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Review

Constructed wetlands to reduce metal pollution from industrial catchments in aquatic Mediterranean ecosystems: A review to overcome obstacles and suggest potential solutions

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ABSTRACT

In the Mediterranean area, surface waters often have low discharge or renewal rates, hence metal contamination from industrialised catchments can have a high negative impact on the physico-chemical and biological water quality. In a context of climate and anthropological changes, it is necessary to provide an integrative approach for the prevention and control of metal pollution, in order to limit its impact on water resources, biodiversity, trophic network and human health. For this purpose, introduction of constructed wetlands (CWs) between natural aquatic ecosystems and industrialised zones or catchments is a promising strategy for eco-remediation. Analysis of the literature has shown that further research must be done to improve CW design, selection and management of wetland plant species and catchment organisation, in order to ensure the effectiveness of CWs in Mediterranean environments. Firstly, the parameters of basin design that have the greatest influence on metal removal processes must be identified, in order to better focus rhizospheric processes on specific purification objectives. We have summarised in a single diagram the relationships between the design parameters of a CW basin and the physico-chemical and biological processes of metal removal, on the basis of 21 mutually consistent papers. Secondly, in order to optimise the selection and distribution of helophytes in CWs, it is necessary to identify criteria of choice for the plant species that will best fit the remediation objectives and environmental and economic constraints. We have analysed the factors determining plant metal uptake efficiency in CWs on the basis of a qualitative meta-analysis of 13 studies with a view to determine whether the part played by metal uptake by plants is relevant in comparison with the other removal processes. Thirdly, we analysed the parameters to consider for establishing suitable management strategies for CWs and how they affect the whole CW design process. Finally, we propose monitoring and policy measures to facilitate the integration of CWs within Mediterranean industrialised catchments.

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1. Introduction

The Mediterranean basin has been identified as one of the most vulnerable regions of the world to climatic and anthropogenic changes (Milano et al., 2012). In the Mediterranean region, human pressure is very strong and occurs almost everywhere, while the biological diversity is remarkably high (Médail and Quézel, 1999). Thus conservation of biodiversity cannot be ensured by protected areas alone, but must depend on a balance between human activities and wildlife (Rhazi et al., 2001). Anthropogenic activity in industrialised zones generates wastes and pollutants on the catchment surfaces that may be washed out to water bodies during storm events (Barbosa et al., 2012). Even state-of-the-art treated industrial wastewaters that are discharged continuously may contribute to significant decline in aquatic faunal populations and biodiversity (Stalter et al., 2013). Drainage systems in catchments usually bring stormwaters and effluents to a point of discharge into the receiving water body, and thus pollution generated and accumulated over the whole catchment is transformed into a point source of pollution upon entry into the aquatic environment (Malaviya and Singh, 2012). In Mediterranean aquatic receiving environments, the pressure resulting from industrial contamination is particularly high (Köck-Schulmeyer et al., 2011). During summer, the natural sources of water may dry up, resulting in rivers and streams that are mainly fed with urban and industrial effluents containing metals[1]. Along the Mediterranean coast, there are many lagoons and associated wetlands with very low water renewal, which are all protected areas for biodiversity conservation (e.g. The Camargue in France, Chauvelon, 1998; The Ichkeul wetland in Tunisia, Casagranda et al., 2006). Anthropogenic activities also impose significant pressure on the groundwater quality and may consequently degrade Mediterranean wetland ecosystems that depend mostly on subsurface water flow (Dimitriou et al., 2008). Given the predicted climatic and anthropogenic changes in the Mediterranean basin in coming decades, with elevation of temperature, reduction of precipitation and population increase (Giannakopoulos et al., 2009; Milano et al., 2012), the contamination pressure on aquatic ecosystems is likely to increase in the medium term (Barbosa et al., 2012).

Among contaminants, metals are currently considered as the main toxic and genotoxic compounds present in hydrosoluble fractions (Maceda-Veiga et al., 2013; Omar et al., 2012). Unlike most of the organic pollutants, metals are not degraded through biological processes, and depending on their forms (complexed, adsorbed onto particles or dissolved), they may enter the trophic web or spread into the sediments where they remain stocked until the physical and chemical conditions change (Forstner and Wittmann, 1981; Devallois et al., 2008). Various industrial processes may induce the release of metals in aquatic environments (Yadav et al., 2012). Diffuse pollutions generated after rainfall in industrial catchments (root and road runoffs, leaching of waste incineration and industrial emissions deposited into river catchments,…) also contribute to high metal loads in rivers (Chon et al., 2012). Therefore, it is necessary to implement restoration measures with the aim of improving water quality (Stalter et al., 2013) and to develop new holistic strategies of management of industrialised catchments (Chon et al., 2012) in order to protect aquatic biodiversity from the impact of metals in Mediterranean environments. Solutions of water quality improvement have to be found at all levels: directly at industries' outfalls but also at the points of discharge of stormwaters into receiving bodies.

Constructed wetlands (CWs) are engineered systems that have been designed to exploit the natural processes involving wetland vegetation, soils and associated microbial assemblages for treating wastewaters. They are designed to take advantage of many of the processes that occur naturally in wetlands, by trying to optimise and speed them up (Vymazal, 2005). Worldwide, they have been increasingly used to successfully remove metals from many types of specific pre-treated industrial effluents (Marchand et al., 2010; Stottmeister et al., 2003). CWs are particularly efficient in warm climates and in areas with sufficiently long daytime periods in winter to support plant growth during all seasons, which is the case in the Mediterranean area. According to Ham et al. (2010), CWs are also a suitable approach for treating and controlling non-point source pollution. CWs are now widely used at catchment scale to attempt to reduce agricultural pollution in temperate environments worldwide (Maillard et al., 2011; Ockenden et al., 2012). These systems can be constructed near natural ecosystems, in particular if designed with notions of landscape-fit, as is the case for Integrated Constructed Wetlands (ICWs) (Everard et al., 2012; Harrington and McInnes, 2009). CWs are seen as an effective solution for wastewater processing for agriculture and rural communities (Babatunde et al., 2008) but, to our knowledge, they have never been used for the treatment of metal pollution from industrialised catchments or directly at industrial effluents outlets in Mediterranean environments. The introduction of CWs between industrialised zones or catchments and receiving water bodies could, however, be a suitable solution to restore or protect the endangered biodiversity of Mediterranean natural wetlands.

Depending on the thickness of the substrate layer, and of the level and direction of the water flow, CWs are classified into four categories, namely surface flow CWs, horizontal or vertical sub-surface flow CWs and hybrid CWs (Vymazal, 2005). In this review, we address more specifically the issue of hybrid CWs given that they enable better adaptation of the system to the wastewater and environmental context, which is a prerequisite for catchment pollution treatment. Moreover, we focused our study on rooted emergent macrophytes as they are the most widely used plants in CWs (Vymazal, 2005) and did not consider free-floating or submerged aquatic plants although they can also be used in treatment systems of this kind (Headley and Tanner, 2012). In this review, we aim to point out the different obstacles that may be overcome to promote the implementation of CWs in order to reduce metal pollution in Mediterranean natural ecosystems. With regard to some of the obstacles, we provide an insight into future prospects. In particular, we dwell on the lack of decision making tools and methodologies for wetland plant selection and we also argue in favour of the need to fit CW design to management strategies. Our review focuses on Mediterranean areas context and needs, but many of the aspects that we develop here may be suitable for other environmental contexts.

2. The Mediterranean context, expected changes and problematic linked to metal pollution

The Mediterranean basin is the region of lands around the Mediterranean Sea. It is defined as a biodiversity hot-spot and is one of the world's major areas for plant diversity, where 10% of the world's higher plants can be found in an area representing only 1.6% of the Earth's surface (Médail and Quézel, 1997). Along the coastlines, vast deltas and numerous lagoons are encountered (Accornero et al., 2007; Bragato et al., 2006; Roche et al., 2009).

The region is under Mediterranean climate, characterized by mild winters and hot and dry summers. The marked orography of the Mediterranean basin often triggers intense events that may cause flash floods and the hot and dry weather in summer causes low flows to be long and severe (Quintana Seguí et al., 2010). The typical hydrologic regime in catchments of the Mediterranean region follows a seasonal pattern that is very irregular due to sudden storms that generate high volumes of runoff because of low bedrock infiltration rates (Fernández-Turiel et al., 2003).

Anthropogenic pressure on rivers has drastically increased in the Northern part of the Mediterranean basin (European countries) with the main expansion of industries in the twentieth century resulting in metal contamination increase in water and sediments (Accornero et al., 2007; Fernández-Turiel et al., 2003; Loumbourdis and Wray, 1998; Medici et al., 2011; Palanques et al., 1998; Tuncer et al., 2001). In the Northern countries, despite significant advances in the field of industrial effluent treatment processes, metal pollution from industrial catchments is still partially discharged into rivers. Many types of industrial activities generate metal pollution through atmospheric deposal into catchments or directly through the release of treated effluents, e.g. metal coating industries (Medici et al., 2011; Soupilas et al., 2008), smelters, fertilizer plants (Milovanovic, 2007), or vehicle recycling factories (Simic and Dimitrijevic, 2012). Moreover, metal pollution may also result from past mining or smelting activities (Affholder et al., 2013; Frau and Ardau, 2003; Olías et al., 2004). In the countries of the Southern part of the Mediterranean basin, industrial effluents are most of the time discharged into rivers without any treatment (Giorgetti et al., 2011; Milovanovic, 2007) resulting in rivers highly contaminated with metals (Herut and Kress, 1997). Nevertheless, far fewer studies assessing metal pollution in rivers have been conducted in these countries in which many industrial activities are responsible of metal releases, in particular tannery and textile industries (Giorgetti et al., 2011; Koukal et al., 2004), fertilizer plants (Herut and Kress, 1997) and uncontrolled landfills (Marzougui and Ben Mammou, 2006).

