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CONSTRUCTED WETLANDS AS RELIABLE DECENTRALIZED SYSTEMS FOR WASTEWATER TREATMENT AND REUSE IN THE MEDITERRANEAN AREA: MAIN ISSUES AND CONCERNS



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PhD Thesis 2019

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Dipartimento di Agricoltura, Alimentazione e Ambiente Di3A

UNIVERSITÀ DEGLI STUDI DI CATANIA

"Agricultural, Food and Environmental Science" XXXII CYCLE

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"There is a water crisis today. But the crisis is not about having too little water to satisfy our needs. It is a crisis of managing water so badly that billions of people - and the environment - suffer badly."

World Water Vision Report

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Please note: equations embedded in the text maintain the original numerical order, as reported in the published format, and must be considered in within each contextual chapter or appendix in order of appearance.

Abbreviations and Acronyms

AS	Activated Sludge
BMP	Best Management Practice
BOD ₅	5 days Biological Oxygen Demand
COD	Chemical Oxygen Demand
CDVI	Climate-Demography Vulnerability Index
CFU	Colony-Forming Unit
CSEI	Centro Studio di Economia applicata all'Ingegneria
(Catania	a)
CSO	Combined Sewer Overflow
CSS	Combined Sewer System
CW	Constructed Wetland
DSS	Decision Support System
DTM	Digital Terrain Model
DWTF	Decentralized Water Treatment Facilities
EC	European Community
EU	European Union
ECs	Emerging Contaminants
ECOSA	N Eco-Sanitation
EDCs	Endocrine-Disrupting Compounds
EFR	Environmental flow requirements
E. coli	Escherichia coli
FAO	Food and Agriculture Organization
FaD	Fill and Drain
FC	Fecal coliform
FEM	Floating Emergent Macrophyte
FFM	Free-floating Macrophyte
GHG	Green-House Gas
GIS	Geographic Information System
HSSF	Horizontal Sub-Surface Flow
HRT	Hydraulic Retention Time
H-TWs	Hybrid Treatment Wetlands

IC	Irrigation Consortia
IWMI	International Water Management Institute
Ks	Hydraulic Conductivity at Saturation
LID	Low-Impact Development
MD	Ministerial Decree
MBR	Membrane Reactor
MS	Member State
LD	Legislative Decree
NBS	Nature-Based Solution
O&M	Operation and Maintenance
P.E.	Person Equivalent
PAHs	Polyclic Aromatic Hydrocarbons
PPCPs	Pharmaceuticals and Personal Care Products
R.E.	Removal Efficiency
RW	Reclaimed Water
SDG	Sustainable Developed Goals
SDS	Sewerage and Drainage System
SF	Surface Flow
SIAS	Servizio Informativo Agrometeorologico Siciliano
SSS	Separate Sewer System
SUDS	Sensitive Urban Design Systems
TFWW	Total freshwater withdrawn
TRWR	Total renewable freshwater resources
TSS	Total Suspended Solids
TW	Treated Wastewater
TWs	Treatment Wetlands
UASB	Upflow Anaerobic Sludge Blanket
UN	United Nations
VDF	Vertical Down Flow
VUF	Vertical Up Flow
VSSF	Vertical Sub-Surface Flow
WHO	World Health Organization
WSUD	Water-Sensitive Urban Design

WW WWTP Wastewater Wastewater Treatment Plant

Abstract

Nowadays, there is a wide recognition on global water stress for several reasons, and a real crisis is specially occurring unbalanced and ineffective water resources due to allocation. In this sense, there is an urgent need to prove, adopt and promote, suitable and reliable integrated water management plans. Decentralized water treatment facilities (DWTFs) have been described as one of the best practices (BMPs) for effective water management programs in the circular economy framework. Under this perspective, recovery and reuse of valuable resources, water in primis, are identified and strongly pursued. In this sense, all types of water resources, even unconventional water resource like wastewater (e.g. civil, industrial, agricultural etc.), can be considered valuable to easy the exploitation of freshwater supply and to make more resilient cities in the next future.

At the light of that, this thesis aims at: (i) analyzing main reasons related to the water crisis, describing pros and cons of the most commonly used DWTFs, providing a general evaluation on water reuse scenario and an insight of principal issues and barriers still hindering the water reuse practice, with a special focus on Italy; (ii) promoting strategic approaches and solutions in water resources management of semi-arid areas; (iii) evaluating constructed wetlands (CWs) as reliable and viable DWTFs for unconventional water recovery and reuse in Mediterranean increasing knowledge on climate; (iv) hydraulic performance of horizontal flow (HF) CWs and general treatment efficiency of hybrid-CWs, with different design and applications.

Main findings concern: (i) the validation of a GIS-based decision support system (DSS) for assessing the effective

potential of treated wastewater (TW) reuse, in Sicily; (ii) the feasibility of TW reuse in Sicily, even if limited by unfair local governance; normative and ineffective (iii) experimental evidences on the viability of hybrid-CWs as DWTFs for different applications and operation modalities (i.e. alternative treatment of two types of WW, highly different in terms of nutrient-richness); (iv) the optimization of hydraulic monitoring of HF units, both by combining different methodologies, and defining an easy and precise methodology for K_s estimation through the falling head test, for clogging evaluation.

Sommario

Al giorno d'oggi sono molte le problematiche connesse al tema della carenza idrica. Queste principalmente riguardano una distribuzione disomogenea e inefficace delle risorse idriche. Il trattamento e il recupero delle acque reflue tramite sistemi decentralizzati può considerarsi, a tal proposito, una delle migliori pratiche per una gestione integrata delle risorse idriche nell'ottica dell'economia circolare. In questa prospettiva, il recupero e il riutilizzo di risorse preziose, l'acqua *in primis*, sono stati individuati e devono essere perseguiti.

Alla luce di tali considerazioni, il presente lavoro ha l'obiettivo di: (i) esaminare le principali cause legate alla crisi idrica, descrivendo pro e contro dei sistemi di trattamento decentralizzati; (ii) valutare il potenziale riuso delle acque reflue e le principali barriere che ne ostacolano un'applicazione a scala reale, con particolare attenzione al caso dell'Italia; (ii) promuovere approcci strategici nella gestione delle risorse idriche delle aree semi-aride; (iii) valutare l'efficienza di trattamento di impianti di fitodepurazione come infrastrutture decentralizzate affidabili per il trattamento e riuso delle acque non convenzionali nelle tipiche condizioni di clima mediterraneo; (iv) accrescere le conoscenze sulle prestazioni idrauliche di impianti di fitodepurazione a flusso orizzontale e dell'efficienza di trattamento di impianti ibridi, diversi per tipologie e campi di applicazione.

principali Ι risultati ottenuti riguardano: (i) l'implementazione di un sistema di supporto decisionale basato su piattaforma GIS, per valutare il potenziale reale di riutilizzo delle TW, in Sicilia; (ii) la conferma delle potenzialità del riuso delle acque reflue in Sicilia, sebbene limitata da normative inadeguate; (iii) le evidenze sperimentali sull' affidabilità di impianti di fitodepurazione ibridi come sistemi decentralizzati utilizzati per diverse applicazioni; (iv) la messa a punto di una metodologia speditiva per la stima della conducibilità idraulica a saturazione.

1 General Introduction¹

1.1 Problem Statement

Water is limited, but a renewable vital and primary resource. Water stress results from an imbalance between water demand and water resources. As reported by the United Nations, "water scarcity can mean scarcity in availability due to physical shortage, or scarcity in access due to the failure of institutions to ensure a regular supply or due to a lack of adequate infrastructure". Globally, in fact, there is not a water shortage as such, but nations and regions need to urgently face the critical problems presented by water stress, so then managing demand should be a primary focus. Integrated water resources management offers a wide framework for governments to conform water use patterns with the needs and demands of different users, including the environment. (UN, 2019). As documented in Figure 1, for the period 2000-2015, the levels of water stress by country vary worldwide. This trend will continue, even more evidently in the next decades. In fact, the per capita water use is increasing due to changes in lifestyle, population growth and migrations (climatological, political and economic). This, together with spatial and temporal

¹ A modified version of this chapter was submitted as Ventura, D., Barbagallo, S., Consoli, S., Milani, M., Sacco, A., Rapisarda, R. & Cirelli, G. L. (2019c). On the performance of a novel hybrid constructed wetland for stormwater treatment and irrigation reuse in Mediterranean climate. AIIA Mid-Term Conference Matera 2019.

variations in water availability (exacerbated by global warming), means that the water to produce food for human consumption, industrial processes and all the other uses is becoming scarce. In particular, water withdrawals for irrigation represent 66% of the total withdrawals and up to 90% in arid regions, the other 34% being used by domestic households (10%), industry (20%), or evaporated from reservoirs (4%) (World Water Council, 2017). The indicator used for water stress evaluation (Figure 1), has been described as the ratio between total freshwater withdrawn (TFWW) by all major sectors and total renewable (TRWR). freshwater resources after considering environmental flow requirements (EFR). It is calculated using the following formula (eq. 0):

Water Stress (%) =
$$\frac{TFWW}{TRWR - EFR} * 100$$
 (0)



Figure 1. Levels of water stress by country (%) (2000–2015); Data source: FAO Aquastat and IWMI (*SDG: Sustainable Developed Goals from 2030 Agenda) (FAO and UN, 2018).

The rising anthropic water use does not only reduce the amount of water available for human being wellness, societal progress and development, but heavily injure aquatic ecosystems so much so that environmental balances are disrupted and cannot play their regulating role anymore. Also, the water crisis may be cause of international and national transboundary tensions among different users which are sharing water resources. It is estimated that more than 260 river basins are shared by two or more countries, that without strong institutions, clear agreements and cooperation policies can become points of regional instability and, at worst, of conflicts (World Water Council, 2017). Also, the adoption of suitable water resource management plans still could mitigate both, the causes and the effects of water scarcity and avoid the completion of a water crisis. In this complex panorama, the possibility to protect freshwater supply and ensure access to potable water for all, clean water for different purposes, and sanitation services for human health protection, are the most challenging issues. This is the focus of the 6th goal of the 2030 Agenda for Sustainable Development (UN, 2018). To this aim, the recycling approach is considered as the core perspective for all goods management, and naturally of water, in all its shapes. In addition to conventional natural fresh surface water or groundwater, wastewater (WW) generated as a by-product of specialized processes or that pre-use need suitable treatment are known as unconventional water resources (e.g. desalinated seawater and highly brackish groundwater, black water, grey water, stormwater, agricultural drainage water, etc...). In this sense, all types of WW can be considered as sources of valuable compounds and not as "wastes" anymore, and their reuse can alleviate the pressure on freshwater. According to the Eco-sanitation (Ecosan) concept, in fact, human excreta and nutrients, are finally considered basic resources. The last is a systemic approach, an aptitude (used and well known in the past), in contrast with the conventional water wasting flush toilets-based sanitation, supporting the idea of "sustainable, closed-loop system, which closes the gap between sanitation and agriculture" (Langergraber and Muellegger, 2005). Thus, WW should be collected separately in order to optimize the potential for reuse, and then treated and reused as closest as possible to the site where they are produced for water, nutrients (nitrogen and phosphorous) and bio-energy (produced by organic material) valorisation. In this context, intended uses for reclaimed water, beyond the agricultural, the first in term of demand, should be clear and quality standards functions of those. At the light of this, successful integrated water resources management plans for sanitation and water recovery systems should include a meticulous understanding of all the component required to the intended use pursued. In many cases, also rainwater harvesting, garden wastes and animal manure, can be integrated as sources of precious compounds. The whole system, including social and natural issues should be considered in relation to every local situation and climate, geographical and temporal conditions also evaluated (e.g. hydrology, landscape and ecology). All stakeholders should be involved and aware in order to ensure a strong participatory approach during the planning and decision making processes. This is a strategic feature since it has been widely recognized the importance of rising awareness of people for good and useful policies (Poppe et al., 2018; Berry et al., 2019). The Ecosan perspective is aimed to combine all types of available technologies, ranging from conventional and high-cost installations and

materials, to near-natural WW treatment techniques, but picking them for a final ecological benefit, even if the components of the chosen and planned treatment strategy can be not necessarily "eco" per se. In this sense, considering case by case, and depending on the localspecific needs of rural centres, sub-urban, industrial, commercial or developing urban clusters areas, great funding will be required and solutions will range over the best relative strategies. Sewerage networks, on-site sanitation systems, like pure decentralized plans (WW and valuable by-products are processed, treated and reused locally) or like satellite systems (variably connected to the centralized WWTP) (Gikas and Tchobanoglous, 2007) will be therefore opportunely adopted, combined and integrated. Generally, community-based sanitation solutions are now being adopted in many countries, providing among various benefits, local jobs creation and fostering environment protection. Local and national governments will be asked to manage and regulate sanitation systems as a priority, and will have the chance to strength their authority on the territories, with local community agreement backing. To aim, the development and use of information that management systems, especially in low and middle-income countries, is considered a useful platform, as already demonstrated for GIS-based procedures application as effective support tools in integrated water management (Sharma et al., 2012; Owusu et Asante, 2013; Hama et al., 2019). Finally, in the light of the above, policy makers are considered the main characters of the scene. In fact, they firstly should be able to rethink and reconfigure the actual SDS order, free from business-driven old schemes and conception and willing to have a holistic approach. This would be the only way to cope with what will be known, otherwise, as a real "wastewater crisis" (De Feo *et al.*, 2014), in the near future.

<u>1.2 Decentralized water treatment facilities (DWTF) for</u> <u>effective WW resource management: general considerations</u>

Water recovery and reuse can be pursued through different approach and technologies, intensively engineered or more likely as low cost and energy saving nature-based solutions (NBSs). The latter uses or mimics natural processes to increase water availability (e.g. soil moisture retention and groundwater recharge), improve water quality (e.g. natural and constructed wetlands and riparian buffer strips), and minimize water-related risks by restoring flood plains and constructing decentralized water retention systems such as green roofs. As already mentioned, resilient cities of the future should rely on unconventional and non-punctual water source for easing the exploitation of freshwater supply. Urban scenarios, from the huge massively cemented areas to the smaller ones, could, therefore, benefit from decentralized water treatment facilities (DWTFs) for effective water management. These, in fact, have been described as one of the best practices (BMPs) for promoting the circular economy, protecting hydrological balance, threatened by global warming, reducing the expensive investment for centralized facilities O&M services and for conforming inadequate sewerage and drainage systems (SDSs). On this regard, De Feo et al. (2014) pointed out the unfeasibility to extend existing centralized water and WW systems to cope with the increasing level of urbanization, with consequent extra water demand and waste loads. Furthermore, many WW treatment facilities still remain

vulnerable to the adverse effects of climate variability, with different frequencies in droughts and floods all over the world. Moreover, in developed economies, like those of the European Union (EU) Member States (MS), many infrastructures ageing and deteriorating. while are developing countries are facing challenging and costly investments for upgrading poor SDSs. With the exception of Australia, in most part of the world SDSs are traditionally in combined configuration, leading to Combined Sewer Overflow (CSO) management issues (Liquete et al., 2016), and hindering the benefits from WW resource streams from separated sources. Centralized systems are historically recognized as the unique feasible infrastructures for WW management, since allow to collect them in heavily dense populated areas by mean of extended sewage and a centralized WWTP. In this sense. decentralized infrastructures would not constitute an affordable and immediate alternative, but since investments could be gradually put in place in relation to the type of populated area, the levels of growth and development, they could be considered as flexible and adaptive supporting measures, with the advantage of local WW reuse. Developing commercial and residential districts, as also hospitals, could represent viable examples for effective reuse plans. Generally, all current WW treatment technologies could be used into a DWTF, taking into account that relatively small volume of WW produced by single households or groups of habitations have to be locally treated, usually within a maximum distance of less than 3 km. However, as already their choice and combination should discussed. be attentively evaluated, depending on multiple factors, as also WW strength/variability, performance, low capital and O&M costs, energy inputs, land area requirement ("footprint"), and local reuse needs (Capodaglio *et al.*, 2017).

The most traditional and ordinary decentralized DWTFs are underground septic tanks and waste stabilization ponds (anaerobic, facultative and aerobic). The first, efficient for TSS settlement and some anaerobic digestion, particularly in hot climate, but ineffective for pathogen reduction, also require post-treatment. The second, very simples, with high hydraulic retention times (HRTs), favorable to pathogen abatement, produce suitable effluent for irrigation (high algae concentrations), and reduce nitrates-related risks of ground water contamination. However, their principal disadvantage is the high land area requirement. Also, small activated sludge (AS) plants are still used in rural and developing contexts.

Recent advises from the research on the most common decentralized systems highlighted few treatment processes, ranging from NBSs, like constructed wetlands (CWs), to improved aerobic or anaerobic biological systems (Singh *et al.*, 2015). Among these, aerobic treatment systems, like membrane biological reactors (MBRs), non-membrane/ biomass retention systems and anaerobic treatment systems, such as upflow anaerobic sludge blanket (UASBs) are reported. Main sustainability-related issues of all these technologies are resumed in Fig. 2. In some case, however, costly high-tech applications could be encouraged by advanced remote operating control systems.

DWT	Health & Hygiene	Environment & Resources	Technology	Financial	Socio-cultural & Institutional
CWs	May be set up for solar disinfection (post-treatment)	Natural engineered systems. Energetically almost neutral. Good compatibility with sparsely populated locations. No resources recovery (out possible vegetation harvesting)	Easy to operate. High robustness and low vulnerability to crises. High adaptability if physically possible to expand. High water loss in hot climates.	Investment cost mostly for land plot. Operation close to free if gravity flow possible.	Acceptance good if 'out of the way' and not causing nuisance. Possible poor institutional understanding (nonstandard practice)
Aerobic Conventional	Require post- treatment	Energy intensive. Current mainstream technology. Possibility of tertiary recovery of nutrients (struvite) and energy from sludge	Relatively easy to operate with remote control. Medium robustness and vulnerability (power cuts, discharge toxicity). Expandability possible at medium-high costs. Suitable for cheaper 'package' construction for smaller facilities.	High investment and O&M costs (energy and sludge management).	Acceptance depending on location and past experience. Possible nuisance from odours. Well accepted institutionally.
MBR Aerobic	May be suitable for reuse without post-treatment	Very energy intensive. Smaller footprint than aerobic conventional. Higher efficiency. Possibility of tertiary recovery of nutrients.	More complex operation, with fouling problems in time. Robust towards flow and load variations, vulnerable to power cuts (medium), less to toxicity. Expansion requires high investments. Suitable for cheaper 'package' construction for smaller facilities.	Highest investments and O&M (increased energy, but less sludge to manage)	Acceptance depending on location and past experience. Possible nuisance from odours. Accepted with cost-concerns institutionally
Aerobic Filtration	Will likely require post-treatment	Energy intensive (aeration). Footprint comparable to MBRs, similar efficiency.	Operation simpler than MBRs. Other conditions similar, lower investment for expansion. Suitable for cheaper 'package' construction for smaller facilities.	Higher investment but O&M lower than MBRs. (less energy and sludge to manage)	Acceptance depending on location and past experience. Possible nuisance from odours. Accepted with cost-concerns institutionally
UASB	Require post- treatment	Anaerobic technology can be energy neutral or positive (biogas generation in the presence of strong wastes). Possibility of post- recovery of nutrients.	Relatively easy to operate at optimal conditions. Robust towards flow/load variations, vulnerability low. Expansion at medium cost. Suitable for cheaper 'package' construction for smaller facilities.	Medium investment, Low O&M, sludge and effluent management. Possible high revenue from biogas recovery.	Acceptance depending on location and past experience, considering likely nuisance from odours. Cost-recovery (energy) enhances institutional support.

Figure 2. Sustainability-related issues compared among most used DWTF technologies (Capodaglio *et al.*, 2017).

