Organic and metallic pollutants in water treatment and natural wetlands: a review

K. Haarstad, H. J. Bavor and T. Mæhlum

ABSTRACT

A literature review shows that more than 500 compounds occur in wetlands, and also that wetlands are suitable for removing these compounds. There are, however, obvious pitfalls for treatment wetlands, the most important being the maintenance of the hydraulic capacity and the detention time. Treatment wetlands should have an adapted design to target specific compounds. Aquatic plants and soils are suitable for wastewater treatment with a high capacity of removing nutrients and other substances through uptake, sorption and microbiological degradation. The heavy metals Cd, Cu, Fe, Ni and Pb were found to exceed limit values. The studies revealed high values of phenol and SO_4. No samples showed concentrations in sediments exceeding limit values, but fish samples showed concentrations of Hg exceeding the limit for fish sold in the European Union (EU). The main route of metal uptake in aquatic plants was through the roots in emergent and surface floating plants, whereas in submerged plants roots and leaves take part in removing heavy metals and nutrients. Submerged rooted plants have metal uptake potential from water as well as sediments, whereas rootless plants extracted metals rapidly only from water. Caution is needed about the use of SSF CWs (subsurface flow constructed wetlands) for the treatment of metal-contaminated industrial wastewater as metals are shifted to another environmental compartment, and stable redox conditions are required to ensure long-term efficiency. Mercury is one of the most toxic heavy metals and wetlands have been shown to be a source of methylmercury. Methyl Hg concentrations are typically approximately 15% of Hgt (total mercury). In wetlands polycyclic aromatic hydrocarbons (PAH), bisphenol A, BTEX, hydrocarbons including diesel range organics, glycol, dichlorodiphenyltrichloroethane (DDT), polychlorinated biphenyls (PCB), cyanide, benzene, chlorophenols and formaldehyde were found to exceed limit values. In sediments only PAH and PCB were found exceeding limit values. The pesticides found above limit values were atrazine, simazine, terbutylazine, metolachlor, mecoprop, endosulfan, chlorfenvinphos and diuron. There are few water quality limit values of these compounds, except for some well-known endocrine disrupters such as nonylphenol, phtalates, etc.

Key words | metallic, organic, pesticide, pharmaceutical, pollutants, review, wetlands

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INTRODUCTION

Increasing demands on water resources and the environment make reclamation and reuse of municipal and other wastewater and runoff waters an important component of the water budget. Constructed wetlands are used successfully to treat domestic and industrial wastewater worldwide for secondary and tertiary treatment of domestic wastewater. Effluent standards can be met also under temperate climatic conditions. Treatment wetlands can be an integral part of the water-reuse cycle providing both water-quality enhancement, water volume control and habitat functions. When used for habitat, the bioaccumulation potential of contaminants in the wastewater is a critical consideration. Wetlands include marshes, swamps, bogs and wet meadows, and are seemingly 'magical' areas (Kadlec & Night 1996) with unique properties due to their frequent wetness, alternating chemical, physical and biological properties of soil, water

doi: 10.2166/wst.2011.831

and biota. Historically, wetlands have been used to treat agricultural, urban and industrial wastewaters receiving and transforming pollutants from preferably pre-treated waters into new biomass, soil and gases, with very low inputs usually restricted to some earthwork, piping, concrete structures and pumping. Wetlands are formed in lowlands or in flat parts of uplands, where ample water is provided either by precipitation, surface or groundwater flows.

Wetlands are used in a broad variety of ways to prevent pollution, in order to meet targets set by a regulator, of freshwater, drinking water, groundwater, polluted soils and biodiversity quality, or for polishing treated or untreated wastewaters. Although the use of treatment wetlands is well established for wastewater categories such as municipal waste, stormwater, and acid mine drainage water, their use in treating a variety of industrial and agricultural wastewaters is less well developed. Chemical oxygen demand (COD), biochemical oxygen demand (BOD), trace organics, metals, toxicity, total suspended solids, nitrogen, and phosphorus can reliably be removed from wastewater by treatment wetlands, generally by horizontal subsurface flow wetlands focusing on filtering processes removing, e.g. phosphorous (Figure 1), and vertical flow wetlands for removing nitrogen. Pollutant removal is highly dependent on hydraulic loading and influent concentration and to a lesser extent on internal plant communities, water depth, and hydraulic efficiency. According to a review by Bavor & Mitchell (1994), Bavor *et al.* (1995) and Haberl *et al.* (2003) wetlands have a number of benefits:

- Less expensive to build than other treatment options.
- Simple construction and operation.
- Utilizing natural processes & tolerate flow variation.
- Low operation and maintenance expenses.
- Periodic, rather than continuous, on-site operation and maintenance.
- Able to treat different types of wastewaters.
- Characterized by a high process stability (buffering capacity) and low excess sludge production.
- Facilitate water reuse and recycling.
- Provide habitat for many wetland organisms.
- Fit harmoniously into the landscape.
- Provide wildlife habitat and aesthetic enhancement, open spaces.
- Publicly favourable and environmentally sensitive approach.
- Can treat water polluted with organic compounds.

Some common problems and challenges are:

- Details of processes are typically not available.
- Research is needed to identify microbes, define optimal plant species and quantify rates of individual processes in the field.
- Removal rates, as well as optimal plant species, are substance specific, and also typically not available. Laboratory studies are best suited to gain this data.
- Genetically modified plants should be considered to enhance the treatment performance of constructed wetlands for specific compounds.
- Performance data are needed for microbial activity and contribution of the plants to the overall removal process.
- If a constructed wetland provides different environmental conditions and uses different plant species the treatment efficiency can be improved.

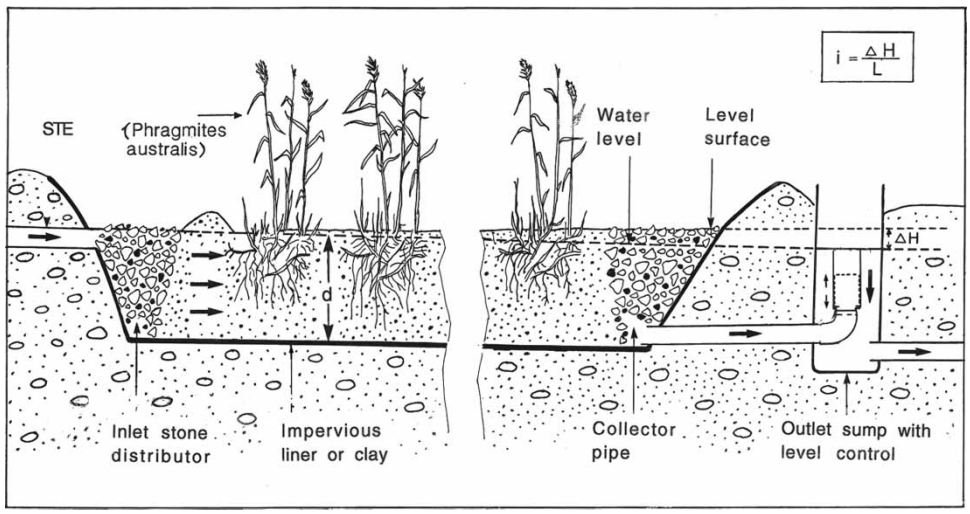

Figure 1 | Layout of typical subsurface flow wetland (SSFW) including plants, filter materials and flow arrangement.

- Removal rates depend on seasons and the age of the wetland system.

Here the occurrence of pollutants in wetlands is presented, focusing on organic pollutants but not excluding heavy metals. The latter can be a problem in wetlands also by generating more toxic forms (methyl Hg, labile Cu).