Future scenarios for water resources in the Mediterranean region suggest changes that will affect water resources qualitatively and quantitatively, enhancing the necessity of improving water management. Many studies have predicted a consistent decrease in river discharge over almost the entire Mediterranean basin mainly because of global warming and increasing water consumption. In addition to changes in the annual amount of water, noticeable alterations have been predicted in seasonality of river flows with probably markedly lower flows in summer (García-Ruiz et al., 2011). In parallel, the predicted population increase (the total population of the Mediterranean basin is projected to reach 269.7 million by the year 2050, while in 2001 only 187 million people lived along the Mediterranean coastline; Milano et al., 2012) will result in an increase of industrial areas in catchments, and thus of metal-polluted water releases into rivers. Aquatic organisms will therefore be concomitantly exposed to natural stressors (reduced surface area for freshwater habitats, changes in velocity patterns and thermal regimes…) and pollution increase. The concurrence of these different stressors may pose tangible risks on both ecosystem integrity and water availability for human use (Petrovic et al., 2011). The flow decrease may have an obvious direct effect on the dilution factor, giving rise to an increase in the concentration of pollutants and thus to a corresponding increase in ecotoxicological risk for the aquatic ecosystems. Moreover, floods can promote sediment resuspension and transport, and thus the subsequent remobilisation of pollutants retained in the sediments. Another specific issue is that sediments often emerge in systems submitted to strong water-level variability, this being the case of Mediterranean aquatic environments. How pollutants accumulated in the top sediment layers will react in contact with the atmosphere, and whether this constitutes a risk for people contacting these sediments during recreational activities (e.g., bathing, fishing, and boating) is unknown (Petrovic et al., 2011).

CWs have been poorly used under Mediterranean conditions to treat variable flows of polluted waters containing metals (Terzakis et al., 2008). It is necessary to encourage field research in the Mediterranean context in order to adapt the technique to the constraints of Mediterranean climate and context. Attention should be paid to flow variability (changing metal concentrations arriving in the CW, risk of flushing of sediments due to storm events, summer dryness for plants), sensitivity of the local biodiversity and ecosystems (high density of protected areas with endangered aquatic biodiversity to be preserved requiring CW environmental integration) and the socio-economical context in certain zones due to water scarcity and predicted evolutions (requiring adapted low cost CW management and monitoring). Therefore, in this specific context, there is a need of finding solutions to overcome these obstacles and to highlight research needs in ecological, organisational and technological plans.

3. Scientific and methodological obstacles concerning CW design

CWs are engineered wastewater treatment systems that encompass a plurality of treatment modules, including biological, chemical, and physical processes, which are all analogous to processes occurring naturally in wetlands (Babatunde et al., 2008). Basically, they are an assemblage of individual basins, one after the other, filled up with mineral substrates and planted with wetland plants (Zhang et al., 2010) (Fig. 1).

In CWs, metal purification performance is based on the combined actions of substrate, plant roots and associated microorganisms (Truu et al., 2009). The active reaction zone of CWs is the rhizosphere or more precisely the so-called mycorhizosphere where physico-chemical and biological processes take place because of the interactions of plants, microorganisms, substrate and pollutants (Stottmeister et al., 2003) (Fig. 2). Macrofauna is another compartment that may have both direct and indirect effects on metal fluxes within CWs via above-cited interactions. In particular, bioturbation is a major engineering process that occurs at the water-sediment interface, including sediment reworking caused by burrowing activities, construction of tubes and burrows, and irrigation of these biogenic structures (Mermillod-Blondin, 2011). On one hand, bioturbation may increase accumulation of metals in substrate (Ciutat et al., 2005) and on the other hand, bioturbation and especially bioirrigation may enhance the mobility of metals and hence their release out of the substrate (Delmotte et al., 2007). The production of galleries by tubificid worms may also increase the supply of oxygen and other electron acceptors at depth into the sediment (Mermillod-Blondin et al., 2008). The influence of macrofauna, as part of the bioremediation processes, should be more taken into account for metal phytoremediation in CWs and studies should explore how to take advantage of these processes for optimising metal treatment.

Therefore, water quality, hydraulics, water temperature, soil chemistry, available oxygen, microbial communities, macro-invertebrates, and vegetation play a functional role and treatment outcomes depend upon how the various components interact (Thullen et al., 2005). Thus, adjustment of the relative size and cover of CWs, the plant species grown on them and the soil media contained within them offers significant potential to control biochemical, physical and chemical and hydraulic conditions to optimise the desired metal removal processes (Tanner and Headley, 2011). On the basis of the existing experiment and reviewed data, several connections between the design variables and physico-chemical and biological metal-removal processes can be made. For example, control of redox conditions can be used to optimise growth of a targeted functional group by altering loading rate, hydraulic design, and mode of operation (Faulwetter et al., 2009).

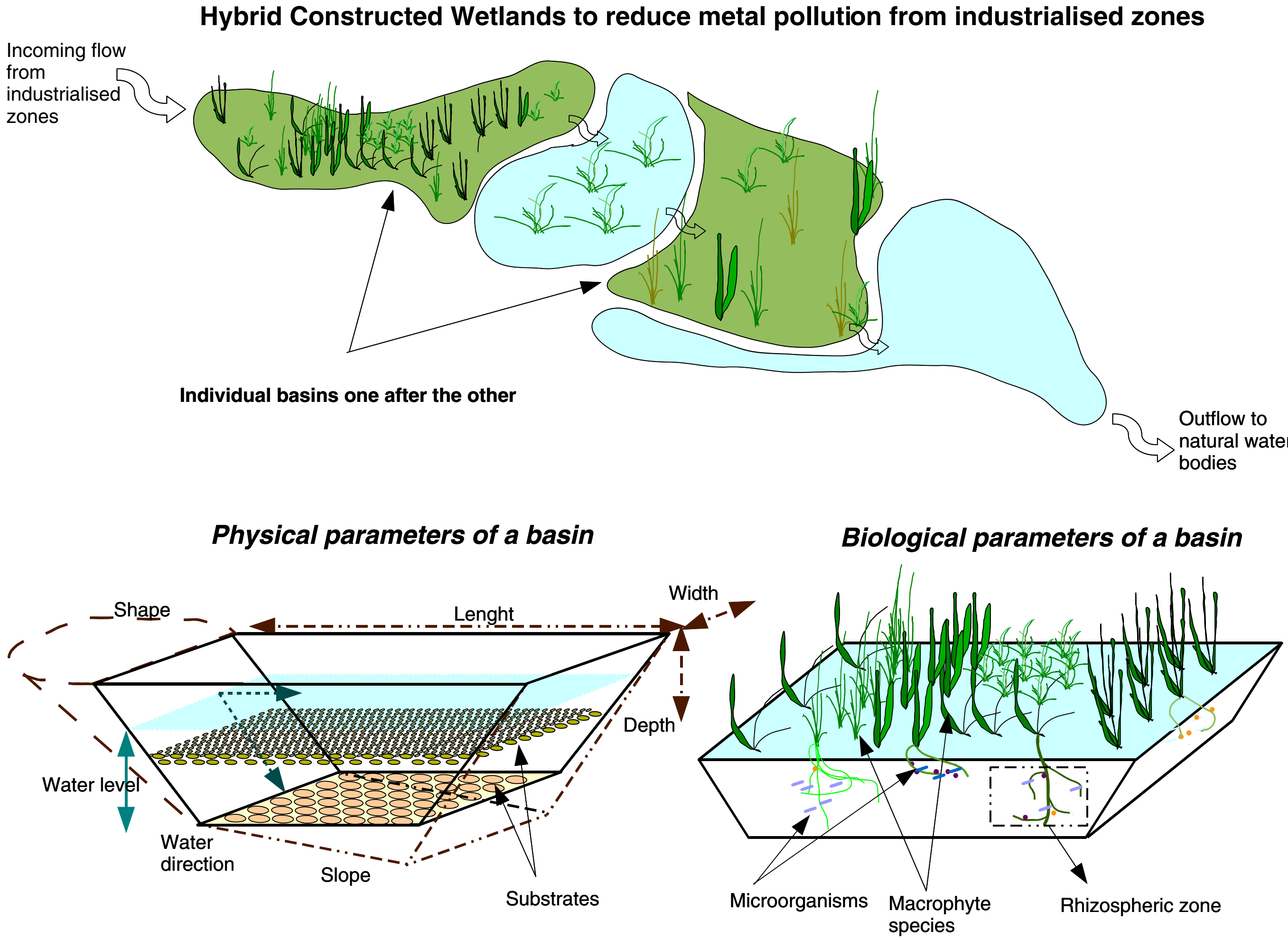

Fig. 1. Schematic representation of hybrid constructed wetlands to reduce metal pollution from industrialised zones.

Nevertheless, in view of the large number of connections between design parameters and metal removal processes, it is necessary to identify the most influential parameters for facilitating CW design. Research concerning CWs has largely dealt with technological design issues, with the active reaction zone of the rhizosphere being essentially treated as a ‘black box’ where the only issues of concern were the inlet and outlet loads (Malaviya and Singh, 2012; Stottmeister et al., 2003). As a result, CWs are often designed with little regard for the numerous interrelated biological, chemical, and physical processes that occur within an individual wetland, and may, therefore, be missing some critical elements for optimising treatment function and system sustainability (Thullen et al., 2005). More research is therefore required to quantify the effects of the design variables on the above-cited processes so that their influence can be optimised for a specific treatment objective (Faulwetter et al., 2009).

To this end, we have summarised on a single diagram the relationships between the design parameters of a CW basin and the physico-chemical and biological processes of metal removal, on the basis of 21 mutually consistent studies (Fig. 3). We noted that selection of the plant species to be planted can serve as an effective tool to alter the wetland's microbial composition (Calheiros et al., 2009; Collins et al., 2004) as well as the physical and chemical reactions with metals. According to Faulwetter et al. (2009) regarding the well-established interactions between plants and microorganisms in agronomic settings, it seems quite probable that manipulation of plant species might be as important for the enhancement of desirable microbial functional groups as wetland type. However, this hypothesis needs to be tested by further research. Nevertheless, vegetation is often the last consideration in the design of CWs (Thullen et al., 2005). Selection of wetland plants, optimization of contaminant uptake by plants as well as determination of the best technical design parameters are still challenging tasks for environmental engineers and researchers (Brisson and Chazarenc, 2009; Zhang et al., 2010). In the following chapters, we focus on the concern of plant species selection and on the link between CW design and plant management, with the aim of providing elements that may offer decision making aid and practical support.