<u>1.2.1 Constructed wetlands as DWTF: main principles,</u> <u>applications and critical issues</u>

Natural wetlands are areas of land where the water table is at or near the surface for at least part of the year and are characterized by the presence of adapted vegetation (hydrophytes) and soil characteristics that have developed in response to the wet and saturated conditions (Mitch & Gosselink, 2007; Kadlec & Wallace, 2009). These ecosystems are highly productive and characterized by a high capacity to transform and store organic matter and nutrients. CWs reproduce, in a controlled environment, the natural purification processes characteristic of wetlands, improving water quality. These variably engineered systems, simple from a technical and operational point of view, exploit the complex interactions of natural ecosystems between soil, vegetation, micro-organisms, water and the atmosphere. In particular, main physical-chemical and biological contaminant transformation processes are: sedimentation and filtration, microbial conversion and degradation, plants and microbial uptake, chemical paths and precipitation, peat accretion and volatilization (gas exchanges). There are many different types of CWs, and since many different design variations can be considered, their classification consists of a complex hierarchy (Fonder & Headley, 2013). Three are the major components of a treatment wetland: the macrophytic vegetation types, the presence of water-logged filtering media and the inflow of contaminated water to be treated. On these basis, hydrology and vegetation characteristics are the main traits for CWs categories differentiation. Particularly, in relation to water flow position and direction, degree of media saturation and position of influent loading, two major groups can be identified: surface flow-based systems with a benthic substrate and those with subsurface flow passing through a porous media. The first can be divided depending on the type of vegetation into surface flow (SF), free-floating macrophyte (FFM), and floating emergent macrophyte (FEM) CWs. The second group always present sessile emergent macrophytes and the flow direction shapes different designs: horizontal sub-surface flow (HSSF), vertical sub-surface flow (VSSF), vertical down flow (VDF), vertical up flow (VUF) and fill and drain (FaD) CWs. All these systems can be used in their standard forms or combined (H-CWs) in order to fulfil many treatment requirements for variable WW quality, and therefore for many different applications (Zhang et al., 2014; Gorito et al., 2017; Gorgoglione and Torretta, 2018; Wang et al., 2018). The system design can, in fact, determine very variable biological and chemical dynamics occurrance, particularly when a H-CW is set up. In fact, thanks to the arrangement of different CWs types all potential pollutants removal mechanisms can be exploited (Stefanakis and Akratos, 2016). Also, intensified CWs variants are known and applied, largely based on different aeration modalities (e.g. tidal flow, artificial aeration and recirculation), with increased energy, chemical or operational inputs. Other variants of vertical CWs for raw sewage and septage treatment for sludge dewatering and mineralization are respectively the French systems (Molle et al., 2005; Morvannou, et al., 2015) and the Sludge treatment systems (Nielsen. 2003: Enhanced treatment Brix. 2017). performance of intensified CWs have been widely documented in the last decade (Vymazal, 2007; Vohla et al., 2011; Zhang et al., 2014). Ilyas and Masih (2017) described

lacking oxygen transfer, poor nutrients removal (nitrogen and phosphorous), and high footprint as the main limiting factors for successful CWs adoption and widespread, even hindering the low cost and overall high treatment performance. In fact, as discussed by the same authors, for selecting the best relative wetland type for a specific practical application, the most suitable indicators are: pollutants removal efficiency, footprint and use of energy. Several works and approaches have been dedicated and tested to deepen the knowledge on reducing footprint besides achieving good quality effluent (Prost-Boucle and Molle 2012; Foladori et al., 2013; Zapater-Pereyra et al., 2015). Among these, great focus has been put on varying the hydraulics by recirculation of treated waters, using of filland-drain cycles or artificial aeration, selecting new and or reactive media and evaluating the effects of solids accumulation in the filtering media. The latter, commonly referred as clogging, is described and approached as a key phenomenon in the hydraulic behaviour of CWs. As part of the treatment process, in fact, filter media will gradually become clogged due to factors mainly related to influent characteristics, system designs and operation activities. The effects of clogging on treatment performance have been quite debated. Some authors agreed about its negative influence on the removal and hydraulic performances of the CW and, also reducing the lifetime of the system (Cooper et al., 2005: Caselles-Osorio and Garcia, 2006: Nivala and 2009). Gorgoglione and Rousseau, Torretta (2018) discussed on the importance of selecting suitable substrates hydraulic parameters for specific (e.g. hydraulic conductivity, HRT, etc...) and therefore to system removal efficiency and Tatoulis et al. (2017) affirmed that the reduction of pore volume of the substrate media, can affect

CWs performance. Other authors, however, did not observe any decrease in treatment performance when clogging phenomena occurred (Vymazal, 2018; Marzo et al., 2018). Since during design phase the hydraulic conductivity is overestimated to face the clogging phenomenon occurring during the CW operation, monitoring and assessment of clogging would be then recommended for evaluating system footprint reduction, as also for its long-term management and lifespan forecasting. The clogging is clearly а widespread operational problem and, for these reasons, it has become increasingly important to identify practical methods for its measurement. CWs require, therefore, a precise understanding of system hydraulics for their correct design and efficient operation. Hydraulic behaviour of CWs could be investigated in situ by means of hydraulic conductivity at saturation (Ks) measurement, clog matter characterizations as well as hydrodynamic visualizations, by means of tracer tests (Headley and Kadlec, 2007; Aiello et al., 2016, Licciardello et al., 2019). The suitability of available measurement techniques, in terms of accuracy, repeatability as well as time and skill required, moreover can vary depending on the substrate type, system design as well as clogging degree and distribution.

<u>1.3 Unconventional water source: general overview on</u> <u>stormwater and its reuse</u>

There is a global rising recognition, particularly in developed countries, that stormwater is a valuable unconventional resource that should be harvested and reused for encouraging overall non-drinking water supply, waterways pollution control and water crisis reduction. In

contrast with the traditional concept of stormwater like something to be disposed of quickly, actual sustainable water management focuses on stormwater reuse as a key resource for securing adequate future water supplies based on the approach of 'water fit for purpose' especially in large urban centres (Begum et al., 2008; Goonetilleke et al., 2017). Definitions of stormwater are very variable, but should be generally referred to the concept of "runoff". Many authorities, worldwide, gave their definition, but all agreed on its importance, since it is a valuable water resource, and strongly impacts on human living spaces, infrastructure and natural environments (Van Leeuwen et al., 2019). According to United States Environmental Protection Agency (EPA, 2019), "stormwater runoff is generated from rain and snowmelt events that flow over land or impervious surfaces, such as paved streets, parking lots, and building rooftops, and does not soak into the ground." Moreover, stormwater picks up all kinds of chemical and biological pollutants encountered on the way, such as many types of industrial wastewaters, litter, dust and soil, microorganisms, trash, chemicals, oils, grease, and also special and sometimes recalcitrant pollutants (e.g., biocides, pharmaceuticals and personal care products (PPCPs), other micro-pollutants, metals, and the less known "per and polyfluorinated alkyl substances" (AGDD; 2016). In particular, through mixing with sewerage effluents, in some cases (e.g. CSO), very high microbial pathogens loads can occur, together with other WW-derived pollutants like antibioticresistant genes (Garner et al., 2017) and endocrinedisrupting compounds (EDCs) (Kalmykova et al., 2013). The rapid anthropic growth has led to an increasing level of urbanisation and so to the expansion of impervious surfaces (there is an increasing number of megacities: population >

10 million (UN, 2014)), which also corresponds to increased stormwater runoff flow rates, volumes and pollutant loads displaced, and obvious detrimental effects on rivers, streams, lakes, and coastal waters. Depending on the type of catchment area and the outflow volumes (urban, rural or industrial), the impact on receiving water bodies can change significantly, also due to the wide variety of pollutants involved (e.g. emerging pollutants cause great concern also considering their combined effects on natural ecosystems) and to very complex load dynamics. Already He et al. (2008) discussed the correlation between stormwater quality and climatological variables, which clearly differ at the regional scale. These are also exacerbated by climate changes, and consequent effects of extreme drought and flood events. Samson et al. (2011) predicted in detail the most vulnerable areas of the world to global climate change through the year 2050, highlighting that less-developed areas will face significant change. To this aim the same authors developed a new metric called climate-demography vulnerability index (CDVI), estimated by subtracting climate vulnerabilities from demographic annual growth rates, displayed as percentage of annual growth rates of human density (Figure 3). Ironically, the hardest-hit are those least-responsible for green-house gas (GHG) emissions and those already dealing with high temperatures and drought conditions.



Figure 3. Global Climate-Demography Vulnerability Index (CDVI). Red corresponds to more vulnerable regions, blue to less vulnerable regions. White areas correspond to regions with little or no population (Samson *et al.*, 2011).

At the light of these considerations, stormwater management presents quality and quantity issues traditionally faced for community protection from adverse sanitary and flood impacts caused by rainfall and runoff. Generally, authorities have been historically focused on formulating guidelines and standards for setting minimum requirements for flood management. Australia witnessed this approach and stormwater runoff has been managed by mean of separated sewer systems (SSS), differently to USA, China, the UK, and some European states, where combined sewer systems (CSS) commonly convey stormwater and wastewater together through single pipework systems (Van Leeuwen et al., 2019). CSS and relative risks of CSO, particularly during high flow events, present various drawbacks in terms of treatment requirements, due both to the difficult elimination of emerging contaminants (ECs) (i.e. EDCs, PPCPs) through conventional WW treatment processes, huge stormwater volumes, and high rate flows. However, Li et al. (2013) documented that SSS not necessarily offer a plenty of benefits when compared to CSS for pollution control and present similar risks, while Park et al. (2017) found that significant ECs concentrations can come also from diffuse sources, like agriculture and urban runoff. Moreover, costly infrastructure for stormwater collection, treatment (without considering the need to provide adequate treatment technologies for still unknown ECs compounds) and distribution to users should not be undervalued. Hence, it is true that SSS has been generally considered effective for stormwater and WW management, and in recent past and present times adopted, also for easing and increasing recycling and reuse processes at various levels. But, on the the approaches other hand. newest to stormwater management suggest not to focus any longer on the point of discharge and to reconfigure the catchment area in order to minimise treatment requirements and flood risks. Recently, stormwater management practices for stormwater retention and rainwater infiltration, have been proposed worldwide as: water-sensitive urban design in Australia (WSUD), the "Sponge City" in China, sustainable urban drainage systems in the UK (SuDS) and low-impact development in the USA (LID) (Zimmer et al., 2007; Begum et al., 2008, Joksimovic and Alam, 2014; Beza et al., 2018; Chan et al., 2018). As part of this perspective, various blue-green infrastructures (e.g. urban forests, CWs, green roofs and green walls) can reshape global cities, preserving aesthetic values, ensuring recreational facilities, promoting natural habitats and water reuse strategies, and reducing the carbon and energy footprint. Many applications for reclaimed stormwater have been described and comprise agricultural, industrial and commercial uses, green infrastructure, non-potable uses (i.e.

toilet flushing and washing), urban green areas and sport fields irrigation (Schwecke et al., 2007; Kazemi and Hill, 2015), and also potable use (Gerrity et al., 2013). Practical treatment measures consist in sand filtration, biofiltration, gross pollutant traps, grassed swales, sedimentation ponds and wetlands (Aryal et al., 2010; Hatt et al., 2006). Nevertheless, as reported by Goonetilleke et al. (2017), stormwater are still globally underestimated, because of related uncertainty and unreliability due to seasonal supply and therefore to quantitative and qualitative issues that could complicate the implementation of storage and treatment planning and also designing strategies. In conclusion, in order to reduce the consequent counterproductive effects of combined quantitative and qualitative constraints (e.g. increased management and treatment costs) and to facilitate the design phase, the intended use is a key concept allowing to use and valorise different water, with highly variable quality.

1.4 The Italian context

Italy can be considered a representative slice of the global panorama in terms of various WW management issues and contradictions. Urban WW collection and treatment are largely lacking and insufficient (where present, these are mostly based on combined sewerage systems), in fact, as reported by the European Community (EC), 620 agglomerations in 16 regions are in breach of the EU rules (EC, 2019). In particular, several EC infringement procedures and one condemnation of the Court of Justice, highlight absent or not complying infrastructures in 62
(>10.000 PE) and 5 (>2.000 PE) agglomerations in Sicily (CSEI, 2018). In the case of first flush rain water and stormwater runoff, Italian normative (Legislative Decree 152/06, Art. 113), specified that the Regions, subject to the authorization of the Ministry of the Environment, can regulate and manage, by means of specific rules and "case by case", these resources. On this regard, only few regions (Lombardy and Emilia Romagna) have promulgated the appropriate regional regulations, while the Sicily region, among others, does not have anyone yet. Stormwater runoff have been compared to industrial WW and their reuse, as well as for domestic and urban WW, is regulated by the Ministerial Decree (M.D.) 185/2003. However, the strictness of this law, which is reflected in costly monitoring activity, the absence of clear standard limits for specific intended use, and also the increasing tariffs to the expense of consumers for covering the reuse costs, generally discourages the spread of this practice (Ventura et al., 2019a). A clear example of that is offered when conducting microbiological monitoring for environmental quality assessment for WW reuse. This is often considered complex for costly time-consuming analyses required, and in the last decades there has been a need to focus on specific and suitable biological indicators. On that regard the Italian National Institute of Health (Bonadonna et al., 2002) already proposed Clostridium perfringens as an effective indicator for old fecal contamination, but it has never been included in the Italian standard limits for water reuse (M.D. 185/2003). Also the recent EU proposal for water reuse (European Commission, 2018) recommended Escherichia coli (E. coli) (already defined as primary indicator of fecal contamination, WHO, 1993) and Clostridium perfringens (indicator of old contamination), respectively, as the most

appropriate indicators for pathogenic bacteria and protozoa assessment. On this regard, as noted by Bichai et al. (2017), there is an important research need to demonstrate the suitability of pathogen "surrogates" for stormwater quality monitoring and assessment because of pathogens high variability in this context, as well as its dependency on operational conditions of the treatment system considered in its individuality. Masi et al. (2018), highlighted, on the other hand, how generally people are not willing to pay for wastewater reuse service and there is a need to demonstrate the reliability of costs-saving treatment technologies on the long term. The EU proposal for water reuse (2018) could be considered, in this context, as a first important step for easing and conforming its promotion and actualization at national level (Ventura et al., 2019a). The Italian M.D. 185/2003, boosts the use of natural systems, such as CWs, for wastewater treatment and reuse by considering more tolerable limits for E. coli and in general for nitrogen and phosphorous in case of irrigation. Nevertheless, a general decentralized and integrate WW management approach is still underestimated by policy makers when compared to conventional solutions, and is far from being considered an effective and systemic action plan to tackle water scarcity and a strategic support framework for agricultural needs in the Mediterranean area, particularly in semi-arid regions. In this sense, despite CWs are well known treatment technologies and largely applied since several decades at international level (e.g.in Germany, since the end of 70's), there is a knowledge gap on their technical performances, management procedures and design criteria (CSEI, 2018). Regarding to that, the possibility to increase and define technical knowledge and management guidance would finally turn it into the chance to knowingly relay on extensive, more adaptive and self-supporting DWTFs, fostering their spread and applications.

1.3 Objectives

The **general objectives** of the thesis are:

- to provide general evaluation on water reuse scenario and promote strategic approaches and solutions in water resources management of semi-arid areas, with specific insights on Italy and Sicily.

- to evaluate the reliability of CWs as viable decentralized wastewater treatment facilities for unconventional water recovery and reuse in Mediterranean climate.

- to increase knowledge on treatment efficiency and hydraulic performance of horizontal CWs, with different design and applications.

The **specific objectives** proposed are:

- to evaluate factors and constrains still limiting treated WW reuse in agriculture, in Sicily.

- to promote a GIS-based decision support system (DSS) for assessing the effective potential reuse of WW for its management in Sicily.

- to estimate the feasibility of WW reuse in Sicily, with regard to quality characteristics and in relation to national and international legislation and guidelines.

- to evaluate the removal efficiency of a pilot scale H-CW, alternatively treating stormwater runoff drained from a parking area and SBR-treated WW produced by a retail

store, for toilet flushing and green area irrigation effluent reuse.

- to evaluate the hydraulic performance of the HF stage of a full-scale H-CW and the effects of partial clogging on the overall treatment efficiency of the system, for effluent discharge or reuse.

- to study the effectiveness of combining different methodologies for monitoring the hydraulic behavior of the same HF unit, as a reliable tool for general approach.

-to monitor the hydraulic behaviour of the HF units of a pilot scale H-CW from the start-up phase for the entire system lifespan.

- to define an accurate and handy methodology for K_s estimation of different gravel filtering media at different spatial scales for clogging phenomena monitoring.

1.3.1 Outline of the thesis

A short overview of the thesis outline is hereby provided with the aim to facilitate the reading of the work and find a common thread among the chapters (2-5).

Chapter 2 illustrates the real potential for reclaimed water use in the Sicilian agricultural context. After discussing its main issues and barriers, a GIS-based feasibility study is proposed by integrating several information. The potential volume of TW by WWTPs to be connected to possible irrigation areas is evaluated in order to fulfil the water deficit. The feasibility survey takes also into consideration the main concerns risen up when comparing international guidelines with Italian standards.

Chapter 3 provides a case study of a full-scale H-CW for tertiary treatment of domestic WW produced by a retail store (SBR effluent), with a special focus on evaluating the influence of partial clogging on its HF unit to the overall performance of the system. As consequence of high number fluctuation in the of customers. frequent overloading peaks observed practically let the H-CW operate for secondary treatment. The hydraulics of the first unit, the HF bed, has been monitored through combined measurements of hydraulic conductivity at saturation (K_s) , tracer tests, and geophysical (i.e. electrical resistivity tomography-ERT).

Chapter 4 provides a case study of a novel pilot H-CW, connected to the same retail store described in chapter 2. The system is operated for alternative treatment of nutrient poor effluent, like the stormwater runoff drained from the parking area, and nutrient rich domestic WW above described. The removal efficiency of a decentralized system for stormwater treatment and recovery in Mediterranean climate is therefore evaluated. In particular, the effects of double feeding (operated to face typical local long dry periods and heavy/short rainfall events) on the overall system reliability are evaluated. The hydraulics of the HF units has been monitored through measurements of K_s .

Chapter 5 reports a study conducted defining a methodology to estimate K_s of gravel-based filtering media in HF units of real scale, pilot CWs as well as in laboratory conditions. Different operating schemes and equations for the Lefranc's test were compared and a new permeameter configuration is also proposed and tested. The focus is put on optimizing the

methodological approach in monitoring operations and data processing phases.

2 How to Overcome Barriers for Wastewater Agricultural Reuse in Sicily (Italy)?²

Abstract

This study reports an up-to-date summary of the principal barriers still limiting reclaimed water use for agriculture in Italy, and particularly in Sicily. Moreover, it provides a geographic informative system (GIS)-based procedure for evaluating the potential treated wastewater (TW) reuse in the Sicilian region as a decision support system for its management. The survey, based on possible economic, morphologic, and design solutions, evidenced a feasible integration of several wastewater treatment plants (WWTPs) with irrigation areas, allowing the water availability enhancement. Overall, the potential volume of TW by WWTPs (connected to irrigation districts) is 163×10^6 m³ y^{-1} , while the water deficit is 66 \times 10⁶ m³ y⁻¹. The feasibility of TW reuse in Sicily was also analysed at the light of the World Health Organization microbial risk assessment. E. coli analyses mostly accomplished these guidelines while conflicting with the restrictive Italian standards. Despite several limiting factors (restrictive legislations, high distance and unfavourable slope between **WWTPs** and irrigable areas, high monitoring and distribution costs) still hamper the exploitation of reclaimed

² A modified version of this chapter was published as Ventura, D., Consoli, S., Barbagallo, S., Marzo, A., Vanella, D., Licciardello, F. & Cirelli, G. L. (2019a) How to overcome barriers for wastewater agricultural reuse in Sicily (Italy)? *Water* 11 (2), 335. <u>https://doi.org/10.3390/w11020335</u>

water use in Sicilian agriculture, some solutions were identified to implement this practice.

Keywords: agricultural reuse; barriers; GIS-based management; irrigation districts; Risk assessment; treated wastewater

2.1 Introduction

Worldwide, arid and semiarid areas have increasingly experienced water scarcity. Such regions mostly use water for agriculture and crop irrigation (up to 70% of total water extracted), relying on groundwater and surface water sources (Al-Isawi et al., 2016a). To reduce stress on limited freshwater resources, non-conventional water like urban wastewater (TW), constitute an important solution for promoting and enhancing the sustainable use of the available water, as evidenced by (Tran et al., 2016). The same authors also highlighted the great potential of TW for irrigation of agricultural fields close to urban centres, also providing a considerable input of required nutrients for plants and reducing their net discharge on sensitive surface waters. As reported by (Salgot et al., 2017), among institutional and socioeconomic causes, a key drawback for agricultural TW reuse practice advancement and its public acceptance is the absence of an adequate international legislation, leading in many cases inhomogeneous quality standards and fairness issues. In fact, as mentioned by (Tran et al., 2016), and discussed in other studies (Salgot et al., 2017; Salgot et al., 2012) the suitability of reclaimed water for specific applications depends on its quality and usage requirements. Generally, in irrigation practices water quality controls should mainly consider factors such as salinity, heavy metals, and pathogens for minimizing any detriment to human, plants, and soils. Beyond the normative barriers, more general factors related to wastewater treatment plants (WWTPs) siting should be considered (Al-Isawi et al., 2016b). Among those, some authors (Leverenz et al., 2011) reported the following: long distances between treatment facilities and agricultural demand areas; construction and maintenance costs of conveying pipe systems; necessity to store TW during fall-winter periods, since TW are continuously produced throughout the year, whereas irrigation demand is generally concentred during crops growing season of dry-summer periods. Moreover, the controversy on the possibility to estimate the true costs of freshwater supply, with respect to the intrinsic value of and González-Villela. (Banderas 2015) may water consequently result in economic disadvantage for recycled water production.

In the attempt to develop their own recycling and reuse criteria, usually proceeding by the most advanced ones (e.g., California and Australia), Mediterranean member states, like Italy, Greece, and Spain, enforced "semiscientific" and too stringent regulations to be really applied (Salgot *et al.*, 2017). In light of this the EU (European Union) recognized the need to implement a common water reuse regulatory instrument at the international level and it is working towards both minimum quality requirements and health and environmental risk-based policies (Gawlik and Alcalde-Sanz, 2018). On 28 May 2018 a "Proposal for a Regulation of the European Parliament and of the Council on minimum requirements for water reuse" was issued (EC, 2018). However, in the perspective to adopt the new EU guidelines for reclaimed water use at the national level, many

constraints will probably remain for a long time, mostly referring to physical and economic ones. In Italy, all previously described issues strongly limit the agricultural use of reclaimed water (RW). Several studies already evidenced the need to enhance TW sustainable reuse and to promote management plans for available water resources in Sicily (Cirelli *et al.*, 2012; Cirelli *et al.*, 2007; Barbagallo *et al.*, 2012). The same authors in fact discussed how, in Sicily, relevant irrigation demand (Barbagallo *et al.*, 2012) is not satisfied mainly because of the increasing drought periods, the impairment of water bodies quality, and the rising civil demand.

Among the aforementioned legislative limitations, the restrictive Italian standards (MD 185/03) lack of quality guidelines for diversified agricultural reuse sectors and a microbiological risk-assessment approach in line with the World Health Organization (WHO, 2006), but present a list of overabundant water quality parameters (e.g., chemical and microbiological compounds) to be analysed Cirelli *et al.*, 2008), with consequent high monitoring costs, mainly for small treatment facilities (Figure 4). Additionally, total costs for developing TW reuse in agriculture are not sustainable and user-friendly when considering the construction, operation, and maintenance of "additional" processes for tertiary and disinfection treatments and RW distribution networks.