METHODS

A search in Scopus in 2009 using 'wetlands, organic, pollutants' produced 474 references, of which 123 are relevant for organic or organo-metallic pollutants and natural or constructed wetlands. Searches in ISI-knowledge and Science direct using the same keywords produced 407 references in 2011. Relevant books and proceedings have also been included. The concentrations have been compared to a number of limit values in water, sediments and biota, see Table 1.

RESULTS AND DISCUSSION

The results in this paper are from the listed references. The references can be found both in the text and in the tables.

Table 1 | Limit values in drinking water (EU 1998, Department of the Environment 2009; World Health Organization 2003; Norwegian Health Authorities 2005; Australian Government 2004); polluted water (SFT 2003; Haberl *et al*. 2003), and natural water (US EPA 2009; SFT 1997; ANZECC 1992). In µg/L

	Drinking water	Polluted water	Natural water
Cd	2 to 5	0.04 to 0.2	0.03 to 0.66
Pb	10	1.9 to 65	0.2 to 7
Hg	0.5 to 1	0.01 to 1.4	0.002 to 0.05
Ni	20	4 to 470	0.5 to 88
Cu	100 to 2,000	2.3 to 130	0.6 to 26
CN	5.2 to 10	n.a.	5.2 to 100
Pesticides	0.1/0.5	n.a.	n.a.
HC	10	35	n.a.
PAH 16	0.01 to 0.7	2 to 3.5	n.a.
PCB 7	n.a.	1 to 5	n.a.
Bisphenol A	n.a.	0.02	n.a.
Phenols	n.a.	0.01	n.a.
BTEX	n.a.	80	n.a.
Formaldehyde	n.a.	17	0.01

n.a.: not applicable.

Heavy metals

The heavy metals Cd, Cu, Fe, Ni and Pb were found to exceed limit values in water (Table 2). Also these studies revealed high values of phenol and SO_4. No samples showed concentrations in sediments exceeding limit values, but fish samples showed concentrations of Hg exceeding the limit for fish sold in the EU. For removal, the harvesting of plants should be organized according to the specified metals and plants in use.

Heavy metals are the most common environmental pollutants, from mining, industry and ordinary wastewater. Cu, Cr and Zn are the most frequently detected heavy metals in modern untreated wastewater in Norway (Blytt & Storhaug 2008), with Cu, Hg and Zn representing the most environmentally challenging of the metals. Mechanical treatment shows much lower removal of heavy metals than chemical/biological treatment. In wetland soils, trace metals can be released due to reducing conditions, possibly by (i) metal release by reductive dissolution of Fe-oxyhydroxides, or (ii) metal release by OM release (Chandra *et al*. 2008). A wetland reduced Cu, Zn and Pb by 60 to 80% in a river water system, increasing over a 5-year period the Cu in the wetland sediments from ca. 4 to 800 mg/kg, higher in the fine sediments than in the organic sediments, although the remobilization was lower in the organic layers (Knox *et al*. 2006). Constructed wetlands working in subsurface flow mode showed higher Cr and Zn removal rates in comparison to surface flow units. Planted systems removed more Cr and Zn than unplanted systems and the addition of an organic carbon source increased the Cr removal rate, but there was not any effect over Zn removal rates (Paredes *et al*. 2007). Surface-based systems showed the lowest Cr and Zn removal rates; however, the addition of an organic carbon source increased the removal rates. Thus, for wastewaters containing Cr and Zn, plant-based systems with subsurface flow are the best option for metal removal, even if the wastewater does not have organic carbon content. A study showed that application of sediment with 100 mg/kg Cd or Zn had little or no beneficial effect on wetland denitrification, while 500–1000 mg/kg decreased it, the toxicity being in the order Cd > Zn > Cu, but it was also dependent on the NH_4 concentration (Guy Riefler *et al*. 2008). Sorption processes were responsible for metal removal by straw filters, chosen based on straw availability and low-cost. However, a disadvantage of using an un-decomposed OC-source such as straw is the mobilizing effect on heavy metals during start-up and the fact that sorption is relatively quickly depleted (<1 year), thus

Table 2 | Heavy metals measured in wetlands

Pollutants Heavy metals (μg/L) *italic* = wetland effluent, **bold** = >limit[a]	What/where?	Concentration	Comment/removal	Reference
Acid mine drainage	Acidity due to sulphide oxidation & metal hydrolysis	Mn: 1–8	Problems: clogging in alkaline cond., channel flow, limit alkalinity production, arid, macrophytes contribute <0.1% in Fe red	Barrie Johnson & Hallberg (2002)
Acid mine drainage	Review 35 passive treatment systems, S Korea	mg/L: Fe 0–**551**, SO_4 2–**4038**	Successive alk. prod. syst. (SAPS). Rem. %: Fe 11–100, SO_4: 0–78	Ji *et al.* (2008)
Acid mine drainage	Review of removal mechanisms. Carbonate ok for Pb, Ni. Sulphide if high in sulphate + TOC	Cu, Ni, Zn, Mn, Cd co-precipitate with Fe, high pH. As, Sb, Se under low pH. Co, Fe, Ni, Zn with Mn. As and Zn on plant roots	Filtering: Cd 75–99.7%, Pb 26, Ag 76, Zn 67 Adsorption: >50%. Se needs to be reduced. Co-precipitation with Fe^{3+}. Al 33%. HCO_3 microbial, limestone good for AMD	Sheoran & Sheoran (2006)
Heavy metals, colour, COD, BOD, phenol	Distillery effluent/Free floating plants in wetland/India	Colour: 180 000; BOD: 28 000; COD: 52 400; SO_4: 3100; N: 668; phenol: 510; Cd: 2.3; Cr: 0.44; Cu: 1; Fe: 84; Ni: 1.2; Pb: 4.4; Zn: 4.6	Increased removal due to bacteria and plants in %: Cd (34–62), Cr (36–58), Cu (33–54), Fe (33–52), Mn (36–83), Ni (36–59), Pb (33–60) and Zn (32–54)	Chandra *et al.* (2008)
Heavy metals	FWS flow wetlands, India, microbial pre-treatment of methanated distillery effluent	*Increased removal 20–80%: Cd, Cr, Cu, Fe, Mn, Zn, Ni, Pb*	Max. 30% effluent, also BOD, COD, phenol, N reduced	Chaturvedi *et al.* (2006)
Heavy metals	Review	Pb, Cr, Cu, Cd, and Zn	*Eichornia* efficient in taking up metals	Dhote & Dixit (2008)
Heavy metals	Acid mine /anaerobic vertical flow wetlands/USA	5 kg/d Fe 1.7 kg/d Al	Alkalinity from limestone and sulphate reduction	Guy Riefler *et al.* (2008)
Heavy metals	Surface wetland of industrial runoff, Calif. USA. Flocs & organic sediments	1–30 *0.4–342*	Cu, Zn, Pb red. 60–80%. Ret. Low in floc layer, high in org. layer. Low ret. for Mn	Knox *et al.* (2006)
Heavy metals	FWS, landfill leachate	Cd: 0–0.4; Cu: 3–10.7; Ni: 6.2–6.4; Zn: 17.6–56.7	Removal: 68 to 83%	Persson *et al.* (2007)
Heavy metals	Impact on denitrification (dn) in FWS from metals in sediment	100 mg/kg did not decrease dn, while 1,000 mg/kg did	Most toxic: Cd > Zn > Cu	Sakadevan *et al.* (1999)
Heavy metals	Microbial processes	Biosorption, sulphate reduction, redox, methylation (Hg, Pb, Zn, As), plant-microbes	Key role in metal mobility, toxicity and bioavailability	Kosolapov *et al.* (2004)
Heavy metals in horizontal wetlands	3 experimental full scale	In: 0.1 to 5658 *Out: 0.08 to 1072*	>90%: Al; 50–78%: Zn, U, Sb, Cu, Pb, Mo, Cr, Ba, Fe, Ga; 25 to 50%: Cd, Sn, Hg, Ag, Se, Ni; Low: V, Li, B, Co, Sn. Negative: Mn, As	Krøpfelova *et al.* (2009)
Heavy metal removal	Subsurface wetland, lab	Co, Ni, Zn: 1 mg/L; Cu: 0.5; SO_4: **2,000**	Be careful ensuring stable redox conditions in CW	Lesage *et al.* (2007)
Heavy metals	River, coastal wetland, France. Uptake in bile in eels	0.02–1,166 μg/L	Highest (Al, Hg, Cu, Ti, Zn, Mn, Ni, Mg Fe) were found in livers	Oliveira Ribeiro *et al.* (2005)