4. Decisional and methodological obstacles concerning emergent wetland plant-selection

4.1. A lack of methodologies for plant diversity selection

Most of the CWs in the world contain low floristic diversity or are even planted with one single species. Only few researchers have tried to evaluate the benefits of polycultural wetlands on purification (Liang et al., 2011). Yet this attempt to improve the depurative performance of CWs is entirely reasonable in the light of various elements. First, it is well known that as the number of plant species increases, so does the complexity of their interactions with each other and with their environment, thereby increasing the resilience of the ecosystem and its ability to adapt to change (EPA, 1994). Previous studies on aquatic microbial diversity found that plant species diversity also confers spatial and temporal stability on several ecosystem functions (Naeem and Li, 1997; McGrady-Steed et al., 1997). Even if the idea that greater plant diversity allows greater plant biomass production is widely accepted, it is only recently that multi-scale experiments have demonstrated the links

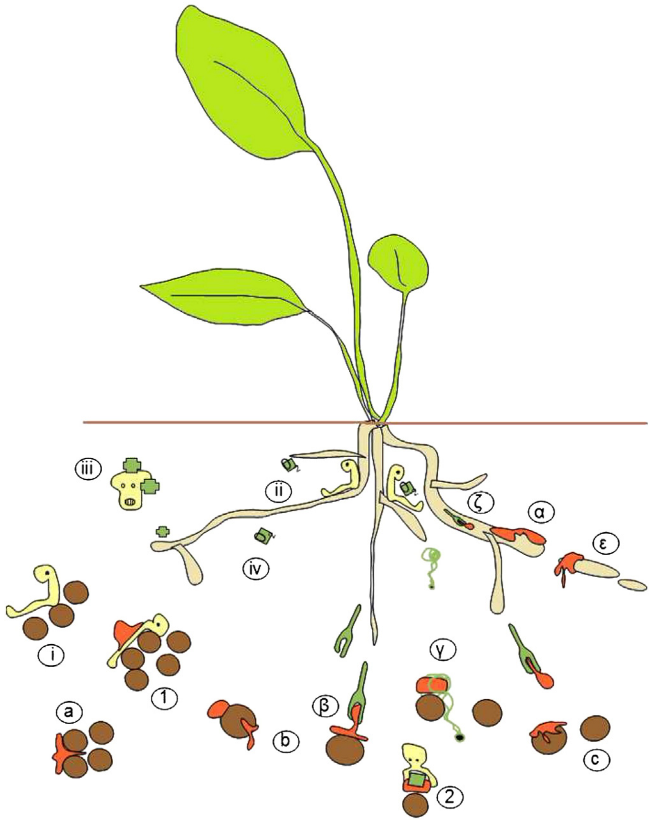

Fig. 2. Schema of the principal interactions that take place in CWs between metals and the purification components. Text associated to the Fig. 2: *Plants–metals interactions*: metals (in orange) can adsorb on root surface (α) or on decaying organic matter (ε) [Guilizzoni, 1991]. They can be solubilised (β) or precipitated (γ) by plant secretions. Solubilised metals can be absorbed by plant, stocked in roots or translocated into shoots (ζ) [Stottmeister et al., 2003]. *Microorganisms–metals interactions*: metals can adsorb on the surface of microorganisms (in yellow) (1), or be absorbed into them [Sheoran and Sheoran, 2006]. Microorganisms may modify the chemical speciation and bioavailability of metallic pollutants (2). *Substrate–metals interactions*: metals can adsorb on substrate (in brown) (c) [Sheoran and Sheoran, 2006], be absorbed into it (b) or they can be mechanically filtered by it (a) [Stottmeister et al., 2003]. *Substrate–plants–microorganisms interactions*: Substrate (i) and plants (ii) are microorganisms' habitat [Marchand et al., 2010], [Stottmeister et al., 2003]. Plants can produce toxic components against pathogenic microorganisms (iii) [Stottmeister et al., 2003]. Plant roots can release O_2 in the rhizosphere, modifying the redox gradients and enabling the formation of many ecological niches that promote a multitude of microbial processes [Faulwetter et al., 2009] (iv) (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

between biodiversity and ecosystem functioning, particularly for aquatic ecosystems (Cardinale et al., 2006; Loreau, 2009). Moreover, wetland plant species also have different root morphology and distribution; that suggests that polycultural CWs may be more efficient because of temporal and spatial compensations in plant root distribution (Liang et al., 2011). Resilience and adaptability, stability of biomass production and root expansion are all useful capacities for a CW that receives toxic and variable discharges from an industrialised zone or catchment (Barbosa et al., 2012). In addition to that, in a context of contaminant mixtures, creating combinations of plant species that vary in their features is a possible strategy to maximise phytoremediation (Zhang et al., 2010).

But it is not only a question of diversity: the choice of plants is also an important issue in CWs, as they must survive the potentially toxic effects of the wastewater and its variability (Calheiros et al., 2007). The lifetime of CWs is also crucially dependent on the selection of plant species (Liu et al., 2007), this being decisive for the improvement of metal removal efficiency (Liu et al., 2010). In Mediterranean environments, we suggest that the macrophytes be selected among local plant species, not only to avoid gene flow and introduction of exotic species but also to develop local biodiversity. Moreover, autochthonous species that are growing in polluted sediments with long-term exposure are certainly the organisms best suited both to local pollution and environmental conditions (Grant, 2010). Although there are several studies involving different macrophytes and types of effluent, there is a lack of studies focusing on ways of selecting the best plant species (Brisson and Chazarenc, 2009). Thus, further research is required to develop some practical methodologies for selecting the best plant species among the biodiversity of aquatic receiving bodies from industrialised Mediterranean catchments.

4.2. Is the metal uptake by plants significant in the whole removal process?

In order to develop methodologies, it is required to clearly determine which plant traits are helpful in the purification processes of metals. For instance, the question of the role of metal uptake by plants in the whole removal process has to be elucidated.

Metal removal processes in CWs are extremely complex and have been well synthesised by many authors such as Guilizzoni (1991), Sheoran and Sheoran (2006) or more recently by Marchand et al. (2010). They are the result of biotic and abiotic interactions that can produce metal precipitation (with phosphates, sulphides, hydroxides, organic salts, etc.), mechanical retention (when they are included in suspended matter), complexation (with organic matter), adsorption/desorption (on Fe and Mn oxides, clay particles, root surfaces, etc.) or their absorption (by plants or microorganisms). On the whole, a general consensus in the scientific literature is that substrate acts as the primary sink for metals (Weis and Weis, 2004; Ye et al., 2001). Nevertheless, concerning the role of plant uptake in metal removal, as has been noted by Lee and Scholz (2007), opinions differ. Depending on the studies, authors either conclude that metal uptake by vegetation is significant (Bragato et al., 2006; Cheng et al., 2002; Collins et al., 2005; Galletti et al., 2010; Grisey et al., 2012; Khan et al., 2009; Liu et al., 2007, 2010; Maine et al., 2009) or that it is of minor importance (Hadad et al., 2006; Manios et al., 2003; Mitsch and Wise, 1998; Nyquist and Greger, 2009; Yang et al., 2006). These contradictory results do not facilitate CW design and management choices for metal removal. The main and most strongly structuring factors for metal uptake efficiency thus remain to be elucidated. Our hypothesis is that ranges of metal concentrations and their bioavailable fractions in wastewater and also types of plant biomasses are part of the answer. We therefore chose a panel of studies in which authors have conducted experiments in CWs (at any scale) to treat effluents polluted with several metals, and in which they have measured both the concentrations of metals in plants and the biomass production. The aim was to conduct a qualitative meta-analysis confronting the authors own conclusions on the role of plant uptake with data being comparable between studies i.e. the percentages of annual pollutant input stored in plant biomass, the CW design parameters, and the effluent characteristics. This qualitative meta-analysis aims at highlighting the main factors that govern the plant uptake efficiency during metal treatment in CWs, independently of climate. Thus, the selected studies were conducted under different types of climate (4 in temperate, 3 in sub-tropical, 3 in continental, 2 in equatorial, 1 in Mediterranean). Nevertheless, in view of having suitable conclusions for Mediterranean areas, the majority of the selected studies used plant species that are also encountered in Mediterranean aquatic ecosystems and used in CWs (i.e. *Carex rostrata*, *Cyperus alternifolius*, *Juncus effusus*, *Phragmites australis*, *Phragmites communis*, *Persicaria hydropiper*, *Typha domingensis*, *Typha latifolia*,…).