Figure 4. RW monitoring cost (MC) vs. RW volume.

The aims of this study are:

- (i) To evaluate the factors hampering the RW agricultural use spreading in Sicily;
- (ii) To provide a GIS-based procedure for evaluating the effective potential of RW use in the Sicilian region as a decision support system for its management; and
- (iii) To ascertain feasible possibility of RW use in Sicily, with regard to RW physico-chemical and microbiological characteristics. In particular, the restrictive Italian microbial standards for reuse (DM 185/2003), were compared with the findings of the WHO microbial risk assessment (WHO, 2006).

2.2 Materials and Methods

2.2.1 WWTPs in Sicily

The Sicilian WWTPs scenario herein presented is an up-to-date survey of the previous one reported in (Barbagallo *et al.*, 2012). With respect to the 259 WTTPs in

operation in the early study, more treatment facilities are herewith reported (Table 1), with a total of 459 urban WWTPs, enclosing also those not in operation and planned.

P.E.	Wastewater treatment plants				
	in operation	not in operation	Planned		
< 2000	87	31	19		
$2001 \div 5000$	80	17	9		
$5001 \div 10,000$	77	11	4		
$10,001 \div 100,000$	95	11	7		
> 100,001	9	1	1		
Total	348	71	40		

Table 1. Classes of WWTPs for P.E.

Most of the WWTPs in operation (70%) treat wastewater of communities lower than 10,000 P.E. (person equivalent), only 3% serve urban communities greater than 100,000 P.E.: while more than 47% of the planned WWTPs serve communities with P.E. lower than 2000. A certain discrepancy is detected between the hydraulic load capacity of the existing WWTPs (of about $320 \times 106 \text{ m}^3$) and the produced volume of TW (about $222 \times 106 \text{ m}^3$). Regarding the location of WWTPs, 93 systems are in the province of Messina (North-Eastern Sicily), 73 in the province of Palermo (Western Sicily), 33 in Trapani (Northwestern Sicily), 40 in Agrigento (South-Western), 15 in Siracusa (Southeastern Sicily) and 31 in Catania (Eastern Sicily). The treatment processes consist of: preliminary treatments, employed in 314 of the Sicilian WWTPs, primary sedimentation, used in 303 of the WWTPs, activated sludge process, the most common type of secondary biological process, covering about 83% of the systems; the trickling

filter technology covers 20.5% of WWTPs. Tertiary treatments, such as coagulation-flocculation, and microfiltration processes are applied for 30% of the systems, treating wastewater for communities greater than 10,000 P.E, and the secondary sedimentation process is operated in 55.4% of WWTPs. Most of the analysed systems discharge neighbouring into rivers. Chemical-physical and bacteriological data on existing WWTPs were available just for a limited number of study cases. Data easily available consist of biochemical oxygen demand (BOD, mg L^{-1}), chemical oxygen demand (COD, mg L^{-1}) and total suspended solids (TSS, mg L^{-1}). Analyses of available data evidenced BOD concentrations generally below the threshold of 25 mg L^{-1} fixed by the Italian law (LD 152/06), BOD concentration within the range of 25–50 mg L^{-1} in 34% of WWTPs under study and concentrations over this range for 31% of the systems. COD values were below the Italian law fixed limit of 125 mg L^{-1} in 73% of WWTPs; in the 22% of the systems COD was in the range of 125-350 mg L^{-1} , and only 5% of the studied cases highly exceeded the Italian threshold fixed for discharge in water BOD₅ies. In 49% of WWTPs, TSS was below the limit of 35 mg L^{-1} fixed by the mentioned standards; 23% of the systems reported TW within the range of 35–70 mg L^{-1} and the remaining 28% greatly exceeded the fixed limit. Above discussed chemical-physical data revealed a weak pollutants removal capacity for the surveyed WWTPs; it is, thus, necessary enhancing the efficiency of these systems by introducing natural treatment systems, such as constructed wetlands and storage reservoirs.

A regional scale GIS-based procedure was implemented according to the methodology described in (Barbagallo *et al.*, 2012). The input data were:

• Land use/land cover data (Corine Project, 2006) (scale: 1:100.000); land uses were classified into five general categories: urban areas, agriculture areas, forest and seminatural area, open water bodies and wetlands. Most of the mapped area is 'Agriculture' (69%), being 'forest and seminatural' the second land use (26%). 'Urban areas', 'water bodies', and 'wetlands' habitats contribute to the remaining land surface area (Figure 5a).

•20-m resolution DTM (digital terrain model) (Italian National Geoportal, 2018) (Figure 5b);

•Characteristics of the Sicilian "irrigation public agency" (from now on named as Irrigation Consortia (IC)) (source: specific surveys at the IC); In Sicily, the water for agriculture use is mainly supplied by 11 IC, divided into Eastern IC and Western IC, with a total of 37 irrigation districts (Figure 5c). The Sicilian region, through the IC, promotes and organizes reclamation actions for water resources and environmental defence, the conservation, the valorisation, and the protection, as well as for the development of territory and the agricultural production. The irrigation area extent is about 18×10^4 hectares, of which 7×10^4 hectares are actually irrigated. Irrigation resources are manly derived from artificial reservoirs, providing about 160×10^6 m³ per year. Actually, 31 artificial reservoirs supply Sicilian irrigation networks; 3 new reservoirs are under construction and they will provide an extra water resource of about 107×10^6 m³ per year. For each district, irrigation demand was derived by multiplying the actual irrigated area (ha) and the water volume (m³ ha⁻¹) fixed by the IC for each crop during the irrigation season. By comparing the water volume availability with irrigation demand, the annual water deficits/surplus were determined for each district (Barbagallo *et al.*, 2012);



Figure 5. GIS-reclassified land use map (a), slope map of Sicily (b), and IC plus irrigation districts (c).

Data were geo-referenced at the same projection (WGS84-UTM zone 33N) and integrated with GIS spatial overlay analysis functions. TW coming from WWTPs in operation were attributed to the corresponding irrigation district with the aim to integrate its collective (by IC) water resources.

A step forward consisted on the definition of a feasibility study based on elevation and land use characteristics at Sicilian regional scale. Therefore, within GIS tools, the "cost-distance" (ArcGis, 2017) function was applied for connecting WWTPs to the irrigation districts on the basis of the nearest distance (Figure 6).



Figure 6. GIS spatial analysis and the cost-distance procedure scheme.

The hypothesis for building a feasible irrigation distribution networks connecting WWTPs with irrigation districts was conditioned by a maximum acceptable slope of 45% (Figure 5b, obtained by a DTM reclassification); the mean elevation of the irrigation districts was defined as the mean elevation of the image pixels falling into each irrigation area. The adopted rules implied also that each WWTP in operation, supplying communities higher than 5000 P.E., was assigned to just one irrigation district. Furthermore, D was the minimum GIS-based distance accordingly to the landscape morphology and the presence of natural obstacles (lakes, urban areas, etc.). On the basis of the different categories of land coverage to be crossed by the distribution networks and depending on the most favourable rout, an economic assessment of pipeline construction can be done.

In order to take into account only significant treatment systems and feasible solutions, WWTPs were selected for TW reuse accordingly to the below reported criteria:

- 1. served communities greater than 5000 P.E., with a corresponding flow rate of about 10 L s⁻¹ considering 150 L P.E.⁻¹;
- 2. maximum distance (D) between WWTP and the nearest irrigation district according to the potential volume (Q), as:
- $D \le 5 \text{ km if } 10 \text{ L s}^{-1} \le Q < 15 \text{ L s}^{-1};$
- $5 \text{ km} < D \le 10 \text{ km} \text{ if } 15 \text{ L} \text{ s}^{-1} \le Q < 30 \text{ L} \text{ s}^{-1};$
- 10 km < D \leq 20 km if Q \geq 30 L s⁻¹;
- 3. TW, exceeding the irrigation request of the nearest district, may be shared between neighbouring IC, exhibiting water deficit conditions; and
- 4. WWTP elevation higher than the mean elevation of the irrigation district or WW pumping (Δ H) up to 50 m.

When ΔH was over the fixed limit of 50 meters, the reuse possibility was evaluated case-by-case, depending on the relevance of the WWTP under study and the presence of areas, within the same district, with altitudes lower than the limit.

In the study, particular emphasis given was to microbiological concentrations found in TW to evaluate the potential risk assessment; to this end, on selected WWTPs, having a minimum dataset of available microbiological data, E. coli concentrations were analysed by comparing the quite restrictive Italian approach for TW reuse (MD 185/03) and WHO guidelines (WHO, 2006). Briefly, the latter is based on a tolerable maximum additional burden of disease of one million of a DALY loss per person per year $(1 \times 10^{-6} \text{ DALY})$ loss pppy). According to the adopted approach (Barbagallo et al., 2012), the model exposure scenario of 'unrestricted irrigation', based on the consumption of TW-irrigated lettuce (Shuval et al., 1997; Ayuso-Gabella et al., 2009), was considered. The selected scenario corresponds to the controlled use of TW to grow crops that are normally eaten raw. For this scenario, results of quantitative microbial risk analysis-Monte Carlo simulation show that the required rotavirus reduction (the pathogen with the highest infection risk compared to the other two pathogens selected in the WHO guidelines) from TW to lettuce ingestion is of 6 log units. This total reduction is achieved by both wastewater treatment and a selection of post-treatment health-protection control measures (i.e., low-cost drip irrigation techniques, pathogen die-off, produce washing and peeling, etc.). If at least 2–3 log unit reduction due to rotavirus die-off between the last irrigation and consumption is considered, TW with E. coli concentration up to 1×10^4 CFU 100 mL⁻¹ are associated with an acceptable median infection risk for rotavirus infection of 10⁻³ pppy (WHO, 2006; NRMMC,

EPHC and AHMC, 2006; Amoah *et al.*, 2007; Abaidoo *et al.*, 2010; Keraita *et al.*, 2010).

2.3 Results and Discussion

2.3.1 GIS Results on Wastewater Reuse

In Sicily, a large amount of wastewater is treated in WWTPs of small-medium size (i.e., 244 WWTPs are below 1×10^4 P.E. and 95 under 1×10^5 P.E.). The mean annual volume of TW (about 222×10^6 m³) is around 30% of the irrigation water yearly requested by the IC (about 750 $\times 10^6$ m³); TW volume could reach 340 $\times 10^6$ m³, corresponding to the available design capacity of these systems. Table 2 summarizes at IC basis the potential of RW use for irrigation purpose in Sicily.

Irrigation Consortia	Mean Altitude (m a.s.l.)	DS* (10 ⁶ m ³)	WWTP (#)	Annual TW volume (10 ⁶ m ³)	Min D (m)	Max D (m)	Mean ΔH (m)
CB1-Trapani	59.3	4.4	4	11.8	20	2,745	36.6
CB2-Palermo	311.0	5.3	21	46.6	0	15719	53.9
CB3-Agrigento	214.1	6.1	16	12.8	0	5193	40.8
CB5-Gela	155.2	0.9	6	5.7	0	14723	22.7
CB6-Enna	343.0	0.8	7	5.8	369	16651	0.0
CB7-Caltagirone	145.7	-0.5	3	6.9	1460	4380	0.0
CB8-Ragusa	106.5	-7.3	12	20.2	0	14647	62.0
CB9-Catania	181.4	42.9	7	33.0	40	13712	22.3
CB10-Siracusa	56.6	13.1	5	19.4	508	15831	43.6
CB11-Messina	537.6	0.1	1	0.6	5280	-	0.0
Tot	al	65.8	82	162.8			

Table 2. Potential availability of TW at the consortium level.

* DS: annual water deficit (+) or water surplus (-).

Within each IC, data were analysed at the irrigation district level (data not shown). Within the WWTPs database, only those systems satisfying the selected criteria were analysed. Among 37 existing irrigation districts, 25 resulted eligible for receiving TW coming from 82 WWTPs in operation (Figure 7). For the remaining areas, no feasible design or economical solutions were defined, showing these areas a limited water deficit, corresponding to about 5% of the water demand.

Within the CB2 of Palermo (Western Sicily), the San Leonardo district shows the highest number of WWTPs (14) that could be connected with; other significant cases are in the districts of Scicli (CB8 Ragusa) with eight WWTPs, Garcia-Arancio (CB3 Agrigento) with seven WWTPs, and Salso-Simeto-Ogliastro (CB9 Catania) with six WWTPs supplying resources. The remaining areas within each IC are potentially supplied by WTTPs in a number ranging from 1–5. The maximum distance (D) between the selected WWTPs and the irrigation districts varied from zero to 16.6 km.

The analysis revealed a potential volume of TW coming from plants connected with irrigation districts of 163×10^6 m³, and a deficit of water resources of 65.8×10^6 m³.

The IC of Ragusa and Caltagirone seemed to be able to completely meet the water demand for irrigation. In these cases, water surplus could be eventually used to enlarge the irrigated area, to save high quality water for other uses etc.) (civil, environmental. or to avoid aquifer overexploitation. Notwithstanding the high volume of TW that could be used in the CB9 of Catania, its water deficit remains quite high. In the case of CB9-Catania, the treated volume could be increased up to 48×10^6 m³ vear⁻¹ with the ongoing revamping of Catania WWTP.



Figure 7. Irrigation districts and the corresponding WWTPs. Same colours indicate WWTP and the irrigation district associated with.

2.3.2 Results of the Risk Assessment Analysis for Selected WWTPs in Sicily

For 11 WWTPs, the health risk associated with the use of TW was analysed by following the referred WHO DALY tool (WHO, 2006). The systems are located in Siracusa, Lentini, San Michele di Ganzaria, Giarre-Riposto, and in Caltanissetta Province (Bompensiere, Gela, Mazzarino, Mussomeli, Riesi, Serradifalco, Villarmosa, Villalba). Figure 8 reports E. coli data from the WWTPs by comparing the 2006 WHO guidelines and the Italian legislation (M.D. 185/03) thresholds. The percentage of samples showing E. coli count lower than the Italian threshold of 10 CFU 100 mL⁻¹, requested for 80% of samples, was up to about 20% for all the selected WWTPs (being null for Caltanissetta Province and 14% and 19% for Lentini and Siracusa WWTPs, respectively). In the WWTP of S. Michele di Ganzaria, 23% of samples had E. coli lower than the Italian threshold of 50 CFU 100 mL⁻¹, required for 80% of samples in the case of natural treatments (i.e., in this case WWTP is integrated with constructed wetlands for tertiary treatment). In this system, 33% of samples had E. coli above the value of 10^4 CFU 100 mL⁻¹ fixed by WHO in 2006 for the 'unrestricted irrigation' scenario; the WHO limit is intended for reaching a median design risk for rotavirus infection of 10^{-3} pppy, considering a 2–3 log units reduction due to rotavirus die-off between the last irrigation and harvest. On this regard Aiello et al. (2013) evidenced, in fact, that although E. coli content of TW is over the limits set by Italian law, the hygienic quality of the irrigated product (i.e., herbaceous crops) can be preserved, according to the WHO. Similar results were obtained by Castorina et al. (2016),

which reported values of *E. coli* concentration up to 80 CFU 100 g⁻¹ for drip and sub-drip irrigated eggplant by TW. In the other WWTPs the percentage of samples below the WHO limit decreases to 1.3% for Siracusa, 1.4% for Lentini and was null for the remaining systems.



Figure 8. Percentage (%) of samples with E. coli concentration exceeding /lower than the limit fixed by the Italian law (M.D. 185/03) (left) and by WHO guidelines (2006) (right) for the selected WWTPs in Sicily. WWTPs in decreasing PE order and with the number of total samples in brackets.

Within the database of WWTPs, used in the study to assess the potential of the reuse practice in Sicily, 14 systems were analysed in terms of physical and chemical quality of the produced TW. Table 3 summarizes these results, showing the mean annual values of the pollutant concentrations. In particular, during the monitored period 2010-2013, the percentages of samples exceeding the standard limits (MD 185/03) (i.e., TSS: 10 mg L^{-1}) varied between 25% (Enna WWTP) and 100% (Gela, Riesi, and S, Michele di Ganzaria WWTPs); in the mean of the systems, TSS was over the limit for 73% of the analysed TW samples. 19% of the samples collected from the analysed WWTPs exceeded the threshold fixed for COD (i.e., 100 mg L^{-1}); the maximum was reached in the Bompensieri (CB5) WWTP with 53% of samples. On the average of the WWTPs, 23% of samples exceeded BOD₅ limit (i.e., 20 mg L^{-1}), with a maximum (50%) in the Giarre-Riposto WWTP. The analysed Sicilian showed fairly high percentages of samples WWTPs exceeding the limits imposed by the Italian legislation for TW reuse on chemical and physical compounds. In particular, high TSS concentrations may affect the performance of the irrigation network in case of direct RW use for agriculture. To enhance TW quality characteristics, efficient cost/benefit solutions should be provided by improving the disinfection phase in existing treatment processes or introducing new eco-sustainable treatments, like natural systems. Some authors (Licciardello et al., 2018) found that the percentage of samples below the Italian threshold of 50 CFU 100 mL⁻¹, raised up to 67% and 80% by using two WW treatment options with different equipment after the constructed wetland system, the first, a natural-based solution, including a biological pond, a storage reservoir and sand and disk filters and the second,

including an ultraviolet system besides sand and disk filters. Both options ETTS allowed to match WHO guidelines (2006) for the "unrestricted irrigation" scenario, as well as the B quality class as defined in the proposed European Directive on water reuse (EC, 2018). Moreover, E. coli contamination on tomato and eggplant never reached significant values by using WW by both option systems. Based on several experiences in different Mediterranean countries, it is expected that rapidly expanding waterrecycling practices will quickly provide sustainable, lowenergy, and cost-effective options to improve water availability based on criteria of quality and recycling capacity (Ait-Mouheb et al., 2018) but to provide a more comprehensive understanding of the health risks of wastewater use in agriculture, future research should consider multiple exposure routes, long-term health implications, and increase the range of contaminants studied, particularly in regions heavily dependent on wastewater irrigation (Dickin et al., 2016).

Selected Sicilian WWTPs	Mean values of chemical-physical compounds in the effluent of WWTPs			Percentages effluent samples over the limits (%)		
	TSS (mg L ⁻¹)	$\begin{array}{c} \text{COD} \\ (\text{mg } L^{-1}) \end{array}$	$\begin{array}{c} \textbf{BOD} \\ (\textbf{mg } \textbf{L}^{-1}) \end{array}$	TSS	COD	BOD ₅
Villalba (CB5)	14	80	16	61	6	11
Mussomeli (CB3)	16	96	17	90	40	0
Bompensieri (CB5)	17	90	15	87	53	13
Serradifalco (CB5)	14	76	12	67	0	0
Gela (CB5)	19	94	17	100	46	38
Riesi (CB5)	18	89	16	100	10	0
Giarre-Riposto (CB9)	10	17	5	48	50	50
S. Michele Ganzaria (CB7)	21	28	19	100	13	35
Ragusa Consortium (CB8)	42	85	21	98	2	48
Ragusa Municipality (CB8)	31	66	23	90	0	33
Scicli-sea (CB8)	19	29	10	38	0	19
Assoro-Leonforte Consortium (CB6)	10	71	17	31	23	31
Enna (CB6)	7	47	10	25	0	32
Siracusa (CB10)	19	79	20	80	25	13
Mean values	18	67	16	73	19	23

Table 3. Chemical-physical characteristics of selected WWTPs in Sicily.

2.3.3. Barriers Limiting TW Reuse in Agriculture

TW reuse in agriculture involves a careful evaluation of regulatory, socio-economical, health-safety protection, agronomic, and technical features, which makes even the most rigorous management approach very complex. As formerly evidenced (Cirelli et al., 2008), the Italian standard limits for water reuse (MD 185/03) enclose 54 parameters to be monitored for TW quality assessment, 20% of which are the same fixed by the regulation for water human consumption (LD 31/2001) while 37% are neither considered by the same law. The microbiological limits are also so restrictive to be in line with a zero risk approach. Moreover, different alternatives among which urban non potable, industrial and agricultural TW reuse are not accounted. Similarly, no attention is paid to differentiate between restricted and unrestricted irrigation (and the crop types), as well as among the irrigation options (drip irrigation, spray irrigation, etc.).

Temporal and spatial gaps, constituted respectively by the need for TW storage during the cold seasons and the WWTPs siting with respect to the irrigation areas open to important criticisms. In the first case, the six dry months of irrigation demand require reliable reservoirs for maintaining desirable quality effluent, since the storage systems could "rarely act as perfectly mixed reactors" (Cirelli *et al.*, 2008); in the second case treatment facilities are usually too distant and altimetrically unfavourable with respect to the eventual water distribution pipelines connected to the interested areas. The Italian law establishes that the additional costs (including operation and maintenance costs) related to reuse of wastewater treatment burden on citizens, while distribution and monitoring costs burden on the final users (farmers, golf courses, etc.). This is a hampering factor for the development of TW due to the fact that the users do not have direct benefits by reusing treated wastewater, but water tariff has to be increased to recover the water reuse costs (exceeding the actual mean of $0.13 \notin /m^3$ for water supplies faced by farmers) (CSEI Catania, 2011).

Under these conditions, the total cost requested for reclamation, since the construction phase, to operation and maintenance ones, in addition to the costs for water distribution and reuse system monitoring, might result hardly determinable and really sustainable just for large WWTPs.

At the light of these considerations the key problems to be focused on are:

(i) the reliability of the "offer" (based on a strengthened quality control system for incoming wastewater to the sewerage system);

(ii) the need to guarantee the TW quality (WWTPs efficiency and the adoption of effective refinement treatments); and

(iii) the certainty of the "demand" (based on TW reuse economic convenience for integrating or replacing conventional waters, and promoting reward and tax relief systems for users' benefits).