(*continued*)

Table 2 | continued

Pollutants Heavy metals (μg/L) italic = wetland effluent, bold = >limit[a]	What/where?	Concentration	Comment/removal	Reference
Heavy metals	Cu in wetland sediments	Low strength: 0.3–1 ppm High strength: 35–50 ppm	Low strength: organic bound. High strength: sulphide bound	Sobolewski (1996)
Heavy metals in grass	SSF wetland, single or multiple harvest	Single harvest best: As, Ba, Co, Cr, Cu, Fe, Ga, Hg, Mn, Ni, Pb, Sb, U	Multiple harvest best: Al, Cd, Li, Mo, Rb, Se, Sn, Sr, Tl, Zn	Vymazal *et al.* (2010)
Heavy metals in *Phragmites*	SSF wetland, stems leaves flowers roots	Above ground conc. (mg/kg): 0.01 to 185, below ground: 0.05 to 4,228	Concentrations: Roots > rhizomes > leaves > stems	Vymazal *et al.* (2009)
Hg + methyl Hg in water, sed. And frogs	9 ponds in natural wetlands, USA	Water (ng/L): Hg: 1.3–8.4/ MeHg: 0.08–1.05, Sed: (mg/kg): 0.02–0.19/0.0009–0.002, Frog: 0.03–0.11/0.003–0.03	Tot. Hg and Me Hg correlated with water DOC. Beavers might be a good indicator of Hg pollution	Bank *et al.* (2007)
Hg, methyl Hg	831 lakes in US incl. 277 for MeHg	*ng/L 0–19.5 HgT* *0.20–0.59 MeHg (mean)*	Me Hg 15% of total, high in wetlands. Long retention time high Hg	Dennis *et al.* (2005)
Hg	Sequestration in forest and peat, USA Deposition/yr: 0.04 mg/m^2	Forest: 0.1 mg/m^2 (from atm.) Forest floor: 1 Soil: 10 Peat: 20	Hg affinity to org. with reduced S 1 to 1.5% MeHg	Grigal (2003)
Hg in streams	47 stations with & without wetlands, fire, US	Median 1.2 ng/L, max 5.1	No difference on burning, difference with wetlands and river DOM	Peckenham *et al.* (2007)
Methyl Hg	Naturally produced methyl Hg/ promoted by sulphate red. Bacteria?	*0.00049*	Highest in freshwater wetland/ correlate with humic acid portion of DOC	Hall *et al.* (2008)
Methyl Hg	Natural wetland/USA	*0.00027* MeHg	MeHg from 2–6% through wetland	Selvendiran *et al.* (2008)
Methyl Hg	Experimental wetlands, 16 m^2, USA	6–71 ng/L *5–40*	Temp. and SO_4 correlate to methyl Hg output	Sexauer Gustin *et al.* (2006)
Methyl Hg	Occurrence in 25 natural wetlands sed., Canada	300–900 ng/kg	Highly dependent on geology, not wetland type. Biological reasons	Siciliano *et al.* (2003)
Wood preservatives	Lab./USA	*7–***70** labile Cu (CuOH) +	Water from wetland high C/no removal studied	Dubey *et al.* (2007)
Methyl Hg	Natural wetland and lake, USA	*0.2–0.8 ng/L*	Wetland source of half of methyl Hg in lake. Other half internally produced from SBR	Watras *et al.* (2005)
Methyl Hg	Soil, water and fish, USA	Total Hg (ng/g): Fish 182–**942**, Sed. Prehist. 50–150, modern 100–364. Water: 0.3–3.3	The methyl Hg exceeds limit values	Wiener *et al.* (2006)
Trace metals	Release from wetland sediment	pH more important than Eh because of org. matter Cu, Cr, Co, Ni, Pb, U, Th, REE	4 process groups for metal release indicated	Grybos *et al.* (2007)
U	Algal removal of U(VI) from water, Canada		Algae can live under acid and alkaline cond. And remove U	Kalin *et al.* (2004)
Zn, Cr(VI)	Small-scale model wetlands, Colombia	1.5 mg/L	Flow, plants, TOC. More Cr rem. SSF with plants and with org. C. Also for Zn except for TOC	Paredes *et al.* (2007)

[a]limit values, see Methods and Table 1. All concentrations in μg/L unless otherwise stated. alk. = alkalinity, prod. = production, org. = organic, ret. = retention, rem. = removal.

making soluble OC-sources more promising. Once anaerobic conditions are created and sulphate reduction is taking place, precipitation of metals with sulphides provides an additional removal pathway and higher removal efficiencies. Precipitation of metals with sulphides will become the dominant long-term metal removal process. The addition of glucose to unplanted gravel microcosms enhanced sulphate reduction and metal removal. *Phragmites australis* negatively affected the precipitation of metals with sulphides in the root dominated top-layer of all microcosms, evidenced by higher metal levels, less sulphate reduction and higher redox conditions. Caution is needed about the use of SSF CWs for the treatment of metal-contaminated industrial wastewater as metals are shifted to another environmental compartment and moreover stable redox conditions are required to ensure long-term efficiency (Barrie Johnson & Hallberg 2002).

High mobility of Pb was found at a rifle range, due to heavy loading (Deiss *et al.* 2004). The background Pb accounted for the dominant fraction of the Pb leached from the columns.

Mercury is one of the most toxic heavy metals and since wetlands have been shown to be a source of methylmercury (MeHg) this metal is of special interest. The methylation process may occur under anaerobic (e.g. *Clostridium*) or aerobic (e.g. *Neurospora*, *Pseudomonas*) conditions (Mason 1991). Several non-biological transformations of Hg may also occur, but the methylation is biologic. Methyl Hg is exceptionally toxic to many animals. The concentration of MeHg in predator fish can be more than a million times the concentration in the water (Wiener *et al.* 2006). Whenever the wetlands have sulphate-reducing properties there is a potential of methyl Hg production, although this can be influenced by both biotic and abiotic factors. This is a topic for further studies. Often the highest total Hg and CH_3Hg+ concentrations are located far from obvious point sources correlated to topographically flat (and thus wet) areas that we relate to wetland abundances. Aquatic total Hg and MeHg concentrations are generally well correlated with total organic carbon (TOC) and with each other. MeHg concentrations are typically approximately 15% of Hgt. MeHg in wetland sediments has been shown to vary with lithologies, while other parameters such as pH, acid-volatile sulphides, sulphate reduction and anions were not important (Dennis *et al.* 2005).