Above all, it appears that it is quite difficult to compare the part of metal removal linked with substrate and the part linked with vegetation because it depends on the plant species, the sampling time (tissue age and period of the year) and frequency, organs analysed (stems and/or leaves and/or flowers, lateral and/or main roots and/or rhizomes, whole plants, etc.) (Guilizzoni, 1991), the effluent parameters (e.g. the metals present and their chemical forms, the total flow rate), design characteristics of CW (size, shape, type of substrate), its age, the

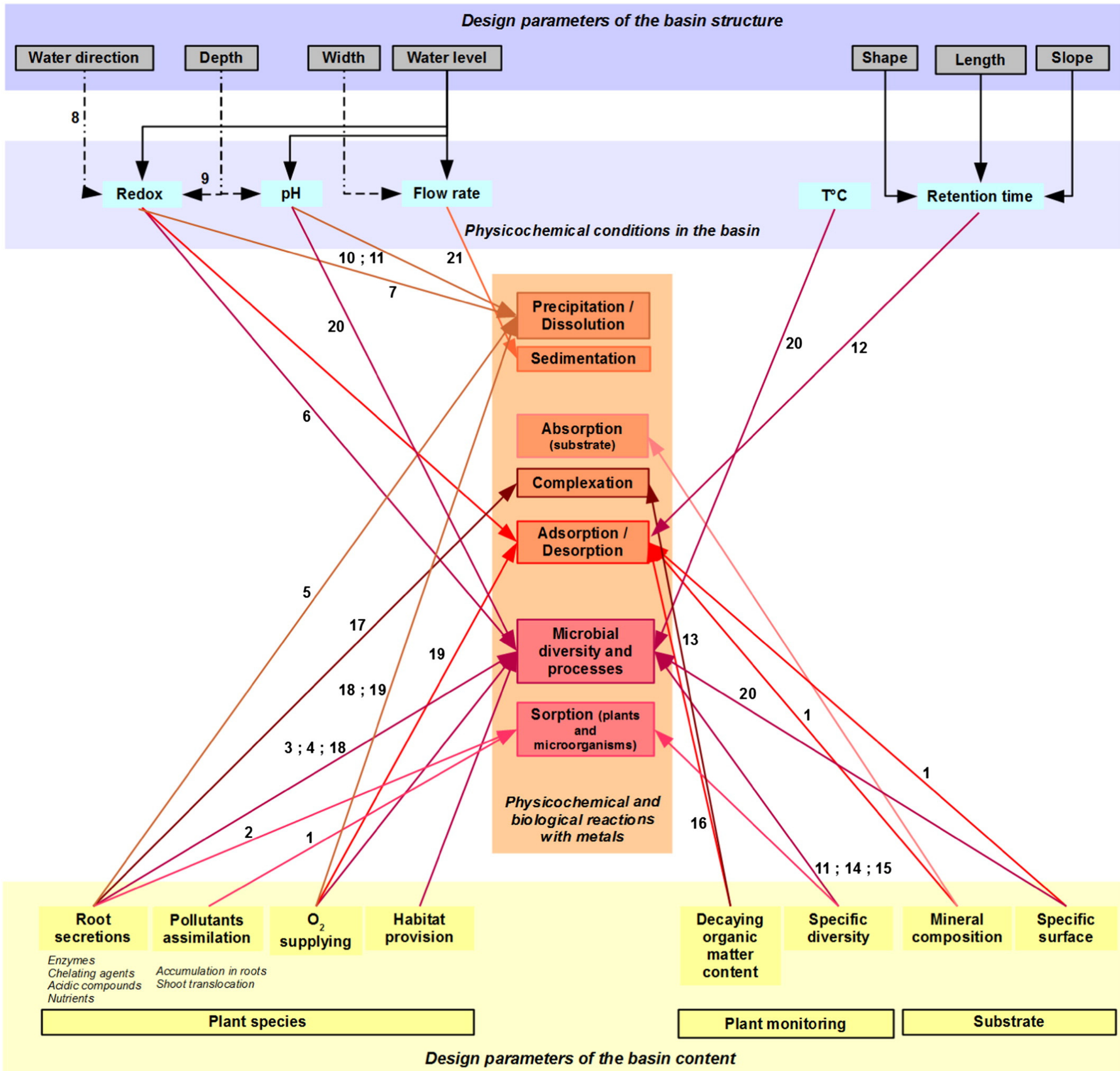

Fig. 3. Connections between design parameters of a CW basin and physico-chemical and biological reactions with metals. Text associated to Fig. 3: [1] Sheoran and Sheoran, 2006; [2] Kabata-Pendias, 2004; [3] Edwards et al., 2006; [4] Huang et al., 2012; [5] Yang et al., 2005; [6] Faulwetter et al., 2009; [7] Marchand et al., 2010; [8] Yalcuk and Ugurlu, 2009; [9] Yadav et al., 2012; [10] Guilizzoni, 1991; [11] Collins et al., 2004; [12] Gerhardt et al., 2009; [13] Banat et al., 1974; [14] Calheiros et al., 2010; [15] Zhang et al., 2010; [16] Le Goff and Bonnomet, 2004; [17] Tanner and Headley, 2011; [18] Stottmeister et al., 2003; [19] Hinsinger, 2001; [20] Truu et al., 2009; and [21] Zhang et al., 2012.

working scale, the climate, and other parameters, that are not necessarily given in the Materials and Methods sections of the papers or even known by the authors. To better take into account the role of these neglected parameters, we selected only the studies in which the authors gave enough data about the metal content in plant or substrate and the parameters that might affect this repartition. These parameters are summarised in Tables 1 to 4. In order to compare all the studies, final results were expressed as percentages of annual inflow rate stored in plant biomass with regard to the loading rates (input of metal in g/m^2 of the CW/day). We also specified the analysed plant organs and the metal concentrations found in the biomass. Finally, we mentioned the author's own conclusions and we classified the studies in two groups (Tables 3 and 4) depending on their opinion regarding the efficiency of plant uptake in the whole purification process.

Among the authors that came to the conclusion that the proportion of metals exported in plants is minor in comparison with the proportion stored in substrate, two groups can be distinguished:

– The first group includes studies 1 to 3 in which the effluent to be treated is related to acid mine drainage (AMD). This type of effluent contains very high metal concentrations compared to the other effluents concerned by our analysis. This can be seen in the differences of loading rates (the Fe average loading rate for the AMD effluents is 10 times higher than the Fe average loading rate for the other effluents) in Tables 2 and 4. In addition, the pH is generally low in AMD. In this group of studies, Nyquist and Greger (2009), Mays and Edwards (2001), and Mitsch and Wise (1998) reached the conclusion that plant metal uptake is of low importance compared to retention via

Table 1
Details of the selected studies—part 1.

Study	Reference	Geographical location	Climate	Scale	Type of CW[a]	Size (m^2)	Depth (m)	Substrate	Duration of pollutant implementation before collecting (months)	Plant species (in which metals concentrations were measured)	Type of effluent
1	Nyquist and Greger, 2009	Northen Sweden (65° 04N, 18° 44E).	Continental	Pilot	SF	22	0.3	Mine tailings	**24**	*Carex rostrata*, *Eriophorum angustifolium*, & *Phragmites australis*	Acid mine drainage
2	Mays and Edwards, 2001	USA, Tennessee	Temperate	Extensive	n.d.	5700	0.15 to 0.3	Sediments	**84**	*Scirpus cyperinus* (L.) Kunth, *Typha. latifolia* L., & *Juncus effusus* L.	Acid mine drainage
3	Mitsch and Wise, 1998	USA, Ohio	Humid continental	Extensive	Mixed	3869	0.6 to 1.4	Spent mushroom compost, clay, limestone	**12**	*Typha latifolia*	Acid mine drainage
4	Ye et al., 2001	USA, Pennsylvania	Humid continental	Extensive	SSF	1300	0.45 to 0.6	Spent-mushroom compost, native soil material	**24**	*Typha latifolia*	Coal combustion by-product leachate from an electrical power station
5	Lee and Scholz, 2007	Scotland, Edinburgh	Cold Temperate	Small	SSF	0.031	0.83	Gravel, sand	**24**	*Phragmites australis*	Urban runoff
6	Lesage et al., 2007	Zemst, Belgium	Temperate	Extensive	SSF	1300	0.5	Gravel	**42**	*Phragmites australis*	Pre-treated-domestic wastewaters and stormwaters

[a] Type of CW. SF: Surface flow; and SSF: Subsurface flow.
Duration of pollutant implementation before collecting is given in bold.

the filling media. In fact, even though the metal concentrations in plant biomass can be very high (e.g. 14050 μg Fe/g DW of *P. australis* belowground biomass, or a mean of 2076 μg Mn/g DW aboveground biomass in *S. cyperinus*, *T. latifolia*, and *J. effusus*), the quantity exported in plants remains negligible compared to the annual input of metals that is trapped in the substrate. Among these studies, only one looks at the chemical speciation of metal loads (Mays and Edwards, 2001), by determining the extractable forms of metals in influents and in sediments. Nevertheless, these authors do not give the ratio between extractable and total metal content that would have been an indicator of metal bioavailability.

– The second group includes studies 4 to 6 in which the authors conclude in a minor role of plant metal uptake on the basis of its aboveground (harvestable) biomass only. In fact, Lee and Scholz (2007), Ye

Table 2
Details of the selected studies—part 2.

Study	Reference	Geographical location	Climate	Scale	Type of CW[a]	Size (m^2)	Depth (m)	Substrate	Duration of pollutant implementation before collecting (months)	Plant species (in which metals concentrations were measured)	Type of effluent
7	Cheng et al., 2002	Germany, Cologne	Temperate	Small	SSF	2	0.7	Gravel, sand	**5**	*Cyperus alternifolius*	Pre-treated domestic effluents
8	Hadad et al., 2006	Argentina, Santa Fé	Humid subtropical	Pilot	SF	18	0.3	Soil	**1 to 14**	*Typha domingensis* & *Panicum elephantipes*	Wastewater from a tool industry
9	Liu et al., 2010	China, Changzhou	Equatorial	Small	SSF	2	0.25	Sandy soil	**1.5**	19 species[i]	Artificial wastewater
10	Galletti et al., 2010	Italy, Ferrarra	Mediterranean	Pilot	SSF	30	0.6 to 1.2	Gravel	**10**	*Phragmites australis*	Domestic wastewater
11	Maine et al., 2009	Argentina, Santa Fé	Humid subtropical	Extensive	SF	2,000	0.5 to 0.8	Sediment	**16**	*Eichhornia crassipes*	Wastewater from a tool factory
12	Maine et al., 2009	Argentina, Santa Fé	Humid subtropical	Extensive	SF	2,000	0.5 to 0.8	Sediment	**36**	*Typha domingensis*	Wastewater from a tool factory
13	Liu et al., 2007	China, Changzhou	Equatorial	Small	SSF	2	0.25	Sandy soil	**1.5**	19 species[i]	Artificial wastewater

[i] *Polygonum lapathifolium* L., *Polygonum hydropiper* L., *Eclipta prostrata* L., *Aster subulatus* Michx, *Cyperus iria* L., *Cyperus difformis* L., *Fimbristylis miliacea* (L.) Vahl, *Oryza sativa* L., *Isachne globosa* (Thunb.) Kuntze, *Phragmites communis* Trin, *Eleusine indica* (L.) Gaertn, *Digitaria sanguinalis* (L.) Scop, *Zizania latifolia* (Griseb.) Stapf, *Echinochloa oryzicola* (Ard.) Fritsch, *Echinochloa caudata* Roshev, *Echinochloa crus-galli* (L.) Beauv, *Alternanthera philoxeroides* (Mart.) Griseb, *Monochoria vaginalis* (Burm. f.) Presl, *Aeschynomene indica* L.
Duration of pollutant implementation before collecting is given in bold.