In spite of all these barriers, perhaps, in Italy and in particular in Southern regions, TW cannot be considered an additional water resource, and their indirect reuse is widespread in many agricultural areas. Treated and untreated wastewater are in fact discharged into water bodies and taken further downstream for irrigation (in many cases only partially diluted with conventional waters).

Several strategies can be promoted to increase the effective reuse of urban TW. Among those, the collection and reuse of TW produced in urban cities located along the coasts, as well as in small and medium-sized urban sites all over the country. On this regard, meaningful examples can be found in Sicily, where about 80% of the population lives on the coastline, and about 70% of urban centres (such as in the rest of Italy) have a population of less than 1×10^4 inhabitants; all of these generally discharge into surface water bodies or directly into the sea. Additionally, the role of wastewater reservoirs, especially for small and medium urban cities located in the internal areas, can be central within a general water resources management and a wastewater reuse policy (more attention should be paid to reservoirs design, operations, and removal efficiencies). The zero risk approach for managing the health-safety issue in TW reuse, must be reconsidered because economically unsustainable, and specific interventions for minimizing the risk of infections could be adopted passing throughout accurate pre and post-harvesting microbial reduction procedures, as also suitable irrigation management and techniques depending on the specific crops.

Since the complex Italian legislation requires a strict control and monitoring of the TW reuse systems with a consequent increasing of the costs, potential users should rely on the support of private or public agencies. In fact, the Italian Irrigation Consortia should have a fundamental role in promoting the reuse of TW in relation to their legal and technical expertise.

2.4 Conclusions

The proposed GIS-based procedure confirmed its validity as a useful decision support system in TW reuse management. The implemented cost-distance spatial analysis allowed to evaluate a possible and desirable integration of WWTPs with the Sicilian collective irrigation districts, in order to increase water resources for the crop water demand fulfilment and to move conventional water resources to other uses (civil, environmental, industrial), substantially reducing water shortages. Overall, the potential volume of TW produced by the plants connected to irrigation districts is $163 \times 106 \text{ m}^3 \text{ y}^{-1}$, while water deficit is $66 \times 10^6 \text{ m}^3 \text{ y}^{-1}$. The feasibility of TW reuse in Sicily was also analyzed at the light of the microbiological risk potentially induced; data on E. coli concentrations were, in most of the analyzed cases, below the limit fixed by WHO for the contamination diffusion through the ingestion of products eaten raw. The results of the microbiological quality conflict with the more restrictive rules imposed by the Italian legislation, that may be revised also in this sense. Additionally, chemicalphysical data did not fulfil the stringent national standards, particularly in the case of TSS, confirming the need to improve the actual treatment facilities with more effective tertiary conventional technologies or natural treatment systems. The work evidenced the strategic role that could be played by the IC for reclaimed water use implementation in Sicily. However, to this aim, the economic framework should be carefully evaluated for each district in order to select, among different reuse projects, those with the greatest potential for success and to better allocate public funds.

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3 Hydraulic reliability of a horizontal wetland for wastewater treatment in Sicily³

Abstract

The purpose of this study was to evaluate how the hydraulic behavior of a horizontal subsurface wetland (HF), that is part of the hybrid wetland (hybrid-TW) of the IKEA[®] store in Eastern Sicily (Italy), influences the overall wastewater treatment performance. The HF unit experiences frequent overloading peaks due to the extreme variability in the number of visitors at the store, and after 2 years of operation it showed signals of partial clogging at the inlet area. The hydraulics of the HF unit has been monitored through measurements of hydraulic conductivity at saturation (K_s), tracer tests, and geophysical (i.e. electrical resistivity tomography-ERT) measurements carried out during the years 2016 and 2017. Results indicated a general good agreement between the performed measurement techniques. thus their combination, if adequately performed and calibrated, might be a reliable tool for detecting those wetland areas mainly affected by clogging conditions. The results also indicated that partial clogging had no significant effect on the quality of the discharged water.

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Keywords: clogging, geophysical methods, tracer test, horizontal wetland, wastewater treatment

3.1 Introduction

Effective wastewater (WW) management is essential for protecting public health, safeguarding the environment, adopting reuse strategies (Castorina *et al.*, 2016; Angelakis, 2017; Salgot 2017), and reducing the use of conventional water in agriculture (Toscano *et al.*, 2013). WW treatment includes the use of centralized plants, which collect, treat, and discharge large amounts of effluent. Construction of these systems, which include large sewage collectors, are expensive and the costs are prohibitive in areas with low population density (Barbagallo *et al.*, 2003; Barbagallo *et al.*, 2012; Ye and Li, 2009).

In the last decade, decentralized WW treatment systems, such as treatment wetlands (TWs), have been increasingly used for small communities because of their low operation and maintenance requirements (Cirelli *et al.*, 2007; Barbagallo *et al.*, 2011). These decentralized treatment systems allow the collection, treatment, and disposal or reuse of effluent close to the WW point source (Massoud *et al.*, 2009).

Thus, TWs are environmentally friendly systems that are now widely used to treat different type of WW with high range of organic and inorganic pollutants. TWs reduce these through a combination of physical and biochemical processes (Vymazal *et al.*, 2008; Avila *et al.*, 2014). TWs are reliable, cost effective, and easily managed, and are therefore important options for small settlements, individual houses, and establishments (schools, shops, restaurants, hotels), especially when they are not connected to centralized sewage systems.

Recently developed hybrid-TW systems are now used to satisfy the more stringent standards for WW treatment processes, so that the effluent can be reused or discharged into public water bodies. These systems include various types of TWs (horizontal sub-surface flow, vertical sub-surface flow, and free water surface flow) that are placed in series to increase the efficiency of the overall treatment process (Vymazal, 2005; Vymazal, 2011; Wu *et al.*, 2014). However, clogging can reduce the life span of a TW (Barreto et la., 2015; García *et al.*, 2016; Knowles 2011; Stefanakis *et al.*, 2014; Vymazal, 2018), mainly because of the reduction of hydraulic conductivity due to saturation of the TW porous media.

The biological, physical, and chemical treatment processes that occur in TWs may lead to a gradual clogging of the substrate, leading to hydraulic faults and/or reduced performance (Nivala et al., 2012). The process of clogging is inevitable due to sedimentation of suspended solids (SS) introduced with the water flow, biofilm formation in the filtering substrate, and invasion by plant roots (Vymazal, 2018). The traditional approaches used to monitor the extent and the impacts of clogging are measurements of hydraulic conductivity (K_s), tracer tests, and physical-chemical characterization of the clogging material. In general, K_s values are measured in situ to directly evaluate the severity of clogging. Tracer tests (using sodium chloride, rhodamine, or potassium bromide, fluoroscein) are used to monitor the effect of clogging on flow through the porous medium (Aiello et al., 2016). The major limitation of these two methods is the need for measurements at numerous time points. Furthermore, K_s measurements, which approximate
the saturated hydraulic conductivity of the porous medium to the measured vertical conductivity, do not consider horizontal flow. In addition, a certain amount of compaction of the substrate occurs when inserting tubes used for K_s measurements (Pedescoll *et al.*, 2011). Other disruptive methods performed on the clog permit quantification of the extent and nature of clogging in laboratory and/or in situ.

Despite the availability of different approaches, researchers have reported that no single method can provide comprehensive and quantitative assessments of clogging in TWs (Aiello et al., 2016; Nivala et al., 2012). Integrated approaches may therefore provide a better characterization of the complex hydrology and development of clogging in TWs. For example, Aiello et al. (2016) performed full-scale investigations of clogging phenomena in subsurface flow TWs by combining K_s measurements with quantification of clogging material and visualization of flow paths using tracer tests with NaCl. Other research used complementary in situ K_s measurements and rhodamine tracing experiments to evaluate the hydraulic conditions in a clogged TW used for tertiary WW treatment (Knowles et al., 2010). De Paoli and Von Sperling (2013) characterized accumulated solids using hydraulic conductivity measurements at specific points, and compared planted and unplanted wetlands. Butterworth et al. (2016) compared the performance of four full-scale aerated horizontal flow TWs in terms of removal of ammonia, removal of solids, and hydraulic conductivity, by combining tracer tests within situ measurements of K_s. However, all of the above-mentioned techniques are invasive, and involve some disturbance of the sample (Nivala *et al.*, 2012).

Some limitations in the monitoring of clogging in TWs may be overcome by use of geophysical techniques, which rely

on measurements of electric and electromagnetic properties of the porous medium. These methods are generally less disruptive than techniques that require sample extraction or taking the system offline. Moreover, geophysical surveys may provide information on a TW before clogging by detection of potential failures (Tapias et al., 2012). Recent research has shown that electrical resistivity tomography (ERT) is useful for investigation of the geometry of a subsurface flow TW, because it provides information on its internal structure, silting up, and clogging (Casas et al., 2012; Mahjoub et al., 2016). Therefore, use of ERT is a potentially effective approach for monitoring problems in TWs (Arjwech and Everett, 2015; Cassiani et al., 2006; Cassiani et al., 2015; Consoli et al., 2017; Vanella et al., 2018), such as evaluation of clogging, at high spatial resolution.

The study aims at: (i) evaluating the hydraulic performance of a horizontal sub-surface wetland (HF), part of the secondary hybrid-TW of the IKEA[®] retail store in Eastern Sicily (Italy), and (ii) observing any possible failure of the overall treatment efficiency at the hybrid TW due to the HF hydraulics behavior.

An evidence of partial clogging at the HF inlet (e.g. after two years of operation a progressive sludge matter deposition was detected) essentially motivated the study. The high fluctuation of customers visits at the store most probably caused an inadequate management of the existing conventional WW treatment system (i.e. sequential batch reactor-SBR). This was associated with an overloading of HF with untreated WW.

The hydraulics of the HF unit, accounting for partial clogging conditions, was therefore monitored by combining measurements of hydraulic conductivity at saturation (K_s)

with the use of traditional tracer tests, and geophysical surveys (ERT).

3.2 Materials and Methods

3.2.1 Hybrid-TW at the IKEA[®] store in Catania

The hybrid-TW system at the IKEA[®] store of Catania (Eastern Sicily, Italy; latitude $37^{\circ} 26'$ N, longitude $15^{\circ} 01'$ E, altitude 11 m a.s.l.) is the tertiary treatment unit for this store (Figure 9).



Figure 9. Hybrid-TW system at the IKEA® store of Catania (Eastern Sicily)

It includes a screening system and a sequential batch reactor (SBR) (Figure 10), as secondary treatment unit. This area of Sicily has a semi-arid climate, with average annual precipitation of about 500 mm, and the air temperature can reach 40 °C during summer. This SBR was designed for treatment of WW produced by toilets and the food area of the store. It has a maximum flow rate of 30 m³ day⁻¹ and a total nitrogen (TN) concentration of 135 mg L⁻¹. The

screening unit and SBR operations started when the store opened in 2013.



Figure 10. Design of the hybrid-TW system at the IKEA® store in Catania (Eastern Sicily).

The IKEA[®] Store in Catania opened in March 2013, employs 250 workers, has an average of about 6000 visitors per day, and up to 23000 visitors on some days. The store has a wide shop space, bar, and restaurants, and has significant hydraulic and organic load variability during the day and year. In particular, during holidays, pre-holidays, and weekends, WW flow rate can be 2-4 times greater than that during normal working days, and NH₄ can be greater than 200 mg L⁻¹ on these busy days.

Due to these large fluctuations of organic load, the SBR system alone was inadequate as the sole treatment system. Thus, in August 2014 a hybrid-TW system that had 3 beds in series, was added. This hybrid-TW system is discontinuously fed. It was designed to receive 30 m^3 of daily effluent from the SBR and 20 m^3 of daily effluent from the SBR and 20 m^3 of daily effluent from the screening unit, which bypasses the SBR unit during "overflow" (when the amount of WW exceeds the capacity of the SBR). In actual operation, the feeding phases did not match the design parameters. In fact, the incoming

volumes ("total inflow") strictly followed the SBR discharge phase (related to overflow volumes), which occur during a few hours of the day (discharge peaks: 12.00-3.00 p.m. and 5.30-7.30 p.m.). From August 2014 to December 2015, the SBR discharge phase had 2 cycles per day; in 2016, to improve the feeding operation of the hybrid-TW system, it was modified to 3 cycles per day; in March 2017, it was modified again to 4 cycles per day by reducing the flow rate per hour. After the startup phase, the duration of aeration was reduced. SBR and this increased the sedimentation time and inactivated the final phase of disinfection. reducing filtration and thus energy consumption by about 20%. A plot of daily total inflows during December 2016 (Figure 11) shows peaks of 50-60 m³ d^{-1} and more than 50% of overflow entering the system. The total inflow (m³ d⁻¹) in Figure 11 indicates the contribution of the SBR treated effluent ("SBR outflow") and of effluent that bypasses SBR treatment ("overflow").



Figure 11. Daily flow rates (V, m3) entering the hybrid-TW system during December 2016 at the IKEA® store in Catania.

The ratio between the SBR outflow and overflow was 1:1 in April-June 2016 and December-February 2017, 4:1 in March-April 2017, and to 6:1 in May-July 2017. The implementation of the hybrid-TW at the IKEA[®] store and the use of tertiary treatment also improved the aesthetics of the site. Thus, particular attention was given to the selection of different ornamental plants.

The first stage of the hybrid-TW is a horizontal subsurface treatment wetland (HF, Figure 10), that reduces organic matter (biochemical oxygen demand [BOD₅] and chemical oxygen demand [COD]) and suspended solids (SS) from the WW. The HF has a surface area of about 400 m² (12×34 m) and is filled with volcanic gravel (grain diameter: 8-12 mm) to a depth of about 0.60 m. The surface is planted with *Phragmites australis* (which is highly tolerant to pollutants) at a density of 3 plants per m⁻², with a ~1 m vegetated strip of *Iris pseudacorus* close to the HF outlet.

The second stage is a vertical subsurface flow treatment wetland (VF1, Figure 10) that is designed for further reduction of organic matter and SS and promotion of nitrification. The VF1 has a surface area of about 580 m² (24 m \times 24 m). and The surface is planted with *Cyperus papyrus* var. siculus and *Canna indica* at a density of 2 plants per m-2. The substrate of VF1 consists of volcanic sand (5-15 mm) on the top 0.45 m, and coarse gravel (25–40 mm) on the bottom 0.30 m.

The third stage consists of vertical subsurface wetland (VF2, Figure 10) that have the same design as VF1 (size, area, porous medium), but the vegetation consisted of Typha latifolia and *Iris pseudacorus* at a density of 4 plants m⁻², with a few *Cyperus papyrus*. rhizomes that were planted in January 2016.

The hydraulic loading rate (HLR) of the HF varies between 75 and 125 L m⁻² d⁻¹. WW effluent from the HF fills a plastic tank, where a submersible pump, equipped with a water level sensor, is located for intermittent loading of VF1 every 4 h, with a maximum flow rate of 10 m³/cycle. The WW effluents from VF1 fill another plastic tank, from which VF2 is discontinuously fed, so the HLR is the same as VF1.

3.2.2 Assessment of clogging in the HF system

3.2.2.1 The hydraulic conductivity at saturation

Evaluation of clogging at the HF unit was conducted using "falling-head method" to determine hvdraulic the conductivity at saturation (K_s, m d⁻¹) (NAVFAC, 1986; Caselles-Osorio and García, 2006; Pedescoll et al., 2009). Nine measurement points were arranged in 3 rows, and small holes were dug into the granular medium until the water table was reached (Figure 12). The device consists of a steel tube (internal diameter: 0.10 m, length: 1.5 m), and it was inserted into the medium using a mallet to a depth of about 0.25 m. A pressure probe (Sensor Technik Sirnach, AG), connected to a computer using a data logger (CR200-R, Campbell Scientific) was positioned inside the steel tube. The tube was filled with water in pulse mode, the decrease of water height inside the tube was monitored until the water level reached the HF water table, and pressure data were then converted into water heights. The experimental monitoring time was 60 s for each test, and the pressure probe was configured to collect 2 water height measurements per s. The hydraulic conductivity at saturation (K_s, m d⁻¹) was calculated using a formula described in NAVFAC (1986), which was obtained by combining the mass conservation principle with Darcy's law:

$$K_s = \frac{2\pi R + 11L}{11t} \ln\left(\frac{H_1}{H_2}\right)$$

(1)

where R is the radius of the tube (m), L is the submerged length of the tube (m), t is time (s), and H1 and H2 are the water levels (m) in the permeameter cell at times t1 and t2 (s), respectively.

To obtain the best fit of modeled H2 (Eq. 2) and measured H2, a least squares procedure was used to estimate K_s , using an iterative, nonlinear procedure in Excel solver (Frontline Systems, Incline Village, NV):

$$\sum_{t=0}^{n} \left(H_{obs}(t) - H_{est}(t) \right)^{2} \to 0$$
(2)

where H_{obs} is the water table height (m) inside the tube at time t during the test and H_{est} is the modelled data from Eq. (2).

 K_s was monitored during 2016 (June 15) and 2017 (January 31, April 27 and July 21) at the points indicated in Figure 12. K_s data (4 replicates per measurement point) were recorded along 3 transects (1-2-3; 4-5-6; 7-8-9) that were transverse to the main direction of flow, and along the three transects (1-4-7; 2-5-8; 3-6-9) that were parallel to the main direction of flow.



Figure 12. Setup of the HF unit showing the locations for measurement of hydraulic conductivity at saturation, tracer tests, and ERT surveys.

3.2.2.2 Residence time distribution

Under ideal plug-flow conditions, all water parcels move through the wetland at the same velocity, and reach the exit at the same time. This exit time is referred to as the nominal hydraulic residence time (nRT), which can be calculated as:

$$nRT = \frac{V}{Q}$$

(3)

where V (m³) is the flow volume in the system and Q (m³ h^{-1}) is the system volumetric flow rate.

In practice, plug flow, in which there is a single nominal residence time, is difficult to measure due to diffusion, turbulence, and mixed flow, which are common in wetlands (Holland *et al.*, 2004; Wahl *et al.*, 2010). Therefore, analysis of the retention time distribution (RTD) function from a tracer test is most commonly used to assess the hydraulic performance of TWs (Barbagallo *et al.*, 2011; Aiello *et al.*, 2016; Guo *et al.*, 2017). In this study, sodium chloride (NaCl) was used for tracer tests in the HF unit of the hybrid-

TW system. The tracer solution was prepared in a bucket by adding 50 kg of NaCl to 150 L of water, followed by complete mixing to dissolve the tracer. Then a pump was used to pulse-inject the tracer solution (in about 10 min) into the inlet pipe of the HF system. WW electrical conductivity (EC) was measured and recorded using 10 conductivity probes (Delta OHM – HD 2106.2) at the hybrid-TW outlet and at 9 sampling points within the HF using piezometers (Figure 12). Measurements began few minutes before the injection, and were recorded every 15 min for the whole duration of the test.

The EC values (μ S/cm) were converted to NaCl concentrations (mg L⁻¹) using a linear calibration curve (R2 = 0.99), after subtraction of background EC. HF volumes were measured at the outlet using a flow measurement device. No rainfall occurred during the tracer testing.

The hydraulic parameters of the HF unit were derived from the RTD using the method of moments (Martinez and Wise, 2003, Wang and Jawitz, 2006). The first moment of the RTD function, the centroid of the curve, gives the average time of effluent in the HF, also defined as actual residence time (aRT, h). This was calculated after subtraction of the NaCl background as:

$$aRT = \frac{\int_{0}^{\infty} t \cdot C_{out}(t) dt}{\int_{0}^{\infty} C_{out}(t) dt}$$

(4)

where t is the sample time (h), dt is the time between the adjacent data point (h), and C(t) is the outlet tracer concentration at time t (mg L⁻¹). Then, the total tracer mass

at the HF outlet was determined as described by Whitmer *et al.* (2000):

$$M_{out} = \int_{0}^{\infty} Q_{out}(t) \cdot C_{out}(t) dt$$
(5)

where M_{out} is the total recovered tracer mass at the outlet (kg), $Q_{out}(t)$ is the outflow rate at time t (m³ h⁻¹), C(t) is the tracer concentration at time t at the outlet (mg L⁻¹), and t is the sample time (h). The relative tracer mass recovery (%) was defined as:

Mass recovery =
$$\frac{M_{out}}{M_{in}} 100$$
 (6)

where Min is the total mass of NaCl added to the HF inlet (kg), in this case 50 kg.

Finally, to produce a more general and comprehensive measure of the hydrodynamic conditions in the HF unit, the hydraulic efficiency parameter (λ) (Persson *et al.*, 1999) was calculated as:

$$\lambda = \frac{t_p}{nRT} \tag{7}$$

where t_p is the time of maximal tracer concentration (h). Higher λ values indicate suitable HF utilization, and the largest amount of WW in the wetland retention process (Persson and Wittgren, 2003; García *et al.*, 2004; Guo *et al.*, 2015; Rengers *et al.*, 2016).

In this study, two tracer tests were conducted during 2017 within the HF unit, from January 31 to February 7 (Tracer Test 1, TT1) and from May 8 to May 12 (TT2). In TT2

some EC data were not recorded due to unexpected malfunction of the probes located in piezometer 7, 8 and 9. In order to compare the RTD curves for both tracer tests, only the common sampling points were considered (piezometer 1-6 and outlet).