Several lines of evidence suggest that wetlands may be a major source of methylmercury (MeHg) to receiving waters, perhaps explaining the strong correlation between concentrations of waterborne MeHg and dissolved organic carbon (DOC) (Bank *et al.* 2007). Wetlands also export MeHg in high concentrations (0.2–0.8 ng/L).

Hg in natural waters is complex and transported by organic carbon. Data suggest that the quality (i.e., composition) of the organic carbon varies in its ability to complex and transport Hg. This means that the Hg-DOC relationship should vary by source of DOC (Hall *et al.* 2008). Indeed, it is a surprising observation that on a per DOC-unit basis, headwater forest watersheds have the capacity to transport more Hg than wetlands. A study showed that Hg increased with decreasing wetland elevation, a cumulative impact of wetlands and riparian zones. The highest concentrations of Hg occurred during high spring base flow. The construction of large-scale wetlands could potentially improve river water quality through nutrient, sediment and total Hg removal. Since most of the Hg is bound to fine clay-sized particulate matter, the hydraulic retention time and flow rates associated with a wetland system could affect the efficiency of Hg removal. Phosphorus and N removal would also be influenced by the hydrologic conditions and degree of vegetation in the wetlands. One important management consideration is that sediment-bound Hg deposited to the wetland could be remobilized and may enter the river if a flood event occurred. As those mesocosms with clean water sediment were a greater source of MeHg than those with Hg polluted sediment, water chemistry is thought to be an important factor influencing MeHg production, from higher SO_4 and TOC concentrations, higher temperature, and lower pH and total suspended solids (TSS) concentrations. These factors combined would all promote methylation of Hg in sediments interacting with this water relative to MW, suggesting that use of CW for a wetland system could exacerbate production of MeHg.

It is widely recognized that wetlands, especially those rich in organic matter and receiving appreciable atmospheric mercury (Hg) inputs, are important sites of methylmercury (MeHg) production. Measurements show that MeHg concentrations were greatest in surface water of freshwater wetlands and lowest in the non-vegetated regions of a studied lake and river. Concentrations of total Hg and MeHg in filtered surface water were positively correlated with the highly reactive, aromatic (hydrophobic organic acid) fraction of DOC. These results suggest that DOC plays an important role in promoting the mobility, transport and bioavailability of inorganic Hg in these environments. Further, elevated pore water concentrations in marine and brackish wetlands suggest coastal wetlands are key sites for MeHg production and may be a principal source of MeHg to food webs.

Concentrations of methylmercury in a game fish study substantially exceed criteria for the protection of human health. Geologic sources of mercury were small, based on analyses of underlying bedrock and C-horizon soils, and nearly all mercury in the O- and A-horizon soils was derived from atmospheric deposition. A sediment core from five lakes showed that most of the mercury accumulated in lake sediments during the 1900s was from anthropogenic sources.

Wetlands have attracted much attention from the mining industry treating a range of industrial wastewater, in particular as passive acid mine drainage (AMD) systems. The biogeochemistry and microbiology are highly complex, however, and there is limited knowledge of many of the fundamental processes. Programs of basic, multidisciplinary research into these complex ecosystems will only answer using constructed wetlands and compost bioreactors to remediate AMD described. In wetlands flocculation is enhanced by high pH, high concentration of suspended solids, high ionic strength and algal concentration (Sheoran & Sheoran 2006). The removal rate through subsequent filtration for Ag, Cd, Zn and Pb was 26 to 100%. More than 50% of heavy metals can easily be adsorbed onto particulates in wetlands. The removal of Fe and Al, and Mn, but at a much higher pH, can be important processes taking place in wetlands when increasing the pH or the adding of oxygen, also inducing co-precipitation of other metals. Cu, Ni, Zn and Mn are co-precipitates with Fe, and Co, Fe, Ni and Zn with Mn (Sheoran & Sheoran 2006). The formation of carbonates can also take place if there is substantial bacterial formation of bicarbonate alkalinity or the wetland contains carbonate rich materials, removing Mn at Ni at 79 and 25%, respectively. Metal sulphide formation is another removal process in wetlands, indicated by a high bicarbonate/sulphate ratio, forming highly insoluble sulphides with Cu, Pb, Zn, Cd, As and Hg (Sheoran & Sheoran 2006). Often the most important removal process in wetlands is biological by uptake in roots, stems and leaves in plants (Sheoran & Sheoran 2006), with the highest per area unit uptake rate for metals in herbaceous plants and macrophytes.

Peat-based wetlands have been shown to remove metals both by organic matter and sulphide binding, with Cu mainly bound organically and as oxides when treating high strength runoff (Cu 35–50 ppm), and as sulphides when treating low strength (0.3–1.0 ppm) runoff (Sobolewski 1996).

A three step process for the effective removal of uranium (U) from dilute wastewaters involved the sequestration of U to aquatic plants such as algae, following removal of U-algal particulates from the water column to the sediments (Kalin *et al.* 2004). Step 3 involves reducing U (VI) to U (IV) and transforming the ions into stable precipitates in the sediments. The algal cells provide organic carbon and other nutrients to heterotrophic microbial consortia to maintain the low Eh, within which the U is transformed. Among the microorganisms, algae are of predominant interest for the ecological engineer because of their ability to sequester U and because some algae can live under many extreme environments, often in abundance. Algae grow in a wide spectrum of water qualities, from alkaline environments (Chara, Nitella) to acidic mine drainage wastewaters (Mougeotia, Ulothrix). If they could be induced to grow in wastewaters, they would provide a simple, long-term means to remove U and other radionuclides from U mining effluents.

Cyanide is together with fluoride, a variety of metals, and some petroleum hydrocarbons a common constituent present in groundwater from historical aluminium industry landfills (Gessner *et al.* 2005). Physicochemical removal processes are expensive and energy intensive. Both total and free cyanide were effectively reduced during 7 d detention, by 56 and 88%, respectively, in shallow, planted basins. Gasoline range organics and diesel range organics were reduced by 67%. These removals are lower bound estimates, because effluent concentrations were often below detection. First order areal removal rate constants were in the range 13–100 m/year for the various constituents. Preliminary studies indicated little volatilization of the cyanide, but significant microbial degradation, and essentially no harmful by-products.

Organic pollutants

The findings of organic pollutants are listed in Table 3. In water samples, PAH, bisphenol A, BTEX, hydrocarbons including diesel range organics, glycol, DDT, PCB, cyanide, benzene, chlorophenols and formaldehyde exceeded limit values. In sediments only PAH and PCB were found exceeding limit values.

