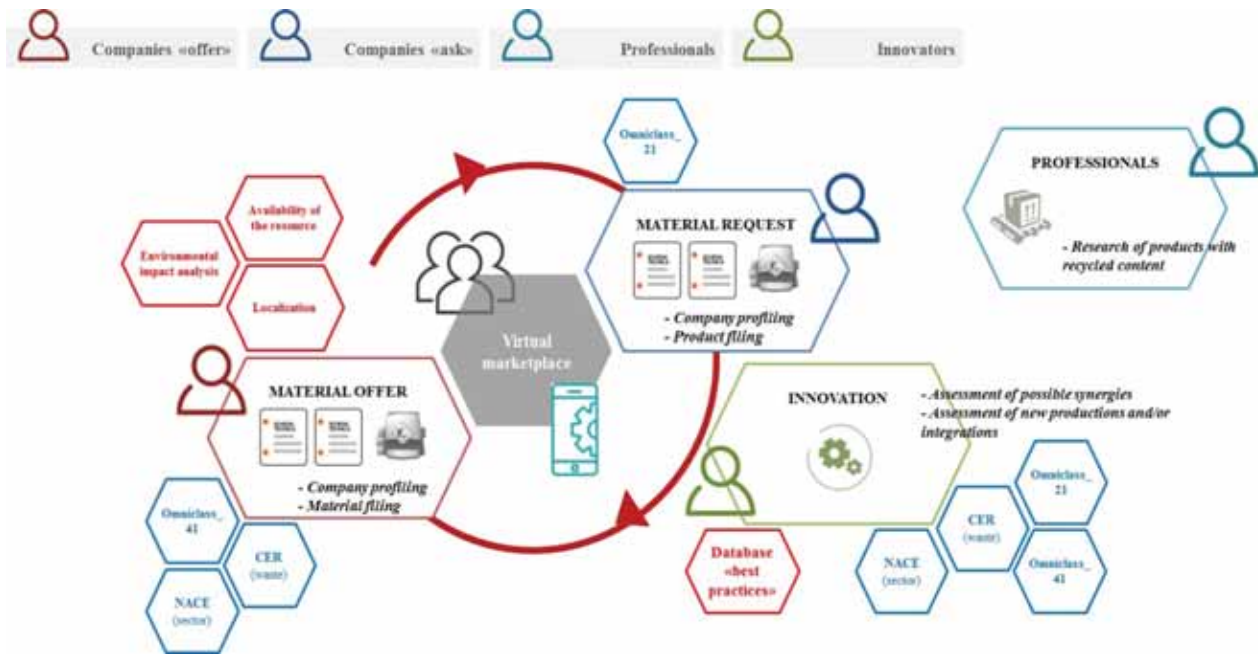


Fig. 3. Structure of marketplace

A greater quantity of information increases the chances of establishing connections, which corresponds to the principles of circular economy.

4. Conclusions

The research work presented focuses on the importance of the close relationship between the parties and the importance of unique and well-understood information. The reuse and exploitation of waste represents a difficult problem to manage due to stringent regulations on the sector. However if from industrial processes we learn to produce only products and by-products (previously called waste), there would be no waste production or at least this would be reduced. This operation becomes simpler and more immediate with the use of a virtual marketplace, because different users are related to different purposes, and the material that otherwise would be destined to landfill is made available.

Following the logic adopted, waste is no longer such, but becomes by-products, with standardized and well-declared characteristics. This simplification is the real strength if we want to promote the valorization and recovery of materials. It has to be noted that currently many resources are wasted (through landfill disposal) because we do not have full knowledge of what is available, and we do not know the hypothetical users of the resource.

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- The text of the paper should be divided into **Introduction, Materials and methods (or Experimental), Results and discussion, Conclusions, References** (for papers dealing with environmental management, policy, education etc., the Experimental part can be replaced by case-studies presentation).

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State the objectives of the work and provide an adequate background, avoiding a detailed literature survey or a summary of the results.

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3. Results and discussion

Results should be clear and concise. Discussion should explore the significance of the results of the work, not repeat them. Avoid extensive citations and discussion of published literature.

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Formulae, symbols and abbreviations:

Formulae will be typeset in Italics (preferable with the Equation Editor of Microsoft Office 2003) and should be written or marked as such in the manuscript, unless they require a different styling. The formulae should be numbered on the right side, between brackets:

$$a^3 = 3M / 4N \quad (1)$$

Always refer in the text to Equations as (Eq. 1), Eqs. (1-4) etc.

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Faber K., (2000), *Biotransformations in Organic Chemistry – A Textbook*, vol. VIII, 4th Edition, Springer, Berlin-Heidelberg-New York.

Handbook, (1951), *Handbook of Chemical Engineer*, vol. II, (in Romanian), Technical Press, Bucharest, Romania.

Symposia volumes: Names and initials of authors, year (between brackets), paper title, symposium name, volume number, place, date, page numbers:

Clark T.A., Steward D., (1991), *Wood and Environment*, Proc. 6th Int. Symp. on Wood and Pulping Chemistry, Melbourne, vol. 1, 493-498.

Journal papers: Names and initials of authors, year (between brackets), full title of the paper, full name of the journal (italic), volume number (bold), first and last page numbers:

Tanabe S., Iwata H., Tatsukawa R., (1994), Global contamination by persistent organochlorines and their ecotoxicological impact on marine mammals, *Science of the Total Environment*, **154**, 163-177.

Patents: Names and initials of authors, year (between brackets), patent title, country, patent number (italic):

Grant P., (1989), *Device for Elementary Analyses*. USA Patent, No. 123456.

Dissertations: Names and initials of authors, year (between brackets), title, specification (PhD Thesis, MSc Thesis), institution, place:

Aelenei N., (1982), *Thermodynamic study of polymer solutions*, PhD Thesis, Institute of Macromolecular Chemistry Petru Poni, Iasi, Romania.

Star K., (2008), *Environmental risk assessment generated by natural hazards*, MSc Thesis, Institute of Hazard Research, Town, Country.

Legal regulations and laws, organizations:

Abbreviated name, year (between brackets), full name of the referred text, document type, author, URL address:

ESC, (2007), Improving access to modern energy services for all fundamental challenge, Economic and Social Council, ENV/DEV/927, On line at: <http://www.un.org/News/Press/docs/2007/envdev927.doc.htm>.

EPA, (2007), Biomass Conversion: Emerging Technologies, Feedstocks, and Products, Sustainability Program, Office of Research and Development, EPA/600/R-07/144, U.S. Environmental Protection Agency, Washington, D.C., On line at: <http://www.epa.gov/Sustainability/pdfs/Biomass%20Conversion.pdf>.

EC Directive, (2000), Directive 2000/76/EC of the European Parliament and of the Council of 4 December 2000, on the incineration of waste, Annex V, *Official Journal of the European Communities*, L 332/91, 28.12.2000, Brussels.

GD, (2004), Governmental Decision No. 1076/2004 surnamed SEA Governmental Decision, regarding the procedure for strategic environmental impact assessment for plans or programs, *Romanian Official Monitor*, Part I, No. 707 from 5th of August, 2004.

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