<u>3.2.2.3 The electrical resistivity tomography for</u> <u>identification of clogging</u>

ERT (Binley and Kemna, 2005) is a non-destructive technique that used to assess the reliability of the monitoring of clogging in the HF unit. This is an active-source geophysical method, in which a low-frequency electrical current is galvanically injected into the ground between two electrodes, and the potential between the electrodes is measured. In two-dimensional (2-D) surveys (as in the present study), many combinations of transmitting and receiving electrodes are repeated along a line, providing a cross section of electrical resistivity (ER, Ω m; inverse of EC) of the subsurface. The general goal of ERT is to derive the distribution of electrical properties and produce a resistivity model that satisfies the observations of transfer resistance (ratio of measured potential difference and applied current) for different electrode configurations within a specified tolerance. In this study, ERT measurements were used to identify horizontal and vertical variability of the HF flow distribution over the examined profile. ERT was also used to derive information on effluent lateral losses and potential clogging, which produce anomalies in the distribution of ER. At the HF unit, 2-D ERT surveys were performed in 2016 (August 4 [ERT1] and September 21 [ERT2]) and in 2017 (May 19 [ERT3] and July 14 [ERT4]) (Figure 12).

For each ERT survey, stainless steel electrodes (length: 0.15 m length, width: 0.03 m) were placed directly on the surface of the HF substrate at a spacing of 0.1 m. The lengths of ERT1 and ERT2 were 7.1 m, and the profiles were located at the HF inlet, at 2 m from the inlet, and at 3.5 m from the right side of the HF. ERT data were collected using a linear array of 72 electrodes. The lengths of ERT3 and ERT4 were 16.8 m and the profiles were at the center of the HF system (between piezometers 2 and 8) and parallel to the main flow (Figure 12). In this case, ERT data were acquired using the roll-along technique by combining three composite lines, each with 72 electrodes and with 1/3 overlap.

A ten-channel resistivity meter (Syscal Pro 72 Switch by IRIS Instruments) was used for data collection. For each measurement cycle, the pulse duration was 250 ms and a target of 50 mV was used for potential readings. A dipoledipole scheme was used for all 2-D ERT data acquisitions (72 electrodes simultaneously), leading to about 5000 direct and reciprocal measurements for estimation of measurement error (Binley et al., 1995; Daily et al., 2004). Each 2-D ERT survey lasted about 25 min. Datasets passing the error threshold of 10% between direct reciprocal measurements were processed using ProfileR (Binley 2003). The performance of the inversions from resistance to resistivity was evaluated by counting the number of iterations needed to reach the solution, the number of data points used, and the root mean square (RMS) error between measured and modeled resistance values.

The ER data are presented as box-plot, with separation of data into quartiles, at 0.5 m longitudinal intervals from the inlet to the end of the ERT profile. Box-plots of ER data are also presented for intervals of depth from the HF surface to

the bottom (0-0.05 m, 0.05-0.1 m, 0.1-0.15 m, 0.15-0.2 m, 0.2-0.25 m, 0.25-0.35 m, 0.35-0.45 m, and 0.45-0.6 m).

3.2.3 Hybrid-TW wastewater sampling campaign

WW quality at the hybrid-TW system was monitored 2 or 3 times per month from April 2016 to July 2017 at the inlet and the 3 outlets of each stage (HF, VF1, and VF2). Standard methods (APHA, 2005) were used for analysis of WW quality. The following parameters were measured: total suspended solids (TSS, mg L⁻¹) at 105°C, BOD₅ test run for 5 days (BOD₅, mg L⁻¹), COD (mg L⁻¹), P as PO₄ (P-PO₄, mg L⁻¹), N as NH₄ (N-NH₄, mg L⁻¹), TN (mg L⁻¹), N as NO₃ (N-NO₃, mg L⁻¹), and *E. coli* (CFU/100 mL).

The hybrid-TW efficiency was evaluated as percent removal efficiency (RE, Eq. 1) for each parameter, and as log10 reduction of CFUs for *E. coli*:

$$RE = \frac{C_{in} - C_{out}}{C_{in}} \times 100$$
(8)

where Cin (mg L^{-1}) and Cout (mg L^{-1}) are the pollutant concentrations at inlet and outlet, respectively.

Due to its buffer capacity for the frequent organic and hydraulic loads entering the TW system, the HF unit plays a strategic role to the hybrid-TW efficiency and reliability improvement.

3.3 Results

3.3.1 Hydraulic conductivity at saturation

Figure 13 and 14 show the hydraulic conductivities at saturation (K_s , m d⁻¹) measured within the HF unit during different monitoring periods.

Our data show that K_s tends to increase from the inlet to the middle of the HF unit, and is slightly lower at the HF outlet (Figure 13: a, b, d), with the exception of April 2017 (Figure 13c) in which there was no decrease at the outlet. The pattern is clearly depicted in Figure 14, where K_s data were averaged at the inlet, middle, and outlet zones, with values of 1760, 4079 and 3467 m d⁻¹, respectively. Mean data resulted slight different at the middle and outlet zones of the unit, and in any case of the same order of magnitude.

In January 2017, K_s clearly decreased in all monitored points, and sample points 1 and 2 (close to the inlet) had the greatest reduction. This may be because of the high organic load that entered the hybrid-TW at the end of 2016; in fact, the SBR system was often by-passed due to the high volumes of WW produced at the IKEA[®] store at this time, and this caused a large quantity of organic matter and solids to enter the HF unit. In April 2017, an inlet area of about 80 m² was manually cleaned and an evident layer of organic matter in decomposition (i.e. sludge) was completely removed and disposed. The HF unit at inlet was further cleaned by a high-pressure hydro-jet (about 800-1000 kPa). The wastewater distribution pipeline at inlet was also upgraded (posed on the top of the filtering media) in order to enhance the WW distribution uniformity at the unit. After the described maintenance operations, K_s values continued to increase overall (Figure 14) and remained more similar over time (Figure 13: c).



Figure 13. Hydraulic conductivity at saturation (K_s) at the different sample points within the HF unit during June 2016 (a) and January 2017 (b), April 2017 (c), and July 2017 (d).



Figure 14. Average values of hydraulic conductivity at saturation (K_s) at the HF inlet, middle zone, and outlet during 2016 and 2017.

3.3.2 Tracer tests

Figure 15 shows the retention time distributions (RTDs) of total dissolved solids (TDS) at the HF outlet based on hydraulic tracer tests. The overall percentage of tracer recovery was 85% for TT1 and 70% for TT2, indicating good reliability (Kadlec and Wallace, 2009). The lower recovery for TT2 was probably due to the shorter duration of this test. During TT1 measurements, the average flow rate was 18 L min⁻¹, corresponding to a nominal residence time (nRT1) of 88 h (based on an average depth of 0.60 m and a constant porosity of 0.4). The TT1 measurements lasted for 164 h, nearly 2-fold more than the nRT1, to guarantee the integrity of the data (BOD₅ in *et al.*, 2013). The actual residence time (aRT1), calculated from firstmoment analysis, was 64 h; this is 24 h less than the nRT1, and suggests that flow may not include the entire volume of the HF unit (Kadlec and Wallace, 2009; Seeger et al., 2013). The total sample time for TT2 measurements was approximately 140 h, and the average flow rate was 17 L min⁻¹. The RTD curve at the HF outlet during TT1 had multiple peaks (Figure 15a), possibly indicating some shortcircuiting in the wetland, and suggesting heterogeneity in water velocity (Thackston et al., 1987; Knowles et al., 2010; Kaplan et al., 2015). We believe the most plausible reason for these multiple peaks is the intermittent loading of the HF unit, in which there are peaks at intervals of about 20 h.

TT2 measurements were performed after some hydraulic adjustments and maintenance interventions (removal and replace of filter gravel in the inlet area), and these led to a more uniform RTD (Figure 15b). The aRT2 was 88 h and the nRT2 was 92 h, based on an average flow rate of 17 L min⁻¹. The first peak of tracer was at 51.5 h for TT1 and 60 h for TT2, and each value was less than the corresponding

nominal hydraulic retention time (nRT1: 88 h, nRT2: 92 h). The hydraulic efficiency (λ) was 0.6 for TT1 and 0.7 for TT2. These are satisfactory based on the classification of Persson *et al.* (1999) (good: $\lambda \ge 0.75$, satisfactory: $0.5 \le \lambda < 0.75$, and poor: $\lambda < 0.5$).



Figure 15. Tracer concentration (NaCl) at the HF outlet during TT1 (a) and TT2 (b).

The RTDs measured by the piezometers, located inside the HF during TT1 (Figure 16: a1, a2, a3) and TT2 (Figure 16b1, b2, b3), also provided important information. In particular, the three RTDs in piezometers 1, 2, and 3 (located close to the HF inlet) exhibited lower EC values during TT2 than TT1, and the time peaks of the tracer concentration were much later during TT2 than TT1. These differences could be related to the removal of material near the inlet.



Figure 16. RTD curves measured in the HF using piezometers during TT1 (a1, a2, a3) and TT2 (b1, b2, b3).

3.3.3 Electrical resistivity tomography

The data acquired by ERT had good quality, as confirmed by the RMS errors (Table 4).

Table 4. Performance of the inversion for ERT1, ERT2, ERT3,and ERT4.

Survey	n. of measurements	n. of iterations	RMS
ERT1	649	3	0.87
ERT2	1637	2	0.89
ERT3	3328	3	0.76
ERT4	5717	3	0.78

Figures 17a and 18a show the ER images (Ω m) for ERT1 (August 4, 2016) and ERT2 (September 21, 2016) at the HF unit. Overall, the surveys had a difference of about 16% in ER values (ERT1: 32 ± 23 Ω m, ERT2: 38 ± 16 Ω m).

The ERT2 medians were 28.7% greater than those of ERT1 (Figures 15b and 16b). One ER anomaly occurred 5 m from the HF inlet, in which ERT1 values were higher than ERT2 values. Generally, ERT1 and ERT2 had lower ER values in the first 3 m of the longitudinal profile, suggesting more conductive (clogged) zones. In fact, in situ observations indicated deposition of organic matter, settleable solids, and lower vegetation density in the same zone.

Analysis of ER variability as a function of HF depth indicated similar ER medians for ERT1 and ERT2 (Figures 17c and 18c). More specifically, below 0.25 m from the top, the median ERs tended to decrease. In the first 0.25 m, the mean ER was 40 (\pm 19) Ω m in ERT1 and 43 (\pm 33) Ω m in ERT2. At greater depths, the mean ER as 23 (\pm 6) Ω m in ERT1 and 16 (\pm 5) Ω m in ERT2.



Figure 17. 2-D ERT1 profile (a), longitudinal profile of ER (b), depth profile of ER (c).



Figure 18. 2-D ERT2 profile (a), longitudinal profile of ER (b), depth profile of ER (c).

Figures 19a and 20a show the ER images (Ω m) for ERT3 (May, 2017) and ERT4 (July 14, 2017) at the HF unit. The mean ER value was 76 (± 139) Ω m for ERT3 and 25 (± 28) Ω m for ERT4. Data for the top 0.15 m (higher ER values in ERT3) were excluded due to an excessive presence of air that disturbed the measurements.

Analysis of the longitudinal profiles of ER (Figures 19b and 20b) show stable and homogenous ER values, with an average of 21 (\pm 3) Ω m for ERT3 and 19 (\pm 2) Ω m for ERT4. Conditions were also similar in the depth profiles (Figures 11c and 12c), with an average value of 22 (\pm 5) Ω m for ERT3 and 19 (\pm 1) Ω m for ERT4.



Figure 19. 2-D ERT3 profile (a), longitudinal profile of ER profile (b), depth profile of ER (c).



Figure 20. 2-D ERT4 profile (a), longitudinal profile of ER (b), depth profile of ER (c).

3.3.4 Removal efficiency of the hybrid-TW

and 6 show the physical-chemical Tables 5 and bacteriological concentrations of the hybrid-TW system for April–June 2016 and December–February 2017 (periods I and II) and March-April 2017 and May-July 2017 (periods III and IV). Each period has 6 to 9 WW samples. These Tables also show the limits imposed by Italian regulations for WW discharge into water bodies (LD 152/06) and for agricultural reuse (MD 185/03). Table 7 shows the removal efficiency (calculated from Eq. 1) of the whole hybrid-TW. The high variability of pollutant concentrations at the inlet of the hybrid-TW reflects the large number of customers who visit the IKEA[®] store and produce WW, which can be 3-fold greater on weekends and holidays. In particular, on these busy times, the hybrid-TW often receives WW directly from the screening unit, without passing through the SBR, due to its frequent overload, and therefore has lower quality.

The maximal levels of organic matter (COD, 643.7 mg L⁻¹ \pm 304), BOD₅, 321.9 mg L⁻¹ \pm 152) and TSS (162.6 mg L⁻¹ \pm 25) were during period II. The levels of these same parameters were lower during periods III and IV because of the reduced incoming overflow to the hybrid-TW and the increased settling time in the SBR system. The hybrid-TW units provided efficient reduction of TSS (up to 99.3% \pm 0.4), COD (up to 92.7% \pm 6.8) and BOD₅ (up to 96.6% \pm 3). It is likely that the use of coarse filter material, ranging from 5 to 20 mm in diameter, affected the reduction of organic matter and TSS (Dotro, *et al.*, 2017). Furthermore, the density of the macrophyte root systems growing in the hybrid-TW units may have improved the efficiency of TSS reduction by induction of settling and filtration processes (Brix, 1997; Korkusuz *et al.*, 2005; Toscano *et al.*, 2015).

The major parts of TSS, COD and BOD₅ reduction took place in HF (first stage) (Tables 5 and 6) thus reducing the clogging problem in the other stages. Vertical units contributed (data not showed) to further decrease of organics and TSS, producing a final effluent usually complied with the reuse and discharge standards.

The hybrid-TW system had a high rate of total nitrogen reduction (up to $69.5\% \pm 17$, Table 7), confirming its efficient ammonification and denitrification. Ammonia was oxidized to nitrate at the vertical stages (data not showed), thus quite high amount of nitrate was found at the outlet, generally over the discharge limit. This could be reduced recirculating the effluent to the primary treatment. These measurements of effluent suggest an inversion in the HF unit. In other words, based on measurements of N as (NO₃) at the HF outlet, the unit changed from being a mostly reducing environment (during periods I and II) to being a mostly oxidizing environment (during periods III and IV). This may have been due to the lower water level in the HF during maintenance operations. The removal efficiency of total phosphorous (TP) concentration was low during periods I and II, but increased up to $54.4\% \pm 1$ during periods III and IV (Table 6). The limited efficiency of TP reduction during periods I and II may be due to the composition of the filter medium. Vymazal (2005) reported greater P elimination rate in fine-grained soils due to their greater cation exchange capacity, but fine-grained soils are not currently used for HF systems because of their poor hydraulic conductivity. Vymazal (2005) reported that aerobic conditions favor P sorption and co-precipitation. Thus, the peak in P reduction that we observed could be explained by temporarily aerobic conditions in the HF unit, and this is corroborated by the high level of nitrates (Table 6). The effluent concentrations of TP was in most of the case below the lower limits for discharge in fresh water.

A considerable improvement in microbiological water quality was achieved at the hybrid-TW. In fact, the influent *E. coli* concentration was reduced meanly of 2-3 log unit in the HF and other 1-4 in the VF1 and VF2 (data not showed), obtaining an overall *E. coli* reduction of 4-6 log unit, from TW influent to TW effluent. **Table 5**. Means (standard deviations) of physical-chemical (mg L-1) and bacteriological (CFU 100 mL-1) parameters at the hybrid-TW inlet and outlet, and at the HF unit during monitoring periods I (April-June 2016) and II (December 2016–February 2017).

	April – June 2016(Period I)			December 20	Italian	Italian		
	Hybrid-TW Influent	HF outlet	Hybrid- TW outlet	Hybrid-TW Influent	HF outlet	Hybrid-TW outlet	WW discharge limit ⁽¹⁾	WW reuse limit ⁽²⁾
TSS	81.9 (±14)	11.5 (±0.7)	3(±1.4)	162.6(±25)	15.5(±0.7)	1.2(±0.1)	80	10
BOD ₅	226.5(±66)	9.8(±3.2)	14(±5.5)	321.9(±152)	57.5(±2.8)	31.6(±8.8)	40	20
COD	430.6(±144)	46.6(±44.0)	30(±25)	643.7(±304)	115(±5.7)	62.5(±18)	160	100
TN	85(±33)	38.3(±1.7)	28.9(±3.2)	140(±30)	72.5(±7.8)	44(±0.3)	-	15
$N-(NH_4)^+$	27.6(±33)	20.4(±8.9)	0.6(±0.3)	77.7(±18)	44.5(±13.4)	4.2(±5)	15 ⁽³⁾	2
N-(NO ₃) ⁻	23.1(±10)	$1.8(\pm 1.6)$	17.9(±7.8)	46(±7)	0.2(±0.1)	39(±0.2)	20(3)	-
TP	8.3(±3)	14.8(±5.4)	8.5(±1.2)	22.1(±0.1)	29(±9.9)	21(±2.8)	10	2
E. coli	4.7×10^{5}	1.9×10^{3}	35×10 ⁻¹	3.1×10^{6}	6.7×10^3	2.5×10^{2}	$5.0 \times 10^{3(4)}$	$100^{(5)}$

Footnotes for Tables 5 and 6: (1) Ministerial Decree 185/2003; (2) Legislative Decree 152/2006; (3) Limit for discharge into surface water bodies; (4) Recommended value for P.E. > 2000; (5) Maximum value in 80% of samples.

Table 6. Means (standard deviations) of physical-chemical (mg L-1) and bacteriological (CFU 100 mL-1) parameters at the hybrid-TW inlet and outlet, and at the HF unit during monitoring periods III (March-April 2017) and IV (May-July 2017).

	March - April 2017 (Period III)			May - July 20	17 (Period IV	Italian WW			
	Hybrid- TW Influent	HF outlet	Hybrid- TW outlet	Hybrid-TW Influent	HF outlet	Hybrid- TW outlet	discharge limit ⁽²⁾	Italian WW reuse limit ⁽¹⁾	
TSS	63 (±37)	32 (±8.5)	17.5 (±3.5)	37.8 (±16)	14.5 (±13)	8.1 (±7.1)	80	10	
BOD ₅	83.8 (±28)	12.9 (±1.2)	3.2 (±3.1)	87.4 (±66)	42.4 (±69.1)	37.2 (±69)	40	20	
COD	161.6 (±3)	27 (±1.4)	14 (±6.4)	187.7 (±129.8)	79 (±125)	66.5 (±120)	160	100	
TN	108 (±30)	60.7 (±48.5)	35.5 (±27.5)	80.3 (±6)	43.5 (±21)	30.8 (±2.5)	-	15	
$N-NH_4$	26.7 (±14)	1.8 (±1.8)	0.5 (±0.2)	14.8 (±6)	0.6 (±0.6)	0.3 (±0.2)	15 ⁽³⁾	2	
N-NO ₃	65.9 (±41)	45(±42.4)	30 (±25.5)	57.9 (±10)	21.4 (±17.6)	28.1 (±3)	20 ⁽³⁾	-	
ТР	12.9 (±1.9)	3.2 (±3)	5.9 (±0.9)	9.3 (±2)	5.7 (±0.6)	4.9 (±0.5)	10	2	
E. coli	1.03×10 ⁶	6.7×10 ³	1.26×10^{2}	4.6×10 ⁵	5.2×10 ³	2×10^{1}	5.0×10 ³⁽⁴⁾	100 ⁽⁵⁾	

The microbiological reduction rates were 1.7-4 log units (Table 7), and the *E. coli* concentrations were generally acceptable, according to the stringent limits of Italian legislation for WW agricultural reuse (M.D. 185/03). The RE values of the whole hybrid-TW system provide evidence of the important role of the HF unit for reduction of pollutants (Table 7). This high RE may be due to the higher physical-chemical and microbiological concentrations of the influent entering the system, as well as the performance of the HF unit.

Table 7. Mean removal efficiencies (RE, % for chemical parameters and as log10 reduction of CFUs for E. coli) \pm standard deviations of the HF stage and the total hybrid-TW during the 4 monitoring periods. (*Not significant)

	April-June 2016		Dec 2016-Feb 2017		March-	April 2017	May-Ju	ly 2017
	(Period I)		(Period II)		(Period III)		(Period IV)	
	HE outlot Hybrid TW		HF outlot	Hybrid	UF outlot	Hybrid	HF	Hybrid
	HF outlet	outlet	HF outlet	TW outlet	nr outlet	TW outlet	outlet	TW outlet
TSS	93.2 (±9)	95.8 (±1.4)	90.3 (±2)	99.3 (±0.4)	43.3 (±20)	68.4 (±13)	49.4 (±6)	73.1 (±33)
BOD ₅	92.3 (±2)	93.2 (±3.6)	79.7 (±11)	88.3 (±8)	84 (±4)	96.6 (±3)	66.9 (±35)	72.9 (±38)
COD	88.4 (±13)	92.7 (±6.8)	79.2 (±10)	89 (±6)	83.3 (±1)	91.6 (±4)	69.7 (±32)	76.4 (±32)
TN	40.3 (±10)	55.1 (±7.1)	47.6 (±6)	67.8 (±7)	48 (±30)	69.5 (±17)	45.5 (±52)	61.4 (±6)
$N-(NH_4)^+$	*n.s.	78.2 (±30.8)	38.9 (±32)	95.3 (±5)	94.3 (±4)	98.3 (±0.1)	95.2 (±3)	96.9 (±3)
N-(NO ₃) ⁻	93.6 (±5)	20.7 (±8.3)	99.6 (±0.2)	14.3 (±13)	40.2 (±28)	58.5 (±13)	64.8 (±25)	50.2 (±11)
TP	*n.s.	26.7 (±11.2)	n.s.	5.1 (±14)	76.4 (±20)	54.4 (±1)	37.9 (±10)	46.3 (±9)
E. coli	1.4(±1)	4(±0.7)	1.6 (±1)	2.9(±1)	1.2 (±0.1)	2.9 (±0.3)	1.6 (±2)	1.7 (±2)

3.4 Discussion

The evaluation of the hydraulic behaviour of the HF treatment unit during partial clogging conditions, carried out by performing traditional hydraulic surveys (K_s and RTD) and geophysics techniques (ERT), allowed to overview the system performance by examining its variability in space and time.