Batch loaded constructed wetland systems can be expected to remove large quantities of non-halogenated polar organic solvents from municipal wastewater (Grove & Stein 2005). Biodegradation is identified as the dominant removal mechanism, but as much as 20% of the total removal may be attributed to sorption. Removal is generally better during summer than winter and varied between plant species. In most cases, data from petroleum industry wetland studies indicate that treatment wetlands are equally, or more, effective at removing pollutants from

Table 3 | Organic pollutants measured in wetlands

Pollutants Organic pollutants (μg/L) italic = wetland effluent, bold = >limit[a]	What/where	Concentration	Comment/removal	Reference
4-nitro-phenol. Used in pest. Production	Biological degradation, wetland soil, lab. Florida, USA	*Deg. At conc. From 10–100 mg/kg, no deg. At 10,000*	Increased degradation when inoculated	Laha & Petrova (1997)
Acetone, tetrahydrofuran, butanol (polar solvents)	Jar experimental wetlands, USA	100 mg/L	90% rem. achieved at 3 to > 14 d THF hardest to remove, no rem. In sterile controls	Grove & Stein (2005)
Acid orange 7 Acid dye	Vertical flow CW, Portugal	127 mg/L *30 $\mu M/m^2$* day removal	Up to 99% colour rem. 93% of org. C	Davies *et al.* (2006)
Aliphatic lipids	Natural wetland sediments, Iraq	Occurrence μg/g: alkanes 6.8–31, acids 4.1–5, alkanols 5.9–7.7, alkanoats 0.3–5.9	Sources %: Plants 24–30, microbial residues 30–42, petroleum 27–30	Rushdi *et al.* (2006a)
Benzene	Vertical flow CW, China	**1 g**/L Rem. % 73–90	Correlated with COD, DO, EC, NO_3, BOD	Tang *et al.* (2009)
Bisphenol A	Photodegradation dependence on DOM/ USA/lab	100 mM/L *2* 0–5 mg/L DOM	Deg. kinetics must faster than pure microbial. Photolysis prob. Important deg. Factor	Chin *et al.* (2004)
BTEX	Pilot system upward flow CW sand/gravel	**127–6,680**	Effectively (up to 80%) Aeration increased rem. 20 %	Bedessem *et al.* (2007)
Caffeine	Natural wetland/USA	*0.034–0.196*	Input to wetland/Dilution	Peeler *et al.* (2006)
Chlorobenzene, phenatrene	Desorption resistant organics, wetland plant uptake, green-house, USA	3.8 to 5.7% uptake	Soot should be removed from the sediment organic carbon calculations	Gomez-Hermosillo *et al.* (2006)
Chloroform, tetrachlor-methane	Coastal salt marsh, China. Atmospheric C. static flux chambers	*108 mM/m^2d*	Large sink in growing season,	Wang *et al.* (2007)
DDT and PCB	Glasshouse hyroponic DDT transformed to DDD by *Phragmites*	**50–1000** DDT **50–100** PCB	*Phragmites* root extracts degraded DDT and low Cl PCB, but not *O. sativa*	Chu *et al.* (2006b)
De-icing, glycol, acetate	Degradation on airport surfaces, cold temperature	**20** mg/L	Deg. (day^{-1}) of eth.glycol: 0.082, prop.glycol: 0.073, acetate: 0.033. Removal % as BOD at 8, 4, 1 deg.c: 33, 30, 21	Revitt & Worall (2003)
Disinfectant by-products, TOX, THM, HAA, DOC	3 large CW, USA	75–200	Red. %: Total org. halide: 13–55, tri-halomethan: 78–97, haloacetic acid: 67–96. Also wetland increase pot.DBP	Rostad *et al.* (2000)
Diesel fuel, Pb	Peat columns, USA	2 ppm Pb **897** ppm DRO	Pb mobile Diesel only mobile along bedding	Deiss *et al.* (2004)
Dioxin, furan, PCB, organochlorine pesticides	Natural reserve Krivoklatsko, Czech republic SPMD sampling	*Dioxins: up to 7 pg/L, PCB up to 2 ng/L, pest. up to 0.34 ng/l*	No toxicity found	Koci *et al.* (2007)
Effect of PAH on plant growth	*Baumea juncea, Baumea articulata, Schoenoplectus validus, junc. Subsecundus,*	Not influenced by PAH		Zhang *et al.* (2010)

(*continued*)

Table 3 | continued

Pollutants Organic pollutants (μg/L) italic =	What/where wetland effluent, bold = >limit[a]	Concentration	Comment/removal	Reference
EU-Regulated POP`s	Horizontal sub-surface wetland/Spain Loading: 36 mm/d	Tracer: clofibric acid Input: 2500	Removal >90% lindane, PCP, endosulfan, PC benzene; 80–90%: alachlor, chlorpyriphos; 20% mecoprop, simazine; not removed clofibric acid, diuron	Matamoros *et al.* (2007)
Explosives PETN + nitro-glycerine	Phytoremediation, Germany	1 mM	Enzyme introduced to transgenic plants, improved red	Rosser *et al.* (2003)
Explosives (TNT, RDX)	Hydroponic batch exp	1–1.4 mg/L	TNT 100% rem., decreased when C > 1.5–3.4 mg/L. No rem. with substrate only	Best *et al.* (1999)
Formalin	Biol. Aerated filter + CW	Formaldehyde: 10–**900** mg/L	BAF rem %; formaldehyde 98, TOC 92, CW 81, 25. Combination best for C and tox. Rem	Melian *et al.* (2008)
Glycol, BOD, ammonia	Engineered wetland, airport, USA	>95 & BOD, NH_4	> 4000 kg/d rem. of glycol	Higgins *et al.*, (2010)
Glycol, microbes	Pilot scale wetlands at Heathrow UK	**632–1,180** mg/L	54% rem. for subsurface, 74% for surface wetlands. Mostly aerobic microbial	Chong *et al.* (1999)
Halogenated phenols	Batch lab. experiments	**299–3,305** 'Plant Speciation' important for plant uptake Highest when pKa > pH	Uptake by *Lemna* Phenols with subst. in position 2 Highest uptake 2,4,6-TCP no uptake	Tront *et al.* (2007)
Heavy oil	Pilot scale subsurface flow CW, China Liaohe Delta	mg/L: COD 77, BOD 3.5, TKN 2.2, mineral oil 2.9	Rem. %: COD 81, BOD 89, mineral oil 89, TKN 81 Oil no effect on plants, but high loading negative	Ji *et al.* (2002)
Hormonal active	Mai Po Marsh, Hong Kong	Nonylphenol 0.029–2.59M octyl- 0.011–0.348	Highest in dry season. Correlate with low sediment TOC	Li & Gu (2007)
Hydrocarbons, groundwater	CW for HC poll. gw. natural gas condensate. Gravel + plants		100% rem. with aeration of subsurface, 50 % without	Anonymous (2002)
Hydrocarbons	Pilot system upward flow CW sand/gravel	8–**34** mg/L	Effectively remove diesel range hydrocarbons	Bedessem *et al.* (2007)
Hydrocarbons	Coral reef sediments	Petroleum hc	>100 ppm, 21–35C odd over even pre-dominance, complex, UCM, thermally mature, sterane/hopane	Volkmann *et al.* (1992)
Hydrocarbons (non-aromatic)	Sediments, Perl river estuary, China	EOM: 19–39 mg/kg NAH: 3–7	HC most important fraction of pollutants	Gao *et al.* (2007)
Hydrocarbons, cyanide, rhodamine det. time model	Waste from Al industry, Free Water surf. Wetlands, USA	Cyanide < **0.6** mg/L Diesel gas > 1	56–88% red. free and total cyanide after 7 d	Gessner *et al.* (2005)
Hydrophobic organic pollutants	Lake, Albania SPMD	4–11 μg/SPMD	39 compounds, PAH most abund. Sign. EROD effect	Rastall *et al.* (2004)
Nitro-aromatic sludge	Explosives, pharmaceuticals, vertical flow wetland, Sweden	14–790 ng/L	Planted wetlands better in reducing tox. No genotoxicity	Gustavsson *et al.* (2007)

(*continued*)