Table 3
Studies indicating a minor role of plant uptake for metal removal.

Study	Metal	Loading rate (mg/m²/d)	Metal concentrations (μg/g DW) in belowground biomass	Metal concentrations (μg/g DW) in aboveground biomass	% of annual or total (when < 1 year) inflow rate stored into plant biomass (aboveground, belowground, total biomass)	Authors' own conclusions on the plant interest
1	Fe	**8500**	14050[P.a]	1471[P.a]	<**0.5**	"Emergent plants and the wetlands constructed in this study were thus inadequate to treat the very harsh AMD at the Kristineberg mine site."
	Zn	**180**	2524[P.a]	148[P.a]	<**0.5**	
	Cu	**50**	300[P.a]	34[P.a]	<**0.5**	
	Cd	**0.18**	29[P.a]	0.13[P.a]	<**3**	
2	Fe	**497.2**	28660[mean, fall]	327[mean, fall]	**1**	"Data indicate that Mn, Zn, Cu, Ni, B, and Cr are being accumulated in the plants at all three wetlands, although accumulation of metals by these plants accounts for only a small percentage of the removal of the annual metal load supplied to each wetland."
	Pb	**0.023**	6.1[mean, fall]	0.6[mean, fall]	n.d.	
	Al	**0.23**	1531[mean, fall]	78[mean, fall]	n.d.	
	Mn	**66.67**	2012[mean, fall]	2,076[mean, fall]	**2**	
	Cd	**0.07**	6.0[mean, fall]	0.1[mean, fall]	n.d.	
	Zn	**0.1**	23[mean, fall]	7.5[mean, fall]	n.d.	
3	Fe	**5800**	n.d.	2500[T.l]	**0.07**	"Plants appeared to have very little to do with metal retention in the wetland even though concentrations in the aboveground portions of the plants were generally higher than background concentrations in non-polluted environments."
	Al	**3308**	n.d.	613[T.l]	n.d.	
	Mn	**249**	n.d.	282[T.l]	n.d.	
4	Fe	**776.1**	7440[T.l, cell 1]	268[T.l, cell 1]	**0.91**	"Metal accumulation in the APS wetland, Pennsylvania, tended to be greater in the surface layers of sediments as well as in the rhizomes of cattail. The accumulation of metals in living shoot tissues of cattail and the accumulation in cattail fallen litter […] were relatively minor sinks in comparison with the sediments."
	Mn	**546.1**	1650[T.l, cell 1]	2010[T.l, cell 1]	**4.18**	
	Co	**3.64**	11[T.l, cell 1]	0.76[T.l, cell 1]	**0.19**	
	Ni	**9.1**	16[T.l, cell 1]	2[T.l, cell 1]	**0.38**	
5	Ni	**74**	n.d.	n.d.	**0.3**	"The amount of metals removed by harvesting was negligible (<1% on average) when compared to those retained in the filters."
	Cu	**70**	n.d.	n.d.	**0.1**	
	Cr	**3.9**	n.d.	n.d.	n.d.	
6	Cd	**0.04**	0.47[1]	0.071[l]	**0.5**[*]	"For all metals, less than 2% of the mass removed from the wastewater after passage through the reed bed is accumulated in the aboveground reed biomass and can be removed by harvest."
	Cu	**1.6**	44[1]	2.2[l]	**0.5**[*]	
	Pb	**1.12**	20[1]	1.1[l]	**0.4**[*]	
	Zn	**10.4**	184[1]	23[l]	**1**[*]	
	Cr	**0.16**	22[1]	0.95[l]	**2**[*]	
	Ni	**0.64**	9.3[1]	0.29[l]	**0.5**[*]	
	Al	**39.52**	2534[1]	21[l]	**0.04**[*]	
	Fe	**34.4**	6433[1]	114[l]	n.d.	
	Mn	**3.2**	58[1]	60[l]	n.d.	

[P.a]: in *Phragmites australis*; [T.l]: in *Typha latifolia*; [s]: sediment; [mean]: mean between plant species; [fall]: in autumn; [cell 1]: in the first cell of the CW; [1]at 1 m from the inlet; [l] in leaves; and [*]percent of the metal mass removed from the wastewater.
Loading rate and % of annual or total (when < 1 year) inflow rate stored into plant biomass are given in bold.

et al. (2001) and Lesage et al. (2007) did not determine either the belowground biomass production or the metal concentrations in belowground biomass (for experimental reasons). In this latter study, total and dissolved metal concentrations were determined, and this fraction represented only between 10 and 35% of total metal concentrations. Lee and Scholz (2007) added heavy metals in the form of salts, but altered the initial pH by adding NaOH, thus modifying concomitantly the chemical forms and their bioavailability. Ye et al. (2001) only determined the total metal content, which makes it impossible to draw conclusions on the influence of chemical form in metal uptake.

In seven other studies (7 to 13) in which the authors measured the capacity for metal uptake by the whole plant biomass (including in its belowground parts) and in which there is no AMD effluent to treat, the authors came to the conclusion that metal uptake by plants does play a role in metal removal. Among these studies, those of Cheng et al. (2002) and of Liu et al. (2007, 2010) have used reconstituted wastewater with heavy metals added in the form of salts. In this case, added metals – totally in bioavailable forms – are more easily absorbed by plants: results showed that between 90 and 100% of metals were removed with a significant proportion retained in plants. In the studies of Liu et al. (2007, 2010), metals were mainly stored by *Alternanthera philoxeroides*, *Zizania latifolia*, *Echinochloa crus-galli*, *Polygonum hydropiper* (in above- or belowground parts, depending on the metal considered) and Cheng et al. (2002) found that metals were mainly stored in lateral roots by *C. alternifolius*.

Several other studies (Bragato et al., 2006; Collins et al., 2005; Khan et al., 2009; Yang et al., 2006) present conclusions that are consistent with our meta-analysis but we did not include them in our table analysis because the authors did not give enough data to calculate the percentage of annual pollutant input stored in plant biomass. Thus, on the basis of our meta-analysis, the quantity of metals that is removed in plants compared to the quantity stored in the substrate may be significant if the bioavailability of metal is high and the belowground biomass is taken into account, except in the case of AMD treatment. According to Stottmeister et al. (2003), the proportion of metals removed by plant uptake is insignificant when mine drainage waters are being treated because the amount that can be accumulated is only a small fraction of the total load of metals in such concentrated wastewater. Indeed, even if the uptake of metals increases with increasing external concentrations, the uptake is not linear in correlation to the concentration increase due to a saturation effect in plant tissues (Greger, 2004).

The lifetime of the substrate could be notably enhanced with a frequent harvest of plants on the basis of the percentages of the annual inflow rates stored in the plant biomass found in some of the studies previously mentioned (e.g. studies 7, 9, 11, 12 and 13). Moreover, given that metals can be released from the substrate of the CW to water when

Table 4
Studies indicating an important role of plant uptake in metal removal.

Study	Metal	Loading rate ($mg/m^2/d$)	Metal concentrations (μg/g DW) in belowground biomass	Metal concentrations (μg/g DW) in aboveground biomass	% of annual or total (when < 1 year) inflow rate stored into plant biomass (aboveground, belowground, total biomass)	Authors own conclusions on the plant interest
7	Al	**94.8**	596$^{C.a,\ main\ roots}$	27$^{C.a,\ leaves}$	**13**	"In summary, *C. alternifolius* has a great potential for heavy metal phytoremediation, especially for Cu, as well as for Mn and Zn."
	Cd	**1.08**	9.2$^{C.a,\ main\ roots}$	0.3$^{C.a,\ leaves}$	**6.6**	
	Cu	**124.8**	2610$^{C.a,\ main\ roots}$	7.1$^{C.a,\ leaves}$	**32.3**	
	Mn	**36.24**	121$^{C.a,\ main\ roots}$	68.9$^{C.a,\ leaves}$	**37.2**	
	Pb	**1.24**	6.2$^{C.a,\ main\ roots}$	1.2$^{C.a,\ leaves}$	**14.6**	
	Zn	**510**	2490$^{C.a,\ main\ roots}$	77.3$^{C.a,\ leaves}$	**5.1**	
8	Fe	**494**	n.d.	n.d.	n.d.	"*T. domingensis* proved to be highly efficient for the treatment of wastewater."
	Cr	**6.38**	26.5$^{C.a}$; 110$^{T.d}$	28$^{C.a}$; 36$^{T.d}$	**7**	
	Ni	**9.72**	11.5$^{C.a}$; 70$^{T.d}$	27.5$^{C.a}$; 32$^{T.d}$	**2**	
	Zn	**3.22**	74$^{C.a}$; 67$^{T.d}$	97.9$^{C.a}$; 39$^{T.d}$	**4**	
9	Cu	**10.6**	200.2$^{M.v}$	34.2$^{M.v}$	**8.8**	"Sedimentation was the principal process or the removal of heavy metals from wastewater in constructed wetland.[…]. However, phytoextraction was also important for some metals, such as Cr in the present research."
	Cr	**5.3**	142$^{M.v}$	33.5$^{M.v}$	**20.5**	
	Ni	**10.6**	266.7$^{M.v}$	44$^{M.v}$	**14.4**	
10	Cu	**0.61**	17$^{P.a}$	5.7$^{P.a}$	**34**	"Plants, such as *P. australis*, participate in heavy metals removal from wastewaters."
	Ni	**2.37**	4.2$^{P.a}$	2$^{P.a}$	**1.8**	
	Zn	**15.73**	66.2$^{P.a}$	56$^{P.a}$	**6.2**	
11	Fe	**387**	n.d.	n.d.	n.d.	"During the *Eichhornia crassipes* dominance, contaminants were retained in the macrophyte biomass"
	Cr	**0.9**	n.d.	n.d.	**88**	
	Ni	**1**	n.d.	n.d.	**93**	
	Zn	<**2.5**	n.d.	n.d.	**98**	
12	Fe	**387**	n.d.	n.d.		"In the *T. domingensis* dominance stage, contaminants were retained in sediment and in the macrophyte biomass."
	Cr	**0.9**	n.d.	n.d.	**30**	
	Ni	**1**	n.d.	n.d.	**13**	
	Zn	< **2.5**	n.d.	n.d.	**41**	
13	Cd	**2.66**	171.21$^{M.v}$	21.26$^{M.v}$	**19.85**	"The results indicated that the plants, in constructed wetland for the treatment of wastewater polluted by heavy metals, can play important roles for removal of heavy metals through phytoextraction."
	Pb	**10.6**	710.96$^{M.v}$	85.98$^{M.v}$	**22.55**	
	Zn	**26.66**	716.35$^{M.v}$	211.41$^{M.v}$	**23.75**	