The first point to be evidenced is that the obtained results from the three different methods have been consisted each other, confirming their sensitivity in detecting clogging phenomena. This reliability was pointed out by several applied researches mainly for K_s and RTD, but represents a relatively novelty for ERT.

In particular, A comparison of the K_s results with the tracer test results indicated that in January 2017 (when a high organic load entered the HF unit due to bypass of the SBR), K_s values at the HF inlet (piezometer 2) were about 1 order of magnitude lower than on the previous monitoring date. At the same time, the TT1 had peaks at the same piezometer. This may be because of some clogging in the first part of HF, which leads to channelling of flow. In contrast, the TT2 had no peaks before 20 h, and in some case they occurred even before in the middle part of the HF unit. Before TT2, the HF inlet was cleaned, so most of the clogging may have reached the central zone of the HF, thereby improving flow at the inlet. In fact, K_s values increased again in April 2017. In May 2017, the RTD curves confirmed the good hydraulic efficiencies of the HF.

Thus, our results indicate the ERT technique is a valid tool for detecting anomalies in ER patterns that may be associated with changes in the conductivity capacity of the

medium. Variability in the texture and water saturation of the HF substrate are not responsible for differences in temporal changes in ER observed between the ERT surveys, because the HF substrate was fully saturated and the substrate was uniform in size. Instead, our results show a link between the ER and the chemical conditions in HF substrate. In fact, we attribute the lower ER values in the ERT1 and ERT2 profiles (near the inlet) to the higher organic load. This is in accordance with the laboratory experiments of Abdel et al. (2004), who compared biotic and abiotic sand columns, and reported a temporal increase in EC (lower ER) in the biotic column (nutrient, diesel, and bacteria) due to microbial processes. Furthermore, Moreira et al. (2014) reported low ER values corresponded with areas in which there was accumulation of organic matter, due to the higher levels of dissolved salts. The more similar ER values that we recorded in the ERT3 and ERT4 profiles (after removal of clogging matter, in April 2017) were not affected by the development of bacterial biofilm and organic matter. Thus, our ERT measurements confirmed the satisfactory hydraulic performance of the HF unit. However, ERT produces some data "noise" and/or numerical artifacts, so more studies are required to quantify how microbeinduced redox processes contribute to changes in ER (Abdel et al., 2004). Forquet & French (2012) confirmed that simple surface ERT measurements can provide good estimates of filtering areas, but additional methods are required for vertical measurements to detect clogging. In any case, the interpretation of ERT results requires further knowledge of the relationship between electrical properties and water content of the system, and of water conductivity and biological clogging in typical filter media (Forquet et al., 2007).
The results of the present study show that partial clogging did not reduce the capacity for removal of organic matter and suspended solids of the HF system under study, in agreement with the results of Vymazal (2018). The severity and extent of clogging depend on inflow loading, and when the system is not overloaded, the clogging is slow and is restricted to the inflow zone.

We obtained high removal efficiencies in many cases, with an overall quality of the effluent acceptable according to the Italian legislation limits (L.D. 152/06 and M.D. 185/03. It should be noted that the zero-risk approach (US EPA 1973, Seder and Abdel-Jabbar, 2011), which complies with Italian regulatory standards, requires high costs for installation and for operation and management, and use of a disinfection system, such as ultraviolet irradiation. In contrast, WHO guidelines (WHO, 2006) suggest a "calculated risk" approach, based on the presence of not more than 1000 fecal coliform (FC) bacteria per 100 mL for water that is reused for irrigation. The Italian regulations are stricter than the WHO regulations (Cirelli et al., 2008; Salgot et al., 2017). Following of these regulations requires highly intense treatments for the reuse of WW, and this reduces the competitiveness of reuse projects.

HF units are not suitable for ammonia reduction due to the lack of oxygen in their filtration beds (Vymazal, 2018). In spite of that, aerobic conditions occur in some circumstances, and this allows nitrification within the HF unit. Nevertheless, denitrification predominated during the entire monitoring period. In fact, as described by Dotro *et al.* (2017), denitrification can be very effective in HF wetlands because the effluent has sufficient amounts of nitrate and carbon. The efficiencies of nitrate and ammonia removal in the HF unit were variable, but efficiency of TN

removal at the HF outlet and at the final hybrid-TW outlet remained steady, despite the high fluctuations of the nitrogen load from the incoming effluent. In this regard, Schmitt *et al.* (2015) noted the need for further studies to better estimate the relationship of N balance with physical and chemical conditions.

3.5 Conclusion

We analysed the hydraulic reliability of an HF unit that is part of the hybrid-TW at the IKEA[®] store in Catania, and its influence on the overall performance of the system to treat wastewater. Our major conclusions are:

- the HF unit had signs of partial clogging at the inlet zone after about 2 years of operation;

- the partial clogging of the HF was confirmed by in situ measurements of hydraulic conductivity at saturation (K_s) ;

- the beneficial effects of maintenance and restoration operations on the inlet filtration zone were clearly identified by tracer tests and ERT measurements;

- the HF system consistently reduced pollutants during the monitoring period, and had no detrimental effects due to partial clogging during late 2016 and early 2017; and

- the clogging of the HF is a consequence of inflow overloading.

Finally, the overall treatment efficiency of the hybrid-TW was not particularly affected by the HF unit clogging and the hydraulic overloading with untreated effluents. The hydraulic monitoring of the HF stage and the consequent maintenance operations, allowed the hybrid-TW to respect for the most the Italian standards for WW discharge.

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The authors have contributed with equal effort to the realization of the manuscript.

4 On the performance of a pilot hybrid constructed wetland for stormwater recovery in Mediterranean climate⁴

Abstract

The overall efficiency of a pilot-scale hybrid-constructed wetland (H-CW), located on a retail store's parking area in Eastern Sicily, for alternative treatment of stormwater runoff and of sequential batch reactor (SBR) effluent was evaluated. Experimental activities were focused on system performances, including wastewater (WW) quality and hydraulic monitoring. System design, macrophyte growth and seasonal factors influenced the pilot plant performance. Very high removal efficiency for microbial indicators were reported within the subsurface horizontal flow unit (HF) playing a strategic role for Clostridium perfringens. The algal growth occurred in the free water surface (FWS) unit inhibited removal efficiencies of total suspended solids (TSS), BOD₅ and COD, impairing water quality. The whole H-CW showed good efficiency in trace metals removal, especially for Pb, Zn, Cu. Preliminary results suggested the

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reliability of the H-CW technology in decentralised water treatment facilities for enhancing water recovery and reuse.

Keywords: Alternate feeding; decentralised treatment; hybrid constructed wetland; stormwater; hydraulic conductivity

4.1 Introduction

Constructed wetlands (CWs) are green treatment technologies simulating natural wetlands. They have been traditionally used to treat conventional wastewater (WW), but during last two decades their application also included industrial and agricultural wastewaters, landfill leachate, as well as stormwater runoff (Vymazal, 2011). In particular, in recent years, CWs have become more common for urban stormwater treatment in order to remove contaminants, that would be potentially detrimental to the receiving water ecosystem. In many countries (U.S.A., Australia, Malaysia, etc.) the CWs were suggested among the best management practices (BMPs) for stormwater treatment. The European Union (EU) has clearly implemented water protection in its environmental policy through various relevant directives (2000/60/EC, 2008/105/EC), but concerning the stormwater regulation, only a quantitative approach was adopted (EU Floods Directive 2007/60/EC), while the quality issue is still lacking. Italian normative (LD 152/2006, art. 74, 113) has a general content, which does not effectively deal with stormwater runoff treatment and management, relying mainly on regional "case-by-case" evaluations. Sicily, in particular, is among those regions without a consistent

regulation. Moreover, in semiarid regions, the unconventional water source reuse, would represents a key factor for local-scale integrated water management plans, in the light of the recent EU proposal for water reuse (European Commission, 2018), whose adoption at national levels could eventually conform the strict and uneven normative barriers in Mediterranean area (Aiello et al., 2013; Barbagallo et al., 2012; Salgot et al., 2016; Ventura et al., 2019a). A large number of authors have described the CWs performances for the urban runoff treatment (Adyel et al., 2016) and removal efficiencies of major pollutants (TSS, BOD₅, COD) are generally high (58% – 99% for TSS mainly in the subsurface flow systems, and 31-98% for COD). The removals of TSS and organic matter are strongly correlated because most of the COD and BOD₅ is in particulate form while only a few portion is in soluble form. The average efficiency of nutrients widely varies from 1 to 90% for TN and from 10 to 90% for TP due to the large fluctuations of hydraulic loading rates. In general, when hydraulic rates exceed the system's assimilative capacity, threat of nutrient release may occur. The nutrient's removal rates are not usually influenced by CW configuration and input concentrations, suggesting retention time is a critical factor in nutrient removal efficiencies in any system. Finally, performances on micropollutants, like trace metals, vary according to author and substance. The explanation lies in the different physical forms they can assume. When attached on TSS, trace metals are efficiently eliminated by sedimentation and filtration. When mainly present in dissolved form, most trace metals are not degraded. However, main CWs removal processes for metals are: binding with the granular medium; precipitation as insoluble salts and uptake by plants and bacteria (Garcia et al., 2010).

Even if temperature, conductivity and pH variations promote metal release in water (Walaszek et al., 2017). The knowledge on CWs hydrocarbons removal mechanisms still need to be extended, though the biological and microbiological processes are by now considered the preferential degradation paths. In fact, bacterial consortia plant-bacteria mutual associations and in the plant rhizosphere and endosphere can promote the microbial gene expression of catabolic enzymes linked to hydrocarbons degradation (Hashmat et al., 2018). Also, in formed microbial mats, the characteristic communities apt to oilderivatives metabolization, can vary depending on oil and nutrients levels (e.g. ammonia), as also pH, temperature, dissolved oxygen and sulfate concentration (Abed et al., 2014). Also bioaugmentation strategy have been proposed for oil-contaminated soils treatment (Tao et al., 2019) Overall, physicochemical processes (e.g. photoreactions, chemical precipitation, sedimentation and filtration) alone or combined allow to foster the biological hydrocarbon removal and nowadays, are widely studied as the base of novel technologies, particularly for the degradation of the more persistent PAHs (Li et al., 2019; Wang et al., 2019). The latter can be biologically degraded, but a lack of data makes conclusions on the CWs efficiency not reliable. Besides the general focus, based on the development of effective decentralised wastewater treatment-water recovery system, the specific aim of this study was to evaluate the removal efficiency of an experimental hybrid CW (H-CW) alternatively treating stormwater of a parking area and the effluent of a sequential batch reactor (SBR) installed in a retail store (toilets, kitchens and bar). This would avoid feeding shortage and system block during warmer seasons and provide system robustness in Mediterranean climate

changing conditions (heavy rain episodes and dry weather periods), besides offering social amenity for the local community. The hydraulic behaviour of the subsurface horizontal CW (HF) unit, after less than 2 years of operation, allowed to assess the clogging's effects on the wetland hydraulic properties (i.e. hydraulic conductivity at saturation, K_s). Finally, the possibility of reclaimed water use was evaluated.

4.2 Methods

The pilot-scale H-CW consists of a retention pond, followed by two identical parallel lines, each one including a subsurface HF and a free water surface CW (FWS) unit in series (Fig. 21, 22). It is located within the parking area of the retail store Ikea[®], in the industrial district of Catania, Eastern Sicily (Italy) (37°26'54.2"N 15°02'05.2"E). The system has been operated since the end of 2016 with a nominal hydraulic retention time (HRT) of 96 hours per line. Water quality monitoring was divided into three periods (number of sampling per period, n = 15). I period ranged from May to September 2017, with SBR effluent treatment (feeding frequency of 4 cycles/day), while the II and III periods respectively from November 2017 to February 2018 and from March to May 2018, with for feeding. stormwater runoff During stormwater collection, sampling campaigns were carried out trying to match moments immediately after significant rainfall events (>5mm). Water quality and removal efficiency (R.E., %) were analysed for TSS, COD, BOD₅, TN, TP (Marzo et al., 2018), trace metals (As, Cd, Cr, Fe, Ni, Pb, Cu, Zn) and polycyclic aromatic hydrocarbons (PAHs) as the total sum benzo(b)fluoranthene, benzo(k)fluoranthene, of benzo(ghi)perylene, indeno(1.2.3-cd)pyrene. ICP mass spectrometry (Perkin Elmer NexION 350X) and gas chromatography (GC-Perkin Elmer Clarus 500) were respectively for performed trace metals and PAHs. Monitoring points are (circled numbers) reported in Fig.21 as: 1 (In Pond), 2 (Out Pond), 3 (Out HF) and 4 (Out FWS). Statistical analysis was performed using the Kruskal-Wallis test (R Core Team, 2014) to evaluate the two treatment lines differences. Significance was shaped as p<0.05. On February and June 2018 the first two hydraulic surveys were conducted on HF units (HF1 and HF2) in order to evaluate the hydraulic conductivity in saturated conditions (K_s) of the filtering volcanic gravel beds. The traditional in situ falling head test was therefore applied: sampling points are reported in Fig. 21 as grey crosses (A, B, C) in the HF units.



Figure 21. Pilot-scale hybrid constructed wetland (H-CW) layout and monitoring design (grey circles and crosses, respectively, for water quality and hydraulic surveys). Sequential batch reactor (SBR); retention pond (POND) and free water surface CW (FWS).

The falling head permeameter, based on Lefranc's test, has been largely used to measure K_s of CW (Nivala *et al.*, 2012) applying different schemes and equations suggested in literature (NAVFAC, 1986). Among the different schemes and equations, the standpipe method was chosen (Licciardello *et al.*, 2018). Compared to other methods, a standpipe test does not require a borehole to be drilled as the pipe is merely inserted directly into the sediment, which can save much time and money. K_s was calculated by using the following equation (Eq. 1):

$$K_s = \frac{2\pi R + 11L}{11t} \ln \left(\frac{H_1}{H_2}\right)$$
(1)

where, R is the radius of the tube (m), L is the submerged length of the tube (m), t is time (s) and H_1 and H_2 are the water levels (meters, m) in the permeameter cell corresponding to time t_1 and t_2 (s), respectively. In order to obtain the best fit between modelled H_2 and the measured H_1 , the squared difference between the theoretical curve and that obtained in the field was minimized to estimate the value of K_s by means of an iterative, non linear, procedure that can be done easily using Excel solver (Frontline Systems, Incline Village, NV):

$$\sum_{t=0}^{n} = (H_{obs}(t) - H_{est}(t))^{2}$$
(2)

where H_{obs} is the height of the water table level measured inside the tube at time t during the test (m); H_{est} is the corresponding modelled data, by Eq. (2).

In order to fill the permeameter with water in a pulse mode, as required, a permeameter unit was assembled. In particular, an impervious steel tube (internal diameter of 0.10 m and length of 1.5 m) for the insertion into the medium was connected to a plastic water reservoir (6.6 L

volume) by means of a bulb valve. The permeameter was inserted by using a mallet, into the medium to a depth of about 0.32 m, after the creation of a small hole in the granular medium to reach the water table. The decrease of water height inside the tubes was monitored until the water level reached the water table by using a pressure probe (STS - Sensor Technik Sirnach, AG), connected to a laptop by means of a CR200-R (Campbell Scientific) data logger, positioned inside the steel tube. Pressure data were then converted into water heights. For each test, the experimental monitoring time was fixed in 60 seconds and the pressure probe was configured to collect 4 water height data per second.



Figure 22. View of the pilot-scale hybrid constructed wetland (H-CW): single stages (FWS and HF) and overall view (with a detail of the system inlet retention tank, named as "Pond").

4.3 Results and Discussion

Effective data monitoring and collection were the most challenging issues faced when stormwater fed the experimental system. In fact, the Sicilian region was clearly affected by heavy rainfall deficit (>30-40%) in 2017, with respect to the period 2003-2016 (SIAS), and very few significant events were recorded (Fig. 23).



Figure 23. Rainfall deficit recorded in Sicily in 2017, in comparison to the period 2013-2016 (data SIAS) and in situ pilot H-CW meteorological data: total rain (mm/day) mean air temperature (°C) (February 2017 – March 2018).

Mean values of main physical parameters for each period are reported in Table 8. Identical treatment units were further averaged (HF1-HF2: monitoring point, 3; FWS1-FWS2: monitoring point 4), since non-parametric data analysis did not show differences (Kruskal-Wallis test pvalues>0.05). P-values are respectively embedded in Table 8 (see caption ^a) and Table 9 for physical and chemical parameters. EC values varied between I and II-III periods, and DO along the treatment units. The latter has been described as a key indicator of wetland metabolism, and its variability can describe the fluctuating balance between autotrophic and heterotrophic processes. All these features, influenced by seasonal changes (climate and plant phenology), together with multi-compartment CW design and diversified flow conditions (as also different HRT along the treatment units) become very challenging when managing these type of systems (Adyel et al. 2016). The alternation of lentic and lotic phases strongly characterizes many wetlands in Mediterranean environments, clearly impacting on DO dynamics. In this case, the only H-CW unit exhibiting higher hydrodynamic regime was the retention pond placed at the inlet system, probably because of the incoming influent flushing, while the remaining stages mainly presented lentic characteristics. A proof of that could be linked to higher levels of DO and pH recorded in the retention pond (Tab. 8, point 2), while temperature seemed less influent. Also, during the warmer periods, when the exposure to solar radiation was high, the accumulation of thick algal mats as floating mass in the FWS units (Tab. 8, point 4) could have limited both photosynthetic activity and atmospheric DO exchange. Only during the II period, ranging from November to February, with lower

temperature and solar radiation, but higher wind velocity (data not shown), OD in FWS units increased.

Table 8. Average values and standard deviation (SD, \pm) of physical parameters recorded at each sampling point during the three monitoring periods; dissolved oxygen (DO), electrical conductivity (EC, at 20°C), pH and T (°C). (a) Kruskal-Wallis test p-values calculated between identical treatment units (HF1-HF2: monitoring point 3; FWS1-FWS2: monitoring point 4).

	I period				II period				III period			
Point	1	2	3	4	1	2	3	4	1	2	3	4
DO			4.0	5.6			1.6	8.4			2.5	3.3
(mg L ⁻	6.7	8.6	(2.1)	(2.5)	4.9	9.8	(0.6)	(2.3)	3.1	8.9	(0.1)	(0.1)
1)	(1.8)	(0.9)	^a 0.63	^a 1	(1.5)	(1.8)	^a 0.91	^a 0.08	(0.7)	(0.9)	^a 0.6	^a 0.59
EC			2130	2851			319	471			211	218
(µS	1910	2129	(224)	(1443)	114	161	(161)	(291)	160	174	(32)	(30)
cm ⁻¹)	(388)	(236)	^a 0.26	^a 0.87	(14)	(55)	^a 0.25	^a 0.35	(73)	(68)	^a 0.35	^a 0.07
			7.8	7.2			7.1	7.5			7.1	7.4
	7.5	8.4	(0.5)	(0.2)	6.7	7.9	(0.8)	(0.8)	6.7	8.6	(0.01)	(0.01)
pН	(0.3)	(0.3)	^a 0.69	^a 0.57	(0.9)	(1)	^a 1	^a 0.92	(0.7)	(0.4)	^a 0.67	^a 0.91
			24.7	26.5			14.8	15.21			20.4	21.7
Т	28.9	26	(2.2)	(3)	15.2	15.1	(1.3)	(1.4)	23.9	22.7	(0.1)	(0.2)
(°C)	(3.1)	(2.1)	^a 0.57	^a 0.87	(1.9)	(2.1)	^a 0.35	^a 0.75	(6.1)	(6)	^a 0.92	^a 0.75

Table 9. Kruskal-Wallis test p-values calculated for chemical parameters between identical treatment units (HF1-HF2: monitoring point 3; FWS1-FWS2: monitoring point 4).

Period	Point	TSS	COD	BOD ₅	TN	TP	Cr tot	Fe tot	Ni tot	Pb tot	Cu tot	Zn tot
	3	0.26	0.33	0.87	0.87	0.87	-	-	-	-	-	-
Ι	4	0.52	1	0.87	0.23	0.63	-	-	-	-	-	-
	3	0.06	0.88	0.25	0.07	0.39	1	0.6	0.83	1	0.46	0.37
II	4	0.25	1	0.15	0.66	0.56	0.7	0.12	0.6	0.75	0.83	0.5
	3	0.88	0.77	0.66	0.05	0.77	0.41	0.75	0.4	0.91	0.17	0.4
III	4	0.75	0.15	0.08	0.44	0.09	0.88	0.17	0.6	0.74	0.14	0.92

Figure 24 reports concentrations and removal efficiency (R.E., %) of the main chemical parameters during I monitoring period. Higher R.E. took place at HF outlet for TSS and organic matter, while the opposite trend was observed for nutrients. The H-CW presented undeveloped Typha latifolia root systems, that were introduced into the FWS on floating support, to act as further filter for retention. The algal blooms occurred in FWS tanks and determined by increased temperature, caused water quality worsening at the H-CW outlet (TSS, COD and TN values were higher than national standard limits for reuse, Table 10) suggesting the system was still at initial and unstable conditions. On the other hand, as already described by others (Vymazal, 2013; Àvila et al., 2017) FWS confirmed to play a positive role in nutrients removal. Nevertheless, as reported by Avila et al. (2017) the effluent recirculation could be an effective solution for TN removal in hybrid CW systems, when unfavourable conditions occur. Values reported in Fig.24 were generally not complying with the Italian standard limits for reuse (tab. 10), except for COD and TP, but confirmed the self-defeating strictness of this legislation. In fact, according to the table 2 of the European proposal for water reuse (EU, 2018) on reclaimed water quality criteria for agricultural irrigation in case of non-food crops (quality classes B and C), BOD₅ and TSS limits refer to those reported in the EU Directive on urban wastewater treatment (91/271/EEC): respectively 25 mg L⁻¹ and 35 (<10.000 PE) or 70 mg L⁻¹ (2.000-10.000 PE), while COD and nutrients limits are not reported.