Table 3 | continued

Pollutants Organic pollutants (μg/L) italic =	What/where wetland effluent, bold = >limit[a]	Concentration	Comment/removal	Reference
Organo-chlorine pollution	2 Rivers, Argentina, agriculture +horticulture	Endosulfan, DDT and chlordanes high	4 times higher (per TOC) in horticulture	Miglioranza *et al.* (2004)
Organic pollutants and metals	Wetland sediments, chronology, Germany	Common: LAB, HCB, DPDE, PAH Modern: organo-tin, tonalide, triclosan	Common: pre – 70, modern: post-70	Heim *et al.* (2004)
Organic pollutants	Review: 10 groups of org. poll. + leachate + +, wetlands; spec. plants used; vertical flow CW, Austria	Aromatic and nitro-aromatics 2–250 mg/L, *< 0.01–2*	Effective and cheap, knowledge needed	Haberl *et al.* (2003)
PAH Pyrene	2 sed. From vert. flow wetland	200 mg/kg in liquid: 0–40 org. 0–150 min	K = 28 and 2 for org. and min. sed. 8–15% available for desorption	Chevron Cottin & Merlin (2007)
PAH	River, coastal wetland, France. Uptake in eels bile	20–1,600 ng/g	Highest in young eels	Ribeiro *et al.* (2005)
PAH, Cu	Experimental salt marsh wetland Cu uptake influenced by PAH	**100–10 000** *Cu, 1.6 PAH Spiked*	Up to 50% red. of Cu in sol. with PAH	Almeida *et al.* (2008)
PAH, PCB, DDT, chlordanes, heavy metals	Tidal marsh sediments	*Levels of PCB, DDT and As give probable effects*	All stations highly polluted	Hwang *et al.* (2008)
PAH + Hg	Fluorantene + pyrene	10–30 ng/g uptake in mussels	Pyrene has higher uptake and toxicity	Fabbri *et al.* (2006)
PAHs and LAS	pilot-scale CW FWS + SSW+ gravel filter in parallel	PAH: 79% & 56% in SSF 68% & 30% in FWS 73% & 401% in gravel filter	Absorption in solid media. SSF > FWS & gravel filter. Decrease with increase in temp	Fountoulakis *et al.* (2009)
PCB	Uptake in river Carp, Korea	0–5.41 ng/g	Max near factory. 138 & 153, penta- & hexa most frequent	Moon *et al.* (2006)
PCB & PCDD	Landfill leachate	16–960 pg/l PCDD, 23–2,180 pg/l PCB	Concentrations increased with time	Ham & Lee (2007)
PCB removal	Aerobic PCB degradation/ USA	**3,000**	Glucose and biphenyl amendment of soil effective	Luo *et al.* (2008)
Pentachlor-nitrobenzen	Nitroaromatic pesticide deg. In anaerob. Sed. Lab. Surface water DOM	1 μM/L	99% transformation	Hakala *et al.* (2007)
Perchlorate (explosives)	Phytoremediation, 4 species, USA	0.2–20 mg/L. In sed. 3–3,138 mg/kg	Rem. all species except water weed. K: 0–0.013/day	Susarla *et al.* (2000a)
Perchlorate (explosives)	Phytoremediation, 6 species wastes, USA	0.2–20 mg/L. In sed. 0–981 mg/kg	Rem. all species except water weed. K: 0.001–0.007 h^{-1}. Smart weed best, willow worst	Susarla *et al.* (2000b)
Petroleum industry effluents	Review 70 m^2–50 ha BOD, TSS, TN, TP	*Petroleum rem %: BOD: 55–97, COD 38–86, oil grease 54–94, phenol 10–94*	Can be very effective rem%: municipal 53–73, pulp&paper 26–71, livestock 42–65	Knight *et al.* (1999)

(*continued*)

Table 3 | continued

Pollutants Organic pollutants (μg/L) italic = wetland effluent, bold = >limit[a]	What/where	Concentration	Comment/removal	Reference
Phenol bacterial degradation	*Pseudomonas pseudoalcaligenes*	mg phenol/CFU/h, Planktonic: 1.04×10^{-9} root gravel biofilms: 1×10^{-10}	Biofilms much higher in phenol and other organics removal, due to greater bacterial biomass	Kurzbaun *et al.* (2010)
Phenol, p-cresol	Wetland sed. laboratory anaerobic deg. of humic subs. Increase red. of org. pollutants	1.5 mM	Active in wetlands when org. C is easily available	Cervantes *et al.* (2008)
Phytoremediation at market level	Exclusion and stabilisation of metal- and As-cont. soils	Rhizo degradation of organic pollutants	Rhizofiltration/ rhizodegradation + phytodegradation of organics in CW	Mench *et al.* (2010)
Polar compounds	Natural wetland sediments, Iraq	Occurrence μg/g: n alk. Acids C_{8-20} 7.8, alkanols, C_{12-39} 29–122, stereoids 27–175, triterpen-oids 0.7–11	Plants, microbes Residues plankton, bacteria	Rushdi *et al.* (2006b)
POP's	Sediment toxicity in Sunderban wetlands, Ganges, India	ng/g dw. PCB: 0.5–**27**, PAH: 9–**4,250**, DDT, HCH-low	g-HCH most toxic, m-ERM-Q must useful	Binelli *et al.* (2008, 2009)
RDX Explosive Hexahydro-trinitro-triazine	Vertical flow mesocosms wetland /USA	1–10 mg/L 89–96% removal	2 years' degradation Study HRT = 2 days	Lowa *et al.* (2008)
TCE (trichlor-ethylene)	Lab	*1–1,127* 2–4	Volatilization most important, plus mineralization to CO_2	Bankston *et al.* (2002)
Tetrachlor ethylene, bacteria	Exposure to microorganisms in wetlands, Canada	1–5 ppm	No neg. exposure detected	McDaniel *et al.* (2007)
Textile wastewater	Review of lignin-degrading microbial	Wetlands good for removal	Up to 50% rem. Must be below toxic levels	Diaz *et al.* (2007)
Triton-X100 surfactant microbes	Pilot scale reed bed, SSF CW, Italy	30–300 mg/L	60% removal of X100 and metabolites octyl-fenol + OPEO	Sacco *et al.* (2006)
Xenobiotic, hydrocarbons, nitroaromtics, chlorinated aliphatics, pesticides, surfactants	Anaerobic biodegradation, USA	*High Cl: cannot be degraded aerobically Persistent: =slow*		Zhang & Bennett (2005)

[a]See Table 2. All concentrations in μg/L unless otherwise stated.

petroleum industry wastewaters than from other types of wastewater. A CW can be used to upgrade the quality of heavy oil-produced water to an acceptable level. Large amounts of COD, BOD, mineral oil and TKN can be removed through CW. Complex distributions of hydrocarbons occur in most aquatic sediments. Total concentrations can range from a few parts per million in non-polluted intertidal and oceanic areas to parts per thousand in heavily contaminated estuarine, lake and near-shore environments. Petroleum-derived residues are common in coastal and estuarine areas, particularly those near urban or industrial centres.

Contaminant transport risks in peat lands may be more pronounced for lateral flow (along the bedding planes) than for vertical flow (across the bedding planes), especially with respect to diesel range organics (DRO). Macropore transport appeared to be an important pathway for both DRO and Pb leaching, especially for lateral flow. Estimates of the soil adsorption–desorption coefficients for Pb were similar, ca. 1.33 L/mg, which was several orders of magnitude lower than most published values and indicated moderate mobility, probably facilitated by organic complexing. For DRO, the adsorption–desorption coefficient was ca. 1,000 L/mg, indicating low mobility, consistent with previously published work (Deiss *et al* 2004).

A pilot-scale SSF CW was effective in removing BTEX and diesel range organics from polluted groundwater, but not MBTE (Bedessem *et al*. 2007). Forced aeration improved benzene removal. Iron precipitation was a potential pretreatment. The seasonal removal efficiencies for benzene decreased with time, and were much higher for the indoor compared to the outdoor wetlands. The presence of particles improved the benzene removal efficiency through adsorption. However, the presence of plants had no significant impact on benzene removal, with removal efficiencies at ca. 90%, and 73 and 80% for indoor and outdoor constructed wetlands, respectively.