$^{C.a}$: in *Cyperus alternifolius*; $^{M.v}$: in *Monochoria vaginalis*; $^{T.d}$: in *Typha domingensis*; $^{P.a}$: in *Phragmites australis*; ^{leaves}: in leaves; and $^{main\ roots}$: in main roots.
% of annual or total (when < 1 year) inflow rate stored into plant biomass.

physical and chemical conditions change (Zhang et al., 2012), the frequent export of metals through plant harvesting is a solution to reduce the impact of such events. Nevertheless, in many cases, this implies that both the roots and shoots are harvested, which has consequences for the CW design and management (issues addressed in Section 5).

4.3. Which criteria to select plants for metal removal?

Different criteria may be taken into account for the selection of wetland plant species for their metal-uptake ability. There are above all three defined types of metal-accumulating plant species:

– Firstly, the plant species qualified as hyperaccumulators that strongly accumulate metals in their aboveground biomass to a level that would be toxic for other plants. There are several criteria to define a hyperaccumulator species: the threshold value of the metal accumulated in the plant must be above a certain concentration (e.g. 100 mg/kg for Cd), the bioaccumulation factor and the translocation factor must be both greater than 1, and the aboveground biomass of the plant should not decrease significantly at the concentration of the critical value (Sun et al., 2009). However, most work on metal hyperaccumulators has been done on terrestrial plants and to date no emergent wetland plants have been identified as hyperaccumulators (Marchand et al., 2010).
– Secondly, the species that are non-hyperaccumulators but that have a natural capacity to uptake metals (Soda et al., 2012) and that produce a high biomass (Chon et al., 2012). In this perspective, the use of cattails and reeds is very common for metal treatment in CW, as cited above i.e. *P. australis* in 5 studies and *Typha spp.* in 6 studies. Bioconcentration factor (BCF) (from water or sediment to root/rhizome) and translocation factor (TF) (from root/rhizome to shoot) of metals are standard determining criteria for the selection of these plant species (Grisey et al., 2012). A species that is able to concentrate metals within the whole plant at concentrations 100 times higher than that in the wastewater (BCF) should be considered a good accumulator. Nevertheless, the TF value should not always be considered as an important criterion regarding phytoremediation of water (unlike soil phytoremediation) when both roots and shoots can be harvested easily (Ha et al., 2011). Not only does the metal extraction capacity of the plant species matter, but also its biomass productivity. For instance, Quan et al. (2007) found that the aboveground metal pools of *Spartina alterniflora* were significantly greater than those of two native species i.e. *P. australis* and *Scirpus mariqueter* in China and that this was mainly the result of a greater net primary production in plots of *S. alterniflora*. Goulet et al. (2005) found that *Lemna minor* had a higher Al uptake rate compared to *T. latifolia*, but that the latter was responsible for 99% of the total Al uptake because it yielded the highest biomass.
– Thirdly, species working as excluders, restricting metal transport to the aboveground part in order to keep shoot concentration as low as possible and keeping high levels of metals in the roots (Bragato et al., 2006). Considering our meta-analysis, many wetland plants act as excluders.

Among these 3 categories of species, there are unspecific collector species and metal-specific ones. The unspecific collectors such as *A. philoxeroides*, *Z. latifolia* (Liu et al., 2007, 2010), *Canna indica* (Yadav et al., 2012), *Phalaris arundinacea* (Deng et al., 2004) and *Amaranthus* *blitoides* (Del Ŕio et al., 2002) are able to take up several metals at the same time. In contrast, metal specific species (like *Elodea canadensis* for mercury) are able to take up one particular metal better than other species do (Mganga et al., 2011; Mortimer, 1985). Planting a CW should be done favouring all these complementary plant types in order to treat a mixture of metals. First, high biomass plant species with unspecific metal uptake (in aboveground or belowground parts) such as *P. arundinacea* could be used. Moreover, a mixture of plant species each accumulating a different type of metal, in the specific environmental conditions required for the treatment, may also be used.

In addition to their metal-uptake ability, wetland plants possess other capabilities that can be helpful in metal removal. Plants represent a physical obstruction to flow and act as a filter system for particles. Mitsch and Wise (1998) found that vegetation provides sites for iron sedimentation. Triboit et al. (2009) suggest that macrophytes may be used in CWs to prevent wind dispersion of polluted particles in Mediterranean areas during dry periods, given that a non-vegetated CW in which metals are accumulating in sediment could become a source of metal contamination by wind dispersion. Some wetland plants can also be used for their roots' ability to form metal-rich plaques. These structures are composed mostly of iron hydroxides and other metals such as manganese that are mobilised from the reduced anoxic sediments and concentrated in the oxidized microenvironment around the roots (Weis and Weis, 2004). Some metals, such as Zn and Pb, seem to be more easily accumulated on root surfaces with plaque than those without plaque (Yang et al., 2010; Ye et al., 1998). Finally, plant species may be used to enhance specific microbial functional groups (Faulwetter et al., 2009).

Therefore, macrophytes may be selected for the removal of metals in CWs for many different traits. In order to determine the interesting traits of the macrophytes that are candidates for a CW project, preliminary studies remain the best option.

4.4. Preliminary studies remain essential for plant selection

Some authors have analysed metal contents in macrophytes from natural wetlands polluted with metals to determine their accumulation abilities (Del Ŕio et al., 2002). Nevertheless, a higher concentration of metals in the plants usually (Collins et al., 2005), but not always (Bragato et al., 2006; Mays and Edwards, 2001; Yeh et al., 2009), indicates a proportional increase in element levels in the water (Grisey et al., 2012; Soda et al., 2012; Yadav et al., 2012) and/or sediments (Guilizzoni, 1991). In other words, depending on the plant type and on the environmental conditions, the high concentrations of certain metals found in macrophytes may either reflect high external pollution levels, or the efficiency of the plant in concentrating these particular substances. To complicate the situation, different forms of the same metal can have different rates of uptake and different effects on plants (Weis and Weis, 2004). Hence, metal data must be extrapolated with great care from one species to another or even, within the same species, from one study to another (Guilizzoni, 1991). Thus, for one given plant species, as the efficiency of phytoextraction also depends on both the water to treat and the CW characteristics, we would recommend carrying out preliminary studies in mesocosms in order to determine the accumulating properties of autochthonous plants under the CW conditions. We suggest setting up mesocosms which contain identical substrate to the CW that is planned as well as the same depth, to achieve results that might be extrapolated. Stoltz and Greger (2002) have demonstrated that a simple hydroponic screening experiment without any substrate is not a suitable method to investigate uptake and accumulation properties of plant species. They have noted that accumulation patterns differed between the field and hydroponical conditions, suggesting that in the field, plant roots interact with soil particles, root-bacteria and/or mycorrhizal fungi which are not present in laboratory solution and might affect the metal uptake. Therefore, these preliminary laboratory studies might provide a basis for determining whether species are tolerant of the wastewater to be treated and of the CW conditions, which are the first things to verify (Maine et al., 2009). At the least, these studies should take into account the bioavailability of metals present in the effluent, and a chemical speciation of metallic load is thus strongly recommended to further assess the CW efficiency. Uptake of metal is strongly correlated to metal bioavailability (Barghava et al., 2012), and it is therefore important to determine the bioavailable/total metal ratio in the influent in order to better interpret the ability of a given species to treat a given metallic load. This speciation may also be carried out in the substrate (soil or sediment), where metal availability and mobility in the rhizosphere may be altered either by rhizospheric microbes and the root exudates (Chaney et al., 2007), or by siderophores produced by microorganisms that are capable of enhancing the availability of metal for uptake into roots (Devez et al., 2009; Neubauer et al., 2000).

Finally, it is necessary to determine which plant species may constitute a threat to the food web in order to adapt management strategies, as metal-tolerant plants grown in polluted medium that can take up and accumulate metals in their tissues may constitute a health hazard for humans and animals (Lotmani et al., 2011).

4.5. Plant distribution in CWs is also an important parameter

Once the wetland plant species have been selected for a given application, their distribution in the CW may be organized carefully. High biomass, fast growing and accumulating species such as cattail or common reed have attractive non-specific extraction capabilities but they tend to shade other species in CWs (EPA, 1994; Hadad et al., 2006; Maine et al., 2009; Yang et al., 2006). In CWs, competition may be more severe than in other plant communities because of the same or similar growth forms, similar individual size, and similar light demand (few are shade-tolerant) amongst the plants used in CWs (Liang et al., 2011). Thus, in order to maintain specific diversity within the CW, some recommendations should be made. For example, two separate cells positioned one after the other for metal treatment could be used: one with particular collector plant species (chosen in function of the effluent metal content) and another with the unspecific collector proliferating species. Moreover, the density of planting for each selected species is an important design parameter and should be engineered taking into account various parameters: the percentage of each metal in the wastewater, the exporting ability (mg/m^2/year) of each species for the different metals, the toxicity of the different metals for the local environment, and the objectives of purification for each metal. This last criterion is linked with the widest objectives of the CW project given that the desired level of purification will depend on the restoration goals. In particular the functionality of the natural aquatic ecosystem (water reservoir against flooding or for drinkable water production, biodiversity reserve, agricultural production or halieutic resources, tourism zone or cultivation, etc.) that has to be preserved in priority will condition the level of purification to achieve. Evaluating current ecological status of water receiving bodies and determining their vulnerability to metals is important in order to establish the level of pollution management in catchments and the objectives of restoration. In particular, several risk-based approaches have been developed to support decision making for programmes aiming to control diffuse metal pollution (Chon et al., 2012) and may be applied to Mediterranean catchments.