Figure 24. I monitoring period (SBR effluent treatment). Maximum-minimum and mean concentrations of conventional water quality parameters: TSS, COD, BOD, TN, TP at the system inlet (In Pond), the horizontal unit outlet (HF, monitoring point 3) and system outlet (free-water unit, FWS, monitoring point: 4). Removal efficiencies (R.E., %) calculated between the inlet of the system and the intermediate treatment stage (Pond + HF unit) and the overall (Pond + HF unit + FWS unit).

In Figure 25, trace metals detected during both runoff treatment periods (II and III) are reported. Overall trace metals removal efficiencies (%), for Cr, Fe, Ni, Pb, Cu, Zn, were respectively 12 (± 9), 9 (± 1), 6 (± 5), 53 (± 30), 30 (± 28) , 61 (± 5) in the II period and 63 (± 40) , 33 (± 32) , 39 (± 27) , 90 (± 12) , 74 (± 9) , 91 (± 7) in the III. Removal efficiencies mostly enhanced during the III period, possibly more appreciable because of higher concentrations in the case of Cr, Ni and Pb, while Cu and Zn, showed very high removal efficiencies even present at lower concentrations. According to Schmitt et al. (2015), metals in urban runoffs are mainly particle-bound but Cu and Zn are also present in the dissolved fraction (retained both, in the pond and in the filter; the latter in fact constitutes further retention for dissolved fraction), but the biological uptake of plants and bacteria should be assessed as well. However, the overall concentrations were always below the Italian standard limits for discharge in water bodies (Table 3).



Figure 25. Log-scale average concentrations (μ g L⁻¹) and SD for trace metals at the hybrid CW inlet (In Pond) and outlet (Out FWS), during stormwater runoff treatment (II-III monitoring periods).

Italian standard	TSS	BOD ₅	COD	TN	TP	Tot Cr	Fe	Ni	Pb	Cu	Zn	E. coli
limits												21
WW Lingh and a	80	40	160	≈35°	10	2	2	2	0.2	0.1	0.5	5.0×10^{3d}
aischarge"												
WW	10	20	100	15	2	0.1	2	0.2	0.1	1	0.5	100 ^e
<i>reuse^b</i>												

Table 10. Physical-chemical (mgL^{-1}) , trace metals (mgL^{-1}) , and bacteriological (CFU 100 mL⁻¹) Italian standard limits for wastewater discharge and reuse.

^a Legislative Decree 152 (2006), ^b Ministerial Decree 185 (2003), ^c Limit for discharge into surface water bodies (as the rough sum of NH₄, N-NO₂ and N-NO₃), ^d Recommended value for P.E. > 2000, ^e Maximum value in 80% of samples.

The indicator microorganisms (Tab. 11) analysed, among those recommended in the last European proposal for water reuse (European Commission, 2018) were *E. coli* and *Clostridium perfringens*, respectively the most appropriate for pathogenic bacteria and protozoa assessment. Both indicators were efficiently removed during the treatment process. Microbial inlet concentrations were below Italian standard limits (Table 10) but a peculiar path was observed in the case of *Clostridum perfringens*, whom removal (2 u-log) was always reached at the HF outlet. Further investigation might be useful to clarify the possible influence of DO and suitable substrates presence for anaerobic respiration along the length of the H-CW.

Table 11. Average concentrations (CFU 100 ml-1) and removal efficiencies (R.E., u-log) of indicator microorganisms in hybrid CW inlet (In Pond) and outlet (Out FWS) during II monitoring period. Standard deviation (\pm SD) are in brackets. (a) Kruskal-Wallis test p-values calculated between identical treatment units (FWS1-FWS2: monitoring point 4).

II monitoring period	In Pond (CFU 100 ml ⁻¹)	Out FWS (CFU 100 ml ⁻¹)	R.E. (<i>u log</i>)
Escherichia coli	106 (±83)	3 (±1) ^a 0.12	1.5(±0.4)
Clostridium perf.	464 (±623)	6(±3) ^a 0.05	2 (±0.4)

TSS and organic matter (BOD₅, COD) average concentrations (mg L⁻¹) at the hybrid CW inlet and outlet recorded when the system was fed with stormwater were respectively 7 (±0.05), 12 (±5), 21 (±7) and 11 (±0.6), 9 (±2), 26 (±9). Values were below the Italian standard limits (Table 10). Similarly, TN and TP average concentrations (mg L⁻¹) at the hybrid CW inlet and outlet were below the

national standard limits: inlet and outlet TN and TP mean values between II and III monitoring periods were, respectively: 3 (±1.3), 2.6 (0.3) and 0.1 (±0.01) and 3.5 (± 2.4) . A slight TP increase at the outlet was observed, probably influenced by the II period, as also showed by reference data (mean HF and FWS outlet concentrations were respectively (mg L⁻¹) 5.5 (\pm 2.7) and 5 (\pm 3.6)). In Mediterranean climate, the ranging period from November to February corresponds to macrophyte seasonal senescence, which in this case could have reasonably contributed to internal nutrient release along the system (Adyel et al., 2016). PAHs monitored during II and III periods were in very low concentrations (0.006 $\mu g L^{-1} \pm 0.003$) with a reduction of 40% (\pm 23) already at the pond outlet. As previously described, the interaction between plants and microorganisms is a preferential path in hydrocarbons degradation. Despite the absence of macrophytes in the stabilization pond, the slight removal trend observed at PAHs low concentrations suggest that microbial activities and physicochemical processes could have already occurred. On that regard, Schmitt et al. (2015) described sedimentation as a relevant mechanism of PAHs retention in CWs but the present study did not investigate specific removal paths.



Figure 26. K_s values obtained during hydraulic monitoring campaigns of February and June 2018, through Falling head test method on both subsurface horizontal CW: HF1 and HF2.

Figure 26 reports K_s mean values (number of sampling per point n=3) measured in both pilot beds at different distances from the inlet in February and June 2018 (after 1-1,5 operations-years). K_s values measured in June were very similar to those measured in February for all measuring points in both beds. Just in the middle point of HF1 there was a significant reduction between K_s values measured in February and in June equal to 2243 m/d. Ks values measured during the operation period were also compared with K_s value measured for the clean gravel, equal to 12135 m/d (SD=1591). K_s values after the operation periods decreased just in the area close to the inlet in both beds and in the HF2 bed also in the area close to the outlet of about 17%. These reductions were higher than the maximum SD of measurement repetitions in the same place and time and equal to about 1500 m/d. Mean K_s values measured in the middle areas and close to the outlet, were sometimes even higher than the values observed for the clean gravel (i.e. point HF1-A and HF2-C when IMP permeameter was used and HF1-C when P permeameter was used). This behaviour could be due to the high variability of K_s values, being those differences in the range of the measured variability of the parameter.

4.4 Conclusions

Preliminary experimental results on a pilot-scale H-CW are promising and suggest good reliability in the perspective to combine this technology in decentralised water treatment facilities (DWTFs) for enhancing the water recovery and reuse (e.g. green area irrigation and toilette flushing), in Mediterranean climate conditions. Particularly in Sicily, DWTFs by natural system can play a strategic role for promoting the wastewater reuse, as suggested by several authors (Barbagallo *et al.*, 2013; Ventura *et al.*, 2019a). However, previsions on main critical issue became heavier than expected when considering the effect of such a strong rainfall scarcity. The feeding modalities of H-CW, particularly in a climate change scenario, have highlighted the following main critical points:

- the need to properly assess the interaction among nutrientrich or poor effluent/feeding frequency/plant phenology. In fact, as reported above, the feeding switching from nutrientrich SBR effluent to poor runoff at the end of the I period corresponded with the macrophyte senescence starting phase, presumibly causing the TP increase from the inlet to the outlet of the system. Consequently, different feeding wastewater affects the H-CW treatment efficiency;

- the need for recirculation phase could be required in order to improve a multi-compartment CW treatment efficiency and cope with wastewater variability, addressing the most feasible option between reuse or discharge.

- the difficulty to conduct a robust monitoring activity and data collection.

However, general conclusions pointed out that:

- the overall trace metals and PAHs removal efficiencies were good in spite of the low inlet concentrations detected, which were actually much lower than expected; hydrocarbons removal paths and processes occurring in hybrid CWs would be further investigated.

- the hydraulic monitoring of a novel H-CW from earlier operation phases, allows to provide a long-term complete screening of the hydraulic conductivity at saturation, starting from a zero-point survey.

Finally, further research is required to increase knowledge on hybrid CWs "metabolic pathways" in highly variable climate regions.

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5 Assessment of clogging in constructed wetlands by saturated hydraulic conductivity measurements⁵

Abstract

This study aims at defining a methodology to evaluate K_s reductions of gravel material constituting CW bed matrices. Several schemes and equations for the Lefranc's test were compared by using different gravel sizes and at multiple falling-head test spatial scales. The method was steel permeameters implemented bv using two one impervious (IMP) and one pervious (P) on one side. At laboratory scale, mean K_s values for a small size gravel (8- 15×10^{-2} m) measured by the IMP and the P permeameters were equal to 19466 m/d and 30662 m/d, respectively. Mean K_s values for a big size gravel (10-25 x 10⁻² m) measured by the IMP and the P permeameters were equal to 12135 m/d and 20866 m/d, respectively. Comparison of K_s values obtained by the two permeameters at lab scale as well as a and analysis sensitivity а calibration, lead to the modification of the standpipe equation, to evaluate also the temporal variation of the horizontal K_s . In particular, both permeameters allow the evaluation of the K_s decreasing after 4 years-operation and 1-1.5 years-operation of the plants at

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full scale (filled with the small size gravel) and at pilot scale (filled with the big size gravel) respectively.

Keywords: Horizontal flow; K_s measurements; permeameter cell

5.1 Introduction

The process of clogging is inevitable in horizontal subsurface flow (HSSF) constructed wetlands (CWs) due to the slow flowing of treated wastewater through the porous medium. The development of clogging can be detected by the appearance of water on the surface of the granular medium. The phenomenon is very variable in space, being generally most severe within the first few meters (Vymazal 2018) of the CW but sometimes, also occurring close to the outlet area. Due to its complexity, the development and impact of clogging on system design and operation must be taken into account (Knowles et al., 2011). Main causes of clogging can be organized in three categories. In particular, some factors are related to influent characteristics (pretreatment, BOD₅ and TSS concentrations and flow); others are related to the system design (shape, size, operation mode, distribution and collection systems, pipes, media, plants); others are related with bed activities (rhizosphere, biofilm. chemical processes, accumulated matter composition). The factors related to the first two categories can influence the third making the phenomena not currently known in detail (Correia, 2016). In some cases, clogged HSSF-CWs can operate with overland flow and still meet

treatment objectives (Griffin et al., 2008). If the surface loading rates are characterized by BOD₅ <10 g m⁻² d⁻¹, COD <20 g m⁻² d⁻¹, <10 g TSS m⁻² d⁻¹, partial clogging may occur only after about 15 years of operation (Vymazal, 2019). Partial ponding has no significant effect on the quality of discharged water, as well as the replacement of partially clogged inflow zone filtration material does not give any significant improvement (Vymazal, 2018). However, reduced treatment performance or the risk of human contact with untreated or partially treated sewage sometimes attracts the attention of regulatory agencies and Preventative strategies, such stakeholders. as best management practices, inlet and loading adjustments, and changes to hydraulic operating conditions (including intermittent operation, backwashing and/or reversing the direction of flow) can be carried out to delay or minimize the negative effects associated with clogging. Advanced clogging may eventually require restorative strategies such as: excavation of dirty gravel and its replacement with new or washed one, and use of gravel; direct application of chemicals to the gravel bed; and most recently, the application of earthworms to the system (Nivala et al., 2012).

Due to the fact that clogging is a widespread operational problem in HSSF-CWs, as demonstrated by frequent reporting over the past two decades, it has become increasingly important to identify practical methods for its measurement.

Clogging assessment techniques include hydrodynamic visualisations by means of tracer tests (Bowmer 1987; Knowles *et al.*, 2010), the analysis of accumulated solids in filter media (Caselles-Osorio *et al.*, 2007; Pedescoll *et al.*, 2009), and determination of the hydraulic gradients between

points in the filter media (from which the mean hydraulic conductivity can be estimated) (Sandford *et al.*, 1995; Rodgers and Mulqueen 2006; Suliman *et al.*, 2006). The clogging process is very much affected by the size of filtration material and consequently by its saturated hydraulic conductivity (K_s). Common laboratory methods in CW is not advisable, to measure K_s in CW due to the difficulties to take undisturbed media samples. For this reason the measurement of K_s in CWs have been mostly carried out by means of *in situ* methods (Knowles *et al.*, 2011).

 K_s measurements can be conducted by falling and constant head tests specifically developed to detect the potentially high hydraulic conductivity of wetland gravels. Ideally, these methods are able to measure a wide range of hydraulic conductivities of porous media, as determined by media properties and the stage of clogging (i.e., K_s values lower than 500 m/day that might be typical of the transition and completely clogged media). The between clean repeatability and accuracy of the methods used have been previously described in Knowles and Davies (2009) and Pedescoll et al. (2011). However, these methods need further validation tests, as development and application within this scope is relatively recent (Knowles et al., 2011; Aiello et al., 2016).

In particular, the falling head permeameter, based on Lefranc's test, has been used to measure K_s of CWs (Nivala *et al.*, 2012) applying different schemes and equations suggested in literature (NAVFAC 1986).

This study aims at defining a methodology sufficiently accurate and relatively easy to implement, to estimate K_s reductions of gravel material constituting CW bed matrices due to clogging phenomena. In order to meet this aim,

different schemes and equations for the Lefranc's test were compared by using different gravel sizes and at multiple spatial scales (lab, pilot and full plant) employing also a new type of *in situ* permeameter cell specifically developed for falling head tests.

5.2Material and Methods

5.2.1Experimental schemes implemented at lab scale

Different schemes can be applied to determine saturated hydraulic conductivity from falling-head tests conducted in a CW by *in situ* permeameter cell condisting of an open ended tube that may eventually encase the sample to be tested (Morris and Knowles 2011).

Depending on the length of the cavity at the bottom of the piezometric tube, the measured K_s can represent the horizontal, three-dimensional or vertical saturated hydraulic conductivity within the measurement zone (Amoozegar 2002). If the cavity cannot be maintained during measurements due to soil collapse, a well screen can be attached at the bottom of the piezometric tube. According to the geometry of the cavity (that is the ratio between radius, R, and length, L) different schemes can be considered (NAVFAC 1986). Three of them were tested in the present paper and were reported in Figure 27 (schemes 1 to 3), together with the corresponding equations to calculate K_s and their applicability conditions. In particular, scheme 1 and scheme 3 implied the use of an impervious permeameter cell (IMP) with and without gravel inside, respectively; scheme 2 implied the use of a pervious permeameter cell (P) without gravel inside. The scheme 1, also called standpipe test, did not require a borehole to be drilled as the pipe was merely inserted directly into the substrate, which can save much time compared with schemes 2 and 3. Also, excavation of a borehole could be a disturbance to porous medium. As a disadvantage, this method did not allow to evaluate horizontal K_s . This could be not a problem for isotropic gravel (clean or not clogged gravels) but, given that the clogging process is likely not to be isotropic, vertical K_s values obtained by this method could be different from horizontal ones. On the other hand, to our knowledge a scheme for predicting also horizontal K_s using a P permeameter and that does not require a borehole to be drilled was not proposed. Thus, scheme 4, which implies the use of P permeameter with gravel inside was also implemented. An arrangement similar to scheme 4 was already used in literature by Pedescoll et al. (2009), but these authors calculated K_s by equation 2 that properly corresponds to scheme 2 (Figure 27). In the present paper, besides checking this combination (scheme 4-equation 2), scheme 4 was applied using, as a first stage, original equation 1 and then a calibrated version of equation 1, as described below.

In order to test the four proposed schemes, two different steel tubes were used (internal diameter 0.10 m and length 1.5 m), being one impervious (IMP permeameter) and one pervious on one side (P permeameter) for a total area of 3.11 x 10^{-4} m², distributed along 0.25 m from the bottom of the permeameter. The P permeameter was implemented in order to evaluate also the temporal variation of horizontal K_s by repeated measurements in the time. The permeameters were inserted, using a mallet, into the medium to a depth of about 0.32 m, after the creation of a small hole in the granular medium to reach the water table. To instantaneously add

water to the permeameter (in a single-pulse mode) a plastic water reservoir (6.6 L volume) was assembled to the measurement units by means a bulb valve (Figure 28).

Variation of the water level, H, within the permeameter was measured by a pressure probe (STS - Sensor Technik Sirnach, AG), connected to a laptop by means of a CR200-R (Campbell Scientific) data logger. The pressure probe was positioned inside the steel permeameter through a driver added to the device. The pressure value at atmospheric pressure was checked before each measurement started. Four water level data per second were recorded for a duration of 30 seconds. The decrease of water height inside the permeameter was monitored until the water reached the water table.

For each scheme – equation combination tested in the paper, in order to obtain the best fit between simulated and measured water levels, the sum of squared differences between the theoretical curve and that obtained in the field was minimized, to estimate the value of K_s by means of an iterative, non-linear, procedure that make use of Excel solver (Frontline Systems, Incline Village, NV):

$$\sum_{t=0}^{n} = [H_{obs}(t) - H_{sim}(t)]^2$$
(4)

where H_{obs} is the height of the water table level measured inside the permeameter at time t during the test (m); H_{sim} is the corresponding modelled data calculated by the different equations 1,2,3 and 4 depending on the used scheme.

Scheme 1	Scheme 2	Scheme 3	Scheme 4	

Impervious permeameter Pervious permeameter cell Impervious permeameter Pervious permeameter cell cell with material inside without material inside (Cased cell without materials with material inside (Standpipe) hole with uncased extension) inside (Cased hole)



Figure 27. Schemes and equations tested for K_s measurements by falling-head method adapted from NAVFAC 1986). R is the radius of the tube (m), L is the submerged length of the tube (m), t is time (s) and H1 and H2 are the water levels (meters, m) in the permeameter cell corresponding to time t1 and t2 (s).
Ten falling head infiltration tests were performed at lab scale in a central point of a tank of $1m \ge 1 m$ (Figure 29). Two different sizes of clean gravel medium, 8-15 $\ge 10^{-2} m$ ('small') and 10-25 $\ge 10^{-2} m$ ('large') were tested with the same procedure. The effective porosity, n,of both media was 0.4.



Figure 28. Permeameter unit for falling head tests.

5.2.2 Standpipe scheme calibration at lab scale by using both IMP and P permeameters

To make the original equation 1 suitable for scheme 4, a calibration procedure was carried out. The values of K_s obtained by the combination of scheme 4 – original equation 1 (P- K_s) and those obtained by the combination of scheme 1 – original equation 1 (IMP- K_s) were used to modify the original equation 1. In particular, for the P- K_s values the R and L parameters of equation 1 were reduced in order to obtain results closer to IMP- K_s values. To be more specific, R (equal to 5.0 cm) was fixed to 4.9 cm to account for a reduction of the cross permeameter area corresponding to the total area of the small lateral holes (3.11 x 10⁻⁴ m²). Then, it was assumed that P permeameter allowed water to flow out of the P permeameter at a smaller depth (from the level of the ground) than the IMP permeameter. Thus L was

reduced to give P- K_s values closer to those measured with the IMP permeameter. The calibration procedure of the original equation 1 was applied to 'small' and 'large' size of clean gravel medium.

5.2.3 Stand pipe scheme application at pilot and full scale by using both IMP and P permeameters

The combinations mentioned above (schemes 1 and 4 with original equation 1) were also tested at full and pilot scale in two experimental plants. The hybrid CW system working as secondary wastewater treatment system of the Ikea[®], located in the industrial district of Catania, Eastern Sicily, Italy (37°26'54.2"N 15°02'05.2"E, 11 m a.s.l.) was considered as plant test at full scale. The plant is located in a semi-arid area, characterized by climate an average annual precipitation of about 500 mm, and values of air temperature in summer reaching 40 °C. The CW system was added to the pre-existing treatment system (including a sequential batch reactor system and a screening unit) in 2014, in order to respond to the high hydraulic and nitrogen load variability during the year. The sequential batch reactor system (SBR) was designed to treat a maximum flow rate of 30 m³ day⁻¹ (2 batch phases every 12 hours) and Total Nitrogen (TN) concentration of 135 mg L⁻¹. Anyway, during holidays or pre-holidays, wastewater flow rate could be 2-4 times greater than that during a normal working day, and NH_4 can be higher than 200 mg L⁻¹. The hybrid CW system includes three in series beds: a horizontal subsurface treatment wetland allowing a reduction of organic matter and suspended solids concentrations followed by two vertical subsurface flow treatment wetlands. The $K_{\rm s}$ measurements were carried out only in the horizontal subsurface treatment wetland filled with "small size" gravel. This bed covered a surface area of about 400 m² (12 × 34 m) with a depth of 0.60 m, and it was planted (with a density of 3 plants m⁻²) in July 2014, with two different aquatic species *Phragmites australis* (for $\approx 80\%$ of the HF surface area), and *Iris pseudacorus* close to the HF outlet. The bed was designed to be fed discontinuously, receiving 30 m³ daily effluent from SBR (two discharges of 15 m³ each one) and 20 m³ effluent from the screening unit, that by-pass the SBR when the wastewater production exceeds the designed flow rate. The hydraulic loading rate of the bed varied between 75 and 125 L m⁻² d⁻¹. For more details about the full scale hybrid CW system, see Marzo *et al.* (2018).