Reclamation of biologically-treated wastewaters is frequently enhanced with the use of constructed wetlands and wastewater lagoons that remove nutrients and persistent organic pollutants (Bankston *et al*. 2002). Anaerobic transformation was apparent for TCE in a shallow aquifer at a rate, however, not sufficient for its complete attenuation, allowing the TCE to enter a wetland into which the groundwater was discharged. Indigenous microbial communities present in two different wetland soils, one sandy and the other organic, had the capacity to mineralize TCE. The presence of the *Typha latifolia* increased the rate of mineralization of TCE above that observed by the indigenous soil microorganisms, suggesting that using broad-leaved cattails can stimulate natural attenuation of TCE in wetlands.

Most explosives are considered to be a major hazard to biological systems due to their toxic and mutagenic effects. Phytoremediation of explosives (TNT) and (RDX) in groundwater using constructed wetlands is a potentially economical remediation alternative (Best *et al*. 1999). TNT disappeared completely from groundwater incubated with plant and substrate treatments. Highest specific removal rates were found in submersed plants in water star grass and in all emergent plants except wool-grass. TNT declined less with substrates, and least in controls without plants. Mineralization was very low, and evolution of volatile organics negligible. RDX disappeared less rapidly than TNT from groundwater.

Runoff of organic and inorganic contaminants from live firing ranges is a challenging issue because of the extensive size, the variable nature of runoff, the random occurrence of surface contamination, and general inaccessibility (Low *et al*. 2008). This is particularly true with energetic compounds such as hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX). One potentially promising technology for addressing runoff in a passive, sustainable, and low-cost manner is the use of constructed wetlands to intercept flow. Significant RDX removal occurred (89–96%) for all loading rates (160–1,600 mg/(m^2 d)) at a hydraulic retention time of approximately 2 days. RDX degradation occurred in both NO_3 and SO_4 dominated electron acceptor zones. Removal of plants is believed to have a small impact on overall RDX removal.

Bisphenol A (BpA) degradation by direct photolysis is significantly slower than its rate in the presence of DOM (Chin *et al*. 2004). In natural waters, the direct photolytic pathway would be even less important due to light screening effects. Surprisingly, differences in the rate of indirect BpA photolysis were relatively small between DOM samples. BpA photo reactivity was not correlated to the structural components of DOM. The addition of methanol, a hydroxyl radical scavenger, to reaction solutions slowed, but did not completely quench, the indirect photolysis of BpA. This observation suggests that BpA photodegrades via multiple pathways involving other transients formed by the photolysis of DOM. Competitive experiments using 2,4,6-trimethylphenol also reduce the reaction rate of BpA by DOM and imply that other DOM-derived photo transients are involved in the reaction. The reaction rate coefficients reported under solar-simulated irradiance in the presence of DOM are significantly faster than those reported for the microbial degradation of BpA. Thus, in natural surface waters photosensitized transformation of BpA by dissolved organic matter may be as important as biodegradation.

An advanced type of constructed wetland (CW) called an engineered wetland (EW) has been developed which allows more efficient removal of contaminants from wastewaters and stormwaters at rates in many cases an order of magnitude higher than those achievable with ordinary CWs (Higgins *et al*. 2010). In one type of advanced aerobic EW, the wastewater being treated flows subsurface (SSF) beneath an aggregate substrate which is aerated mechanically from below. This ecotechnology allows very much higher removals (>95%) of those wastewater contaminants amenable to

aerobic treatment (e.g., BOD, ammonia). A large 1.9 ha EW system treating glycol-contaminated water is now operating at Buffalo Niagara International Airport (BNIA) in upper New York State, USA. This system, which is successfully treating glycol-contaminated stormwater runoff and spent glycol from cold weather aircraft deicing activities, involves four very large aerated EW cells and is capable of treating up to 4,620 m^3/d of glycol-contaminated water containing up to 4,535 kg/d of BOD (the BOD-equivalent of a city of 50,000 people), even in the coldest weather.

Constructed wetlands are efficient in the treatment of airport runoff containing glycols and the subsurface variant shows best year round performance (Chong *et al*. 1999). Plants and microorganisms show good tolerance to glycols. Large populations of anaerobic but dominated by aerobic and actinomycetes populations were found within the reed bed substrate. Aerobic microorganism dominated the growing season, but was also most adversely affected by dosing experiments.

The non-ionic surfactant triton X-100, in dosage 30 to 300 mg/L produced t-octylphenol (OP) and its mono- di- and tri-ethoxyl derivates, among others, in the outlet of a SSF CW (Sacco *et al*. 2006). About 40% of the metabolites left the CW. More aerobic bacteria adhered to the roots than to gravel. The appearance of new strains showed their adaption to the chemicals.

Octanol-phenol and nonyl-phenol are found frequently in surface water wetlands (Xiang-Li *et al*. 2007). Maximum level was found in dry conditions. Correlation was observed between the levels of OP and NP, and total organic matter. Based on the data that are available in the literature, OP and NP levels may possibly cause histological and biochemical endocrine disrupting responses.

Formalin wastewater containing high concentrations of formaldehyde and TOC was treated and detoxified with biological aerated filter (BAF) and wetland reactors (Melian *et al*. 2008). At loading rates up to 0.9 kg/m^3 d both reactors provided high efficiencies but the BAF was more efficient at treating formaldehyde and TOC with average eliminations of 98 and 92%, respectively. However, the BAF effluent was not always detoxified according to the Lemna minor toxicity test as growth inhibition was above 20%. Although the elimination of formaldehyde and TOC achieved by the wetland reactors (81 and 25%, respectively) was lower than that of the BAF, they provided higher total nitrogen reduction. The combination of BAF and wetland reactors is proposed as a novel and convenient method for the treatment of formalin wastewater as it provided almost complete elimination of formaldehyde and detoxified samples with L. minor.

Pesticides

The findings of pesticides are listed in Table 4. In the water phase the compounds exceeding limit values were atrazine, simazine, terbutylazine, metolachlor, mecoprop, endosulfan, chlorfenvinphos and diuron.

The herbicides are generally water soluble and less toxic, the fungicides intermediate, and the insecticides have very low water solubilities and are highly toxic (Table 5).

The use of vegetated wetlands for accelerating pesticide removal from agricultural runoff is gaining acceptance as a best management practice. Wetlands can effectively remove hydrophobic organic compounds (HOC), and concentrations of herbicides, pesticides, and organic wastewater contaminants decrease 0–99% between inlet and outlet. There are indications of accumulation of HOC in fish, including p,p-DDE and trans-nonachlor (Barber *et al*. 2006).

Alachlor is a good chemical probe for examining indirect photolysis due to its lack of reactivity by either direct photolysis or dark reaction pathways and its ubiquity as an agricultural herbicide. DOM plays a role in promoting an indirect photolytic mechanism that is highly pH dependent.

Wetland monitoring results indicate that high atrazine loadings in response to the storm events were observed. The wetland was able to completely remove atrazine caused by the storm runoff with methanogenesis as possibly the dominant biodegradation process in the lower wetland layer. Atrazine accumulation on sediments was not observed based on the sediment analysis. Three wetland species accumulated atrazine in different amounts. It has been suggested that it is not feasible to predict or model the uptake of herbicides by wetland plants (Cejudo-Espinosa *et al*. 2008).