5. Handling and technical obstacles concerning the management of CWs

5.1. Essential management practices to favour metal retention in CWs

Not only plant selection and distribution in CWs are important because the management practices of plants (cutting/exporting or not) may also affect the retention capacity of the CW.

It is assumed that the removal capacity of the substrate mainly depends on its sorption capacity (conditioned by its specific surface, mineral and/or organic composition) and on the physical and chemical conditions in the basin. This latter mainly depends on the basin dimensions (shape, depth, surface) and on physico-chemical properties of wastewater (Sheoran and Sheoran, 2006). The management practices of plants can also affect the retention capacity of the substrate, given that metals are well adsorbed on organic matter. The frequency with which the substrate is renewed will also play a role as it enables resetting of the sorption capacity of the substrate.

On the other hand, metal-uptake capacity of the total plant biomass mainly depends on the biological traits of the selected plant species that are planted but also on management practices. In fact, the quantity of metals exported in plants is linked to the mean absorption efficiency of metal into plants and to the plant biomass production. The mean absorption efficiency depends on the biological performances of plants, which is expressed in their life cycles and life histories, and by the trophic situation in which plants grow (Guilizzoni, 1991). It also depends on the average bioavailable concentrations of metals in the rhizosphere including both the soluble part of metal in the pore water and the fraction of metals that has been remobilized from the substrate to the water due to the plants' occurrence (Triboit et al., 2009). The latter depends on many parameters such as plant species, pH, redox potential and plants and microorganisms interactions (Guilizzoni, 1991; Yang et al., 2005). The biomass production is affected by plant density and climate. Plant harvesting stimulates plant growth and uptake of contaminants. Hence, harvesting methods of plant biomass (cutting or digging out) and frequency are important management practices. Thullen et al. (2005) argue that vegetation management considerations need to be incorporated into the basic design of wastewater-treatment wetlands.

5.2. Management strategies: environmental but also socio-economic issues

CWs should not only be designed but also be operated with a view to maintaining the healthy plant community upon which so many biological, chemical, and physical treatment processes depend (Thullen et al., 2005). Management of plants is linked with the environmental and socio-economic contexts and has to be adapted to the types of plant species that are used. Management strategies should be determined as a first stage in order to adapt the CW design and construction.

Concerning the environmental context, attention must be paid to the environmental quality of the land that will have to be used in order to set up the artificial system. Uptake of metals by plants exhibits a plateau response at high loading rates and plant physiological factors are responsible for this plateau (Hamon et al., 1999). Hence, plants may be used as metal exporters only if the available land and the wastewater characteristics (i.e. total flow rate and metal concentrations) make it possible to maintain the loading rate within mean ranges. This is the case when the water to be treated has low metal concentrations and when the CW is situated in an area with available land. In contrast, in cases where either the total flow rate or the metal concentrations of the water is high, especially if there is little land surface area available for installing the CW, it may be preferable not to rely solely on metal uptake capacity by the plants, but also on high sorption capacity of the substrate. The best solution may then be to set up a deep and compact wetland using highly adsorbent substrate, perhaps planted with plant species that trap metals in the rhizosphere and do not transfer them in their aerial biomass (Yadav et al., 2012).

Concerning the socio-economic context, the amount of resources that can be allocated to the management of the CW must be considered when designing the system. The frequency of plant harvesting has to be high to ensure the metal export via plant biomasses is significant compared to those of substrates. Liu et al. (2007) claim that in densely-planted wetlands for wastewater treatment, plants will absorb and accumulate considerable amounts of metals that can be removed by frequent harvesting of the plants. A proper vegetation balance (i.e., a balance between minimum biomass for maintaining treatment function and excessive accumulation of plant wastes to manage) is needed for optimum treatment performance and the key to maintaining this proper balance is through vegetation management (Thullen et al., 2005). These actions have to be included within the overall treatment budget but they may become economically viable if the harvested biomass is value added-reused. Even if most of the time, plants enriched in metals are still considered as ultimate wastes and evacuated in landfills after burning, several paths of value-added reuse exist and some of them need further researches. If the plant accumulates relatively low metal concentrations, they may be used to produce biogas and sludge or organic fertilizer by composting (Hansson and Fredriksson, 2004). They may also be burnt for bioenergy production and derived ashes that usually have high nutrient concentrations may be spread in forest and agricultural soils as fertilizer (Bonanno et al., 2013). Ecotoxicity studies should be performed in order to define metal concentration thresholds in plants enabling such uses. Moreover, new opportunities are emerging for using metals accumulated in plants as catalysts in green chemistry (Losfeld et al., 2012). Only a few hyperaccumulator terrestrial plants have already been used for such purpose as it depends on the chemical forms of the metals stored into plant biomass and their possible prospect in green chemistry which is very specific to certain plant species and metals. Given the wide variety of reactions that are catalysed by ecological catalysts, and the ease of preparation of such catalysts, there are many opportunities for the development of catalytic processes (Escande et al., 2014) which lead us to think that such researches should also be undertaken in wetland plants accumulating high metal concentrations. A proportion of metals not exported via plant harvesting may be exported from the CW by the macrofauna and potentially constitute a source of contamination through the trophic web (Ciutat et al., 2005). During the life span of the CW, it is also expectable that physico-chemical conditions change in the CW, conducting to changes of metal speciation and potential release of metals trapped in substrate. Finally, metals that have not been exported by plant harvesting or macrofauna and that have stayed trapped in substrate will be exported with substrate when saturated. While bioleaching is a promising alternative for the recovery of the CW substrate contaminated with metals (Anderson et al., 1998), it still consists in a transfer of metals from a compartment (substrate) to another (water). Unless efficient strategies of metal recovery are developed, metals trapped in substrate will still end their life as dangerous waste in landfills.

For cocktails of metal pollution, multiple harvests may be beneficial for element removal from wastewater. In all cases, depending on the module and its plant content, the management will vary in terms of type and time of harvest. In the module with accumulative species, aerial parts that contain metals have to be harvested and exported. In the module with excluder species, plants have to be collected with the aim of exporting their root system. Additional questions have to be elucidated concerning the best seasons for plant harvesting in both cases.

According to Bragato et al. (2006), in order to maximise metal removal, plant harvesting should be undertaken during the period of maximum content in plant tissues, that is in late autumn after senescence. Windham et al. (2003) demonstrated that metal concentrations in leaves of both *S. alterniflora* and *P. australis* are higher in senescent leaves than in young ones for the same individuals. Other authors claim that rates of metal absorption are lower in senescent than in actively growing plants, given that at the beginning of the growth period, macrophytes show a rapid uptake of metals (Guilizzoni, 1991; Mortimer, 1985), and suggest harvesting just after the growing season. Hence, one factor that does seem fairly constant is that individual leaves acquire greater concentrations of metals over their life span (Weis and Weis, 2004). Nevertheless, variations occur depending on the metal that is considered. Vymazal et al. (2010) found that 13 elements i.e. As, Ba, Co, Cr, Cu, Fe, Ga, Hg, Mn, Ni, Pb, Sb and U are transported to the aboveground biomass late in the growing season. On the other hand, 10 other elements (Al, Cd, Li, Mo, Rb, Se, Sn, Sr, Tl and Zn) are accumulated at the early growth stages. This may be the result of a modified chemistry of the rhizosphere during plant growth periods and consequently change in metal availability at the interface of plant root-sediment (Quan et al., 2007). Therefore it seems that harvesting should globally occur between the end of the growing season and the beginning of senescence, depending on the extent of metals to export.

The seasonal question has to be considered in a different way for the harvesting of whole plants. Further studies are required to evaluate the best period for digging out, with the aim of both exporting the greater amount of metals and of allowing the plant the ability to recover. Cheng et al. (2002) suggested an ingenious way of exporting pollutants for plants (such as *C. alternifolius*) that can store metals underground in their lateral roots forming a continuous layer at the top of the basin. This method involves removing a few centimetres of surface layer at the end of a treatment period. As some authors have noted that metal concentrations in substrate and in belowground biomass tend to decrease with distance from the inlet (Lesage et al., 2007), selective and frequent uprooting and replanting could be performed in a restricted perimeter around the inlet of the modules. However, lack of vegetation cover during the dry period under Mediterranean climate may cause wind transfer of metal pollution from the substrate to the surroundings of the CWs (Triboit et al., 2009). This has also to be taken into account for the choice of harvesting periods in such climatic contexts as well as the periods presenting risks of colonisation of the CW by undesirable plant species.

5.3. Pathway to better suited designs to facilitate CW management

Concerning the phytoremediation of metal-contaminated soils, optimum plant properties for phytoextraction purposes include a high capacity for uptake, transport, and sequestration of metals in aboveground parts (which can then be harvested) (Rabier et al., 2007) because belowground parts of the terrestrial plants can only be exported with difficulty. In the case of the purification of metal contaminated waters in CWs, plants that accumulate metals in belowground parts may also be used given that for many macrophytes species, the underground parts can be easily dragged up from substrate (Liu et al., 2007).