The hybrid pilot scale CW, also located within the parking area of the retail store Ikea[®], consists of a fiberglass retention pond (flow rate of 2 m^3/d), followed by two identical parallel lines (flow rate $1 \text{ m}^3/\text{d}$), each one including in series a HSSF bed and a free water surface (FWS) unit. The system, operating since the end of 2016 with a hydraulic retention time of 96 hours per line, alternatively treated storm water of a parking area and the sequential batch reactor WW produced from the retail store (toilets, kitchens and bar). The K_s measurements were carried out in both HSSF beds (H1 and H2) filled with the "large size" gravel. Both HSSF beds, characterized by a width of 1.5 m, a length of 4.5 m and having a depth of 1.1 m, were planted with Canna indica. The falling head tests were carried out at a distance of 1 m (A), 2 m (B) and 3 m (C) from the inlet (Figure 29). For more details about the pilot scale hybrid CW system see Ventura et al. (2019b).

The K_s measuring points at lab, pilot and full scale plants are represented in Figure 29. Four falling head infiltration

tests were performed for each of the 9 measuring points located in the full scale plant. Mean K_s were then calculated by averaging 4 x 3 K_s data obtained at each transect at the same distance from the inlet (i.e. 1-2-3, 4-5-6, 7-8-9) (Figure 29). Three falling head infiltration tests were performed for each of the 3 measuring points located in the pilot scale plant; in this case, given that the two beds work in parallel, the tests were repeated in both beds. The falling head tests at full scale were carried out in April 2018 whereas at pilot scale in February and June 2018.



Figure 29. Measuring points inside the full scale, the pilot and the lab tank used to measure K_s . A, B, C and D inside the full scale plant indicate repeated infiltration tests conducted around each fix installed piezometer.

5.3 Results

5.3.1 K_s values obtained at lab, pilot and full scale by using NAVFAC (1986) schemes and equations

Table 12 reports mean K_s values (N = 10) for both sizes of clean gravel obtained by using P and IMP permeameters at lab scale, as well as, the ratio between P- K_s and IMP- K_s s mean values. Unexpectedly, the K_s was lower for the 'big' gravel than the 'small 'one, probably due to a lower uniformity and to a prevalent angular shape of bed matrix. As expected, P permeameter gave higher values of K_s than IMP permeameter for both size of gravels; the ratio between P- K_s and IMP- K_s values was 1.6 and 1.7 respectively for 'small' and 'big' size gravels. The highest standard deviation (3903 m/d) was observed in correspondence to the highest mean K_s values (P permeameter filled with 'small' size material).

Table 13 reports mean K_s values (N= 4 repetitions x 9 piezometers = 36) measured around the 9 measuring points at the full scale plant by using P and IMP permeameters and the corresponding standard deviations. All K_s values obtained with the IMP permeameter were lower than the values observed for the clean gravel at lab scale (table 12), with reductions up to about 90% in the area close to the inlet and up to about 55-65% in the middle and in the outlet areas. The behaviour was the same when the P permeameter was used. The reductions were higher than the variability of measurements, being maximum standard deviation value equal to 2022 m/d for the IMP permeameter and 2937 m/d for the P permeameter. Mean K_s values (N= 4 repetitions x 3 piezometers = 12) obtained by averaging the measurements at each transect from the inlet are reported in Figure 30.

Mean K_s values close to the inlet were 5.5 and 4.1 times lower than those measured for the clean gravel by IMP and P permeameters, respectively. In the middle and in the outlet areas of the full scale CW both permeameters gave mean K_s values 2.2 times lower than those measured for the clean gravel.

Table 14 reports K_s mean values (N8 = 3 repetitions x 3) piezometers x 2 beds =18) measured in both pilot beds (H1 and H2), filled with "big size" material, at different distance from the inlet in February and June 2018 by using IMP and P permeameters. At both sampling dates, K_s values were lower than the value measured for the clean gravel (table 12) just in the area close to the inlet in both beds and in the H2 bed also in the area close to the outlet. Anyway, K_s values measured in February and in June were very similar for all sampling points and for both beds, therefore only the K_s reductions with respect to clean gravel will be discussed. The K_s reductions measured with the IMP permeameter were higher than the maximum standard deviation of the repetitions in the same place and time equal to about 1500 m/d. Mean K_s values measured in the middle areas and close to the outlet, were sometimes even higher than the values observed for the clean gravel (i.e. point H1-A and H2-C when IMP permeameter was used and H1-C for P permeameter). This behaviour could be due to the high variability of K_s values, being those values in the range of the measured variability of the parameter. The behaviour of K_s values observed with the P permeameter was similar to that obtained for IMP permeameter. Anyway, K_s reductions after the operation period measured by P permeameter were lower than those observed by using the IMP permeameter and standard deviation of the repetitions in the same place and time reached about 2646 m/d. Again, the highest variation of the measurements was associated to a very high mean K_s value (Table 14). Figure 31 reports the behaviour of K_s values averaged between both pilot beds (N= 3 repetitions x 1 piezometer x 2 beds = 6) after 1 and 1,5 years-operation. Again, it was clear that there was not a significant K_s reduction from February to June 2018. Mean K_s values close to the inlet were 83% and 97% of the K_s values measured for the clean gravel by IMP and P permeameters, respectively.

The saturated hydraulic conductivity values obtained by using the scheme 2 and the respective equation were lower than those obtained by using scheme 1 and equation 1 of about one order-of-magnitude. However, the K_s values obtained by using the scheme 3 and the respective equation were not reliable due to the fast lowering of the water level inside the permeameter. Anyway, schemes 2 and 3 were too time consuming and difficult to apply especially at real scale plants.

5.3.2 K_s values obtained by scheme 4 and calibrated equation 1

As expected, K_s values obtained at each spatial scale, by uncalibrated equation 1 and scheme 4 were overestimated. To assess the degree of overestimation, the ratio between K_s values obtained for clean gravel and after the operation in both full (N=12) and pilot scale (N=6) plants was calculated for P and IMP permeameters.

The ratio for clean gravels was very similar for 'small' and 'big' size materials being 1.6 and 1.7, respectively. After the operation period, the ratio P/IMP of the mean K_s values was different along both plants (considering inlet, middle and outlet areas). In particular, the ratio varied in the range 1.3-1.5 for the 'small' size material and 1.7-2.0 for the 'big' size

material (Figures 30 and 31). The variation could be due to a non-uniform spatial and temporal evolution of the clogging process. In particular, in the middle area of the pilot plant, where there was almost no clogging, and K_s values measured after the 1.5 years-operation period were the same of those obtained for the clean gravel, the ratio value of 1.7 was confirmed. This result can help in the reliability of the proposed methodology.

Results of the sensitivity analysis carried out by using clean gravels at lab scale, showed that K_s values were not very sensitive to the reduction of R up to 10%. On the other hand, the K_s values were very sensible to the variation of L that was changed from 10 to 50% from the initial value of 32 cm. The values that allowed us to obtain P- K_s values closest to IMP- K_s values, and so to calibrated equation 1, were L = 18 cm for the 'large' size material and L = 20 cm for the 'small' size material.

Calibrated equation 1 was applied to K_s values measured at full (Table 13) and pilot (Table 14) scale CWs. The standard deviations among repetitions in the same place or close to each other were of the same order-of-magnitude of those obtained for P permeameter. The variation of K_s values averaged among those obtained at the same distance from the inlet after the operation period of beds and that take into account also the accumulation of clogging in the horizontal direction is also showed in Figures 30 and 31 for full and pilot scale CW beds.

5.4 Discussion

The decrease of K_s after the CW operation period was higher in the full scale plant than in the pilot scale plant, probably due to the longer operation life and a higher

organic load (data not shown). As widely reported in literature (Vymazal 2018; Pedescoll et al., 2012; Knowles et al., 2009), the clogging phenomena were more severe in the area close to the inlet of both plants. However, the observed $K_{\rm s}$ reduction in the area close to the outlet of the H2 were also documented in literature (Correia 2016). The saturated hydraulic conductivity determined with the implemented IMP permeameter and modelled with equation 1 gave values comparable to those reported in literature for clean gravels with similar sizes and porosity. In particular, Reed et al. (1995) for media with 8<D₁₀<16 mm and 30<n<38% reported K_s values ranging from 500 to 10,000 m/d, whereas for media with $16 \le D_{10} \le 32$ mm and $36 \le n \le 40\%$ the K_s values ranging from 10,000 to 50,000 m/d. In other studies, K_s observed with different method (i.e. surveys) are of the same-order-of-magnitude in the area close to the outlet (Knowles et al., 2009; Nivala et al., 2012). Contrarily, in some studies (Caselles-Osorio et al., 2007; Pedescoll et al., 2009) lower K_s values are reported (maximum values at the outlet area 810 m/d). However, these studies applied the scheme 4 with equation 2, thus this combination was also verified in the present paper. The results showed that in this case mean K_s values were lower than those obtained with other schemes up to a factor of 25, however they were of the same order-of-magnitude compared to those of the cited studies. Therefore, results showed that scheme 1 used with equation 1 was the most suitable to measure K_s in clean gravels or unclogged media where the isotropic conditions are still valid and vertical and horizontal K_s are the same. During the operation period of CWs at both pilot and full scales, the clogging phenomenon made the medium nonisotropic, so, vertical K_s alone was not representative anymore. Calibrated equation 1 (by using L and R

optimized values) allowed us to use P permeameter to evaluate K_s variation during the operation period of CWs taking into account also the influence of clogging in the horizontal direction and also saving time. High standard deviations among repetitions carried out in the same place (for pilot scale plant) or close to each other (for full scale plant) and at the same sampling data were often associated at higher K_s values, generally, they were higher for P- K_s values. This phenomenon should be reduced during CW operation period, namely when the use of P permeameter will become more useful, due to the increasing of clogging and the consequent decreasing of K_s . It seems that as soon as the clogging increase, the ratio P/IMP moves away from the value obtained for clean gravels; this behaviour can confirm the reliability of the procedure.

Table 12. Mean and standard deviation of saturated hydraulic conductivity K_s (N=10) at lab scale for both small and big sizes of clean gravel.

Small size (8-15 x 10 ⁻² m)			Big size	Big size (10-25 x 10 ⁻² m)			
	K_s (m/d)	SD		K_s (m/d)	SD		
IMP	19466	1553	IMP	12135	1591		
permeameter P	30662	3903	permeameter P permeameter	20866	1628		
P/IMP	1.6	-	P/IMP	1.7	-		

Distance from	Diagomatan	IM	Р	P - E	EQ1	P - CA	L EQ1
the inlet (m)	Plezometer	K_s (m/d)	SD	K_s (m/d)	SD	K_s (m/d)	SD
7	1	3999	1300	5045	1130	3072	614
17	4	6896	2022	8629	298	6500	1677
27	7	8629	1769	11819	945	7311	585
7	2	4023	1044	6520	1482	4546	961
17	5	8123	1329	10217	2937	10217	2937
27	8	5810	1182	9014	796	5301	716
7	3	2604	1374	2554	512	2046	575
17	6	9061	1646	13659	1283	8449	793
27	9	6661	1458	10843	1183	6310	471

Table 13. Mean values of saturated hydraulic conductivity K_s (N=4) measured in the nine points inside the full-scale plant and their standard deviation (SD) after 4-years operation.

February 2018								
Distance from the		IMP		P – EC	P-EQ1		P – CAL EQ1	
inlet (m)	Position	K_s (m/d)	SD	K_s (m/d)	SD	K_s (m/d)	SD	
1	H1 A	10066	1024	19988	1820	11280	980	
2	H1 B	13807	1380	21132	1902	11925	1400	
3	H1 C	12175	1100	20762	1154	11717	1150	
1	H2 A	10222	922	20368	1042	10783	1010	
2	H2 B	11564	1453	21348	2170	12047	1340	
3	H2 C	8384	1072	19550	2215	11032	1040	
June 2018								
1	H1 A	10484	489	19544	2001	10341	1059	
2	H1 B	11733	1496	19991	2646	10818	1400	
3	H1 C	12125	961	21164	1260	11522	1200	
1	H2 A	9387	1085	18839	942	9969	498	
2	H2 B	11007	1486	20482	1985	10838	1050	
3	H2 C	9614	1075	17745	2545	9390	1347	

Table 14. Mean values of Saturated hydraulic conductivity K_s (N=4) measured along H1 and H2 pilot beds and their standard deviation (SD).



Figure 30. Variation of K_s measured by pervious (P) and impervious (IMP) permeameters, and their ratio (P/IMP), along the full-scale plant after 4 years-operation.



Figure 31. Variation of Ks values measured by pervious (P) and impervious (IMP) permeameters, and their ratio (P/IMP), along the two beds (H1 and H2) at pilot scale plant.

5.5 Conclusions

The falling head method for saturated hvdraulic conductivity measurements was suitable to assess clogging in CWs at different locations in full scale and pilot scale CWs filled with different size of gravels and after different operation periods (from 1 to 4 years). In particular, the IMP implemented permeameter used with the original equation was the most suitable method to measure K_s in clean gravels (i.e. unclogged) media where the isotropic conditions are still valid and vertical and horizontal K_s are the same. After the starting of the clogging process, vertical K_s alone was representative anymore. implemented An not Ρ permeameter used with a calibrated equation allowed us to evaluate K_s variations taking into account also the influence of clogging in the horizontal direction and also saving time. The decrease of K_s during the CWs operation period was higher in the full scale plant than in the pilot scale plant, probably due to the longer operation life and a higher organic load. As widely reported in literature, the clogging process was more severe in the area close to the inlet of both plants.

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6 Conclusions

This thesis faces several issues related to the complex matter of integrate water resource management, with a special focus on CWs-based DWTFs for WW treatment for reuse, ranging from politics and societal considerations to the analysis of technical elements related to design, operation and performance. Case studies examined and reported are set in the context of Sicily, at the heart of the Mediterranean area, and suggest with a local insight (in semi-arid climate), a methodological inductive approach, allowing to reach wider levels of comprehension of the problems, for universal *modus operandi* and perspective. Specific conclusions below listed should be interpreted at the light of these considerations.

In general, H-CWs have attested to be a resilient and reliable technical solution for decentralized WW treatment unconventional water and recovery and reuse. in Mediterranean climate. However. maintenance and operation options must be carefully evaluated for dynamic adaptation to case-by-case requirements and management needs. In this sense, monitoring procedures of treatment and hydraulic performance of the systems proposed are strategic for ensuring their effectiveness in a range of applications and require optimization, and increased knowledge. Also, sustainable cost and simple O&M, with minimum energy requirements, are accompanied by ancillary benefits like added aesthetical value for the local communities

The **specific conclusions** have been drawn from each of the four parts of the work.

- After evaluating all the factors and constrains still limiting treated WW reuse in agriculture, in Sicily, this study has proposed a GIS-based DSS for assessing the effective potential of TW reuse and for its management, confirming its validity as a useful tool. The latter allowed to integrate WWTPs with collective irrigation areas for crop water demand fulfilment, promote sustainable allocation of water resources and reduce water shortage. However, at the light of chemical and microbiological TW quality assessment, this feasibility study evidenced respectively: the need to improve the actual treatment facilities with more effective tertiary conventional technologies or natural treatment systems; clear disagreement between very strict national standard limits and WHO guidelines, hindering the practice of reuse.

- The studies highlight the strategic role that local governance and policy makers should play for reclaimed water use diffusion and put the focus on the need to involve communities, attract public opinion and build social awareness.

- The study on the performance of a HF unit in a H-CW, confirms its very adaptive aptitude when unusual operation conditions occur. In fact, the hydraulic fluctuations due to untreated effluent overloading, were efficiently dealt by the HF, which consequently got partially clogged after 2 years of operation. Nevertheless, its treatment performance, as also the overall efficiency of the hybrid-TW, was not negatively affected.

In situ measurement of hydraulic conductivity at saturation (K_s) , confirmed to be effective for HF clogging phenomena investigation. The favourable results of maintenance operations can be detected by tracer tests and ERT measurements.

The combination of different methodologies for monitoring the hydraulic behaviour of the HF stage represents an effective and reliable tool for long-term CW management.

The hydraulic monitoring of the HF stage and the consequent maintenance operations, allowed the hybrid-TW to respect for the most the Italian standards for WW discharge.

- This study evidenced that in Sicily, WW treatment solutions like conventional WWTPs and CWs in full and pilot scale, generally do not accomplish national normative due to strict and inappropriate standard limits for water reuse. In fact, there is still a strong discrepancy between the Italian legislation for water reuse and the guidelines proposed by the EU, which also require an implementation process for being integrated at national level. Even when considering conventional parameters and microbial indicators for water quality monitoring for reuse. quantitative and qualitative differences appear obvious. Therefore, the overcoming of this gap probably remains a key point to make effective the WW reuse practice in Italy, also because the intended use would be finally considered. In particular, valuable and underestimated resource like stormwater runoff would profit and be valorised. In fact, when considering the last EU guidelines (2018), the effluent water quality reported for pilot-scale CW case study would fulfil the minimum requirements (matching all chemical parameters and *E. coli*) for reuse in green area irrigation, in contrast with the Italian standard limits, which would not be fully complied.

- In semi-arid climate, a conventional tertiary treatment during dry periods is required for CWs application in stormwater treatment.

-A pilot-scale H-CW system exhibited positive performance alternatively treating two types of WW, highly different in terms of nutrient-richness. However, variable treatment efficiency can be expected due to several factors. In fact, the combination of different effluent quality, with the seasonality, the plant phenology, together with the feeding frequency, must be carefully evaluated. In fact, nutrient removal is strongly influenced by vegetation phenology, and is very low in winter time.

- A "maturation period" (6-12 months) is recommended when a novel CW system is started up. A recirculation phase could be strategic to improve H-CW treatment efficiency and cope with WW variability (particularly in case of nutrient rich effluents during spring and summer time), addressing the most feasible option among the intended uses considered.

- In this study, different effects of various treatment units, in function of the indicator or pathogen considered were observed. FWS unit was strategic for *E. coli* removal, while *C. perfringens* was reduced already at the HF outlet. The latter is an obligate anaerobic, but in spite of its resistance to environmental changes and disinfection processes, perhaps could have been inhibited by possible microbial predation

activities and/or unfavourable environmental conditions occurrance (like atmospheric oxygen transferring in plant rhizosphere).

- The studies underline that the HF treatment unit generally plays the main role in overall removal efficiency of a H-CW. In particular, metals and PAHs removal efficiencies were good in spite of the low inlet concentrations detected.

- The study defines and proposes an optimized methodology of the falling head test for hydraulic conductivity at saturation measurement and clogging assessment in HF-CWs. The choice and implementation of specific devices and the use of relative equations for data modelling, are functions of variable degree of clogging, and can be evaluated case-by-case, allowing to save time in monitoring phases. In fact, vertical and horizontal K_s components must be both considered when clogging process starts to occur, differently to clean substrate, where isotropic conditions are valid, and the vertical K_s is enough representative alone.

- The studies evidenced and confirmed that the clogging process was more severe in the area close to the inlet of both H-CWs examined.

- For future perspective, the investigation on CW hydraulics (better if performed since the start-up phase for the whole system age), optimized by the integration of different monitoring methodologies can provide crossed information, for complete treatment performance assessment and design improvement. This could be very useful for reducing CWs footprint and forecasting systems lifespan.

Annex⁶

Figure A1 is reported as supplementary data of chapter 4, providing further information on the H-CW treatment reliability for WW reuse in agriculture.

Among the parameters reported in tab. 8, the EC should be considered together with the SAR index (as defined in eq. 1) for a proper evaluation of the ultimate effect on water infiltration rate for pursuing suitable irrigation management practices (FAO, 1994). In fig. A1(a), the relationship between these two parameters in terms of soil structural stability is plotted, as described by ANZECC (2000) and DNR (1997). The fig. A1(b) reports the mean EC and SAR values observed at the inlet and the outlet of the hybrid CW during the SBR effluent (I period) and runoff (II-III period) treatment. After the treatment, in both cases, an increase of both parameters can be noted. The EC increment could be explained by evapotranspiration (ET) processes taking place in the treatment beds, while the photosynthetic activity occurring in the FWS units could have risen the pH values (as reported in tab. 8) causing cations precipitation like

⁶ The content of this annex was submitted in Ventura, D., Consoli, S., Milani, M., Sacco, A., Rapisarda, R., Cirelli, G.L., (2019c). On the performance of a novel hybrid constructed wetland for stormwater treatment and irrigation reuse in Mediterranean climate. In: Innovative biosystems engineering for sustainable agriculture, forestry and food production, by Springer International Publishing AG, AIIA International Mid-Term Conference 2019

 Mg^{2+} and Ca^{2+} and therefore an increase of SAR. By comparing the two graphs, it seems that in this case, the CW treatment train could have played a positive role by making more favourable the EC and SAR relationship in the perspective of WW reuse for irrigation. However, too high proportion of sodium (Na⁺) ions relative to Mg²⁺ and Ca²⁺ could degrade the soil stability and reduce plants growth. Finally, the SAR index was always within the Italian standard limits for water reuse. However, among the environmental hazards linked to reclaimed water use in agriculture, the EU proposal of 2018 cites the sodicity (the others are salinization, nutrients imbalance, and toxicity) and reminds the key role of MS in gathering knowledge with the aim to limit concrete environmental risks.



Figure 32. Mean EC and SAR values detected at the inlet and outlet points of the hybrid CW during runoff and SBR effluent treatment (a); the relationship between SAR and EC for soil structural stability (b), as it appears in ANZECC (2000), modified from DNR (1997). Note that 1 dS $m^{-1} = 1000 \mu S cm^{-1}$.

$$SAR = \frac{Na^{+}}{\sqrt{\frac{1}{2}(Ca^{2+} + Mg^{2+})}}$$
(1)

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List of Publications

Peer-reviewed journals

Published

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