Nevertheless, without suitable CW design, it would require the provision of considerable means (human and material) to dig out plants each year or several times a year. Chen et al. (2012) have tested and approved a system in which the aquatic vegetation is no longer rooted in a solid matrix; but is growing as a plant root mat, where roots have direct contact with the water. Such a mat of densely interwoven roots enables the plants to anchor mechanically with their roots as in soil and give the aboveground plant parts stability against tilting. Chen et al. (2012) confirm that plant root mats are a variant of CWs without a soil matrix that could be a cost-effective solution for the treatment of distinctively contaminated waters. In contrast, Tanner and Headley (2011) have tested floating CWs to treat stormwaters polluted with metals. They found that they were able to remove dissolved and particulate-bound metals. The fact that plants are grown on a buoyant mat makes them particularly suitable for event-driven stormwater applications where water depths and flow rates can vary significantly over time. Yang et al. (2008) have also developed a floating-raft hydroponic system that has proved to be highly efficient. We assume that these methods may also be particularly suitable for industrialised catchment wastewaters polluted with metals and may prove to be well-suited for use in Mediterranean areas. They represent a means of improving the treatment performance of conventional systems by including the beneficial aspects of emergent macrophytes without being constrained by the requirement of shallow water depth (Headley and Tanner, 2012).

5.4. Biomonitoring tools for low cost CW management

Any ecosystem, even an artificially constructed one, has limits to its ability to cope with disturbance. The performance of CW systems may change over time as a consequence of changes in substrate and accumulation of pollutants in wetlands. Thus, CWs must be monitored periodically to detect eventual evidence of stress so that remedial action, if necessary, can be taken (EPA, 1994). In the context of metal pollution from industrialised catchments or zones, the classical physical and chemical methodologies for monitoring seem inappropriate for two main reasons: the high diversity of contaminants that may be encountered and their variable concentrations over time (that could sometimes be below analytical detection levels) and the high cumulative cost of these analyses. Moreover, these methods do not take into account the state of the biotic environment receiving treated waters and in particular, the toxicity of the treated water for downriver ecosystems. For instance, chemical analyses do not highlight toxicity for biota that may be caused by synergy between pollutants. Therefore, certain biomonitoring tools need to be developed and used in combination with physical and chemical analyses (Zhou et al., 2008). Given that different macrophytes have different sensitivity to pollutants, plants could be used not only for their treatment abilities, but also for their role as sentinels. For instance, Lotmani et al. (2011) have found that variations in germination capacities were observed among populations under metal stresses and growth parameters were differently affected by metal treatments. Therefore, the monitoring of growth and germination parameters of macrophytes and their comparison between the upstream and downstream parts of CWs could provide information regarding the improvement of water quality for biota. Biomarkers of plant performance like H_2O_2 content (Shelef et al., 2013), photochemical efficiency, CO_2 assimilation rate, and cell membrane stability (Shelef et al., 2011) have been proven to correlate with wastewater-quality improvement along the CW and may be used to monitor the CW performance. Non-destructive monitoring are of particular interest given that they are cheaper and easy to use in comparison with destructive monitoring of molecular stress indicators such as reactive oxygen species content and do not alter the phytoremediation performance of plants. For that purpose, easily transportable equipments are being developed for a use directly on field e.g. the Multiplex® (Force A, Orsay) field equipment that uses fluorescence technology with multiple excitations to measure phytometabolites content such as chlorophylls and flavonoids (including anthocyans), both correlated to plant state of stress (Laffont-Schwob et al., 2011).

6. Political and organisational obstacles and potential solutions to study

In recent years, there has been growing concern regarding the consequences of wetland degradation, leading to the adoption of several conventions, directives, and an associated range of natural conservation and restoration actions for their protection (e.g. Natura 2000 sites, Water Framework Directive, Ramsar Convention in European Union, Schleupner and Schneider, 2013). Recently, the European Directive 2010/75/EU on industrial emissions was definitively adopted by the European Council, to reinforce prevention and control (European Union, 2010). Our review suggests that the CW approach could be one of the emerging techniques to be developed and tested by Mediterranean member states to reduce industrial pollution. It would require some preliminary studies conducted by researchers and engineers together to design and test treatment modules for different families of pollutants, including metals.

In Mediterranean environments, in order to reduce the amount of water that would be treated in the CW, a solution could be to by-pass the treatment in CWs for the waters during decreasing floods. It has been shown that applying Best Management Practices (BMPs) treatment early in the season could remove several times more pollutant mass than randomly timed or uniformly applied BMPs under Mediterranean climate (Lee et al., 2004). Another solution could be to implement a separate sewer system for the collection of industrial treated wastewaters. This would involve a new way of operating in industrial zones, as recommended by supporters of industrial ecology principles (Boons and Baas, 1997). The CW could thus be seen as an additional pooling treatment of industrial wastewaters produced within the same catchment or zone. The benefits of this additional pooling treatment are in phase with sustainable development principles. Considering the footprint question, a single CW would entail far less material and energy than several physical and chemical or biological tertiary treatment systems allocated within the industrial facilities of the catchment. Moreover, considering that land is an essential and non-renewable resource, CWs are a suitable solution as they can perform a triple function: water treatment, biodiversity conservation and natural leisure settings (Yeh et al., 2009). However, to consider the cumulative impact of pollutants at catchment scale, it is necessary to be aware of the total pollutant concentrations discharged by industrial sites into water environments. We recommend pooling industrial discharges, not only in order to treat them within a CW, but also in order to be able to determine mutual objectives regarding water quality at the scale of the industrial catchment or zone. This way of functioning would also enable the authorities to determine the degree of flexibility they dispose of for the establishment of a new industrial facility in the catchment. To facilitate monitoring and to complement chemical analyses, the use of plant bioindicators in streams that receive industrial discharges and in aquatic receiving environments could be an appropriate management measure, in accordance with the Water Framework Directive recommendations (Jones et al., 2010). Lastly, regulatory constraints and controls downstream of the CW could be introduced in accordance with local environmental management objectives. Finally from the social point of view, it could generate a feeling of shared responsibility for environmental protection among industrial managers (see the Healthy Catchments Strategy, 2009–2012 of the Sydney Catchment Authority) and stakeholders in the catchment (Orr et al., 2007). It could even be required that a kind of industrial union in charge of making sure water quality is respected be constituted at the local scale. Thus, pressures for improvement of wastewater treatment systems would not only be applied by government agencies, but also by industrial managers within the same territory. That could be a new step towards achievement of the "polluter-pays principle". It could increase the number of industries equipped with a tertiary treatment system and facilitate regulatory controls. In summary, it could improve the involvement of industry in achieving an environmentally responsible way of functioning.

The use of market mechanisms to encourage end-users and industries to implement BMPs at site and local level is an approach that has the potential for successful application and it is being encouraged in many countries (Barbosa et al., 2012). It is worth noting that educational programmes and pilot scale applications are also fundamental to facilitate social acceptance of techniques such as CWs to treat stormwaters containing metals from industrialised catchments. For the most part, as Bulc and Slak (2009) reported, the utility of ecoremediation strategies is in developing and applying an innovative model for a deeper understanding of environmental and social sustainability. The aims are to reduce the risk of natural disasters and threats to human health, to develop appropriate environmental policy strategies and regulatory frameworks, to ensure community participation, to protect biodiversity, ecosystems, landscapes, and local cultural features, and to create educational opportunities to ensure an environmentally stable society in the future.

7. Conclusion

It is necessary to provide an integrative approach for the prevention and control of industrial emissions into water (European Union, 2010), in particular when conveying metals. Constructed Wetlands seem to be a promising ecoremediation technology for the reduction of metallic diffuse pollution and the restoration of Mediterranean water bodies located downstream of industrial catchments (Bulc and Slak, 2009). Nevertheless, further research must be performed to improve CW design, monitoring and management methodologies before extending their use to Mediterranean environments. In this review we have highlighted certain obstacles and proposed pathways for future research concerning hybrid CW design, macrophyte species selection and management and catchment organisation that could be further explored.

We could see that each module of the CW corresponds to a type of depurative ecosystem in which plants, microorganisms and their habitat interact via a network of matter and energy fluxes. In each module, depending on the physical and biological design parameters that have been chosen, some of the contaminants transported by water will change of form, behaviour and toxicity. By playing on the design parameters (shape, depth, slope, etc.) of each module, and in their mineral (substrate) and biological (plants, microorganisms) composition, the energy and matter exchanges within ecosystems may be focused on a specific depurative objective, associated with a group of pollutants. For this purpose, further research must be undertaken in order to facilitate and optimise CW design, and in particular more consideration must be given to plant species selection.

Plant species differ in their sensitivity to pollution (type of pollutants and concentrations) and in their reactions (e.g. accumulative or excluder species for metals) and may be directly helpful in removing metals from water, in particular via metal uptake. Their selection and their management (i.e. cutting and/or harvest or not, frequency and time of harvest, replanting or not) may notably affect removal of metals. We may observe that the selection and management of macrophyte-diversity in CWs for treatment of mixtures of metals also condition some physical design parameters of modules. Thus, before proceeding to the physical design of CWs, it must be decided which plant species are required. The development of practical design and species selection methodologies seems to be an area requiring additional research. These improvements in CW design must be accompanied by monitoring and policy measures to enhance the integration of CWs in catchments.

Until now, purification of the metal pollution from industrialised catchments has had to be undertaken by natural wetlands, which may have deleterious consequences in Mediterranean environments for conservation of biodiversity (Duchet et al., 2010; Roche et al., 2009). The introduction of CWs between industrialised catchments or zones and sensitive Mediterranean aquatic bodies is a promising strategy. Our review suggests that it is worthwhile pursuing research aimed at developing methodological, technical and organisational tools enabling the relocation of the depurative function from natural wetlands to artificial ones located upstream, thereby reducing the impact of metal pollution on natural aquatic environments.

[1]From a chemical point of view, the term heavy metal is strictly ascribed to transition metals with an atomic mass over 20 and specific gravity above 5. In biology, "heavy" refers to a series of metals and also metalloids that can be toxic to both plants and animals, even at very low concentrations (Rascio and Navari-Izzo, 2011). In this review, the term "metals" refers to these potentially phytotoxic elements.

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