Pesticide losses to the environment are unwanted due to possible environmental and health hazards. The pesticides propachlor, metalaxyl and chlorphenvinfos were applied on an arable soil plot in the watershed of an experimental wetland to study the efficiency with respect to retention of sediments, nutrients and pesticides (Blankenberg *et al*. 2007). All pesticides were found in the experimental wetland, with peak concentrations shortly after spraying. In 2003 pesticide retention varied from 11 to 42% and in 2004 retention varied from 19 to 56 %. Comparing eight different wetland filters, we found that wetland models with flagstones and straw, respectively, had higher total pesticide retention than a standard Norwegian surface flow wetland. Statistical analyses showed that the treatments were significantly different from zero in six of the wetlands for removal of propachlor, for removal of metalaxyl none

Table 4 | Pesticides measured in wetlands

Pollutants Pesticides (μg/L) italic = wetland effluent, bold = >limit[a]	What/where	Concentration	Comment/removal	Reference
29 pesticides	FWS (26), SSF (2), 1 pond		Removal: −19 (linden) to 100	Kadlec & Wallace (2009)
7–13 pesticides	FWS	Input in g/day	Removal from −11% (metalaxyl) to 67% (propachlor)	Braskerud & Haarstad (2003) and Haarstad & Braskerud (2005)
6 pesticides	Constructed wetlands, fish USA Summer + winter	*ng/L* *carbaryl: <1, chlorpyrifos: <1, diazinon: 49, dichloro-aniline: 340, prometryn: 13, simazine: <1*	40–99% removal Uptake in fish: most DDE and PCB, removal based on SPMD: good except for DDE	Barber *et al.* (2006)
6 pesticides	FWS and leachate	0.02 to 230	60 to 90%	Haarstad & Mæhlum (2007)
Alachlor	Photolysis in surface agricultural water, USA. Natural and CW	5 ppm M	At high pH deg. Only when N was high. At low pH more deg.	Miller & Chin (2005)
Atrazine	Non-point source pesticide, stormwater events, mountain wetland	**26–259** *Not detected*	Methanogenesis	Kao *et al.* (2002)
Atrazine	Lab. Wetland sediments, USA	1 μg/g	Not detectable after 30 d. 25–30% mineralization	Runes *et al.* (2001)
Atrazine accumulates in wetland soil, plant and water	Pot experiments in greenhouse, Mexico	**4–30** mg/L	Acc. depends on pest. C and plant species. 30% in soil, 40% in plant root, and 10–20 in water	Cejudo-Espinosa *et al.* (2008)
Captan, delta-methrin, glyphosate, iso-proturon, pirimicarb	MPC (Max. permissible conc.): μg/kg/μg/L Captan, 1.3/0.11 delta-methrin, 1.3/0.0003 glyphosate, 150 iso-proturon, 5.3/0.32 pirimicarb, 2.2/0.09	Mpc-100[a] Mpc	Stronger microbial activity reduction at MPC than at 100[a]MPC	Widenfalk (2005)
Carbendazim, 2,4-D	Deg. studies with culture from rice sed. Japan	0.10–3 Mm	Complete degradation	Pattanasupong *et al.* (2004)
Cotton pesticides	Wetlands/open dams	30–110 diuron/pro-methryn, endosulfan, chlorpyrifos	Open dams more efficient than wetlands. More microbiol. Endosulfan slow in light chlorpyrifos quick	Rose *et al.* (2007)
Cotton pesticides aldicarb, endosulfan, diuron, fluometuron	Pilot scale 100 m^2 + 200 m^2 CW, Australia	10–90 *0–15*	Removal %: first season 22–53, second 32–90	Rose *et al.* (2006)
CW, passive samplers	4 weeks study	ng/L: diuron: **192**; simazine: 70; atrazine: 5	*diuron: 94; simazine:30; atrazine: 2*	Page *et al.* (2010)
Dimethyl-isophtalate	Wetland soil degradation/China	Complete degradation	Requires a mix of microorganisms	Li & Gu (2007)

(*continued*)

Table 4 | continued

Pollutants Pesticides (μg/L) italic = wetland effluent, bold = >limit[a]	What/where	Concentration	Comment/removal	Reference
DDT & PCB	FWS	0.1 to 1 mg/l	Both DDT & PCB is taken up and transformed in plants	Chu *et al.* (2006a, b)
Endosulfan	Low org. C clay soil. Soil, water plant	**2–10** *in storm runoff*	Residues in plant, soil plant cover positive	Kennedy *et al.* (2001)
Lindane and DDT	Content in fish. High in *Anguilla bicolor bicolour* low in *Heteropneustes fossilis*	2.1 to 77.9 μg/kg	Lower than ADI	Dhananjayan & Muralidharan (2010)
Mecoprop MCPA terbutylazine	Surface flow CW, Spain. 100 m^3/d WWTP effluent	mecoprop: 7.8; MCPA: 2.0 terbutylazine: **2.3**	Rem. %: mecoprop: 22–91, MCPA: 79–93, TBazine: 1–80	Matamoros *et al.* (2008c)
Metolachlor	SSF CW, USA	Some samples >**10**	Distr. of pest in sed. And water not stereo dependent	Eish & Wells (2008)
Nutrients, metals, pesticides	Golf course, US	*0.01 atrazine, 0.22 simazine* *0.56 MCPA,* In June. No pest. during storms	97% N removal, 74% P 90% C/COD rem. during storm 74% for Cl	Kohler *et al.* (2004)
Organic pesticides	Wetland, China	Atrazine: 0.22, DDT: 0.001, dimethoate: 0.35, lindane: 0.002, malathion: 0.12, parathion: 0.002		Qu *et al.* (2011)
Organochlorine & -phosphor. pesticides	Residues in river water % sed., Mexico	DDT: 2 DDE: 247 ng/g, endosulfan: 814 ng/g	Heavy use of insecticides	Hernández-Romero *et al.* (2004)
Organochlorine pesticides	River, coastal wetland, France. Uptake in eels bile	1–1,100 ng/L	Most common endosulfan. Highest in liver: lindane	Oliveira Ribeiro *et al.* (2005)
Organochlorines in edible fish	Carp (*Cyprinus carpio*), barbell (*Barbus barbulus*)	μg/kg lipid: Carp: 1680, lindane, 410, HCB 70, PCB lower	53–88% DDT, some samples > food limit	Davodi *et al* (2011)
Permethrin PBO (Piperonyl butoxide)	Wetlands,USA + lab.	0.04–0.08 PBO	No detectable levels of PBO to increased toxicity of permethrin	Amweg *et al.* (2006)
Pesticides Ca. 55 compounds	Wetlands/Norway	0.01–0.62 *0.02–0.29*	Removal: 2003: 11–42% 2004: 19–56% Wetlands with flagstones and straw seems better	Blankenberg *et al.* (2007)
Pesticides in leachate wetlands		Bentazon: 0.6, cyprodinil: 0.02, dichloro-prop: 0.18, chlorphen-vinphos: **1.1**, MCPA: 0.02, Mecoprop: **148**	Sediments (mg/kg) bentazon 1.7; endosulfan 19.4; mecoprop 6.5	Haarstad & Mæhlum (2008)

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Table 4 | continued

Pollutants Pesticides (μg/L) italic = wetland effluent, bold = >limit[a]	What/where	Concentration	Comment/removal	Reference
Pesticides in nat. wetland	Tidal estuary	simazine: 0.4–**0.8**; pendimethaline 0.07–0.4; diuron: 22; terbutylazine **52**		Barba-Brioso *et al* (2010)
Phenoxy acids MCPA + clofibric acid	Wetland + support matrix (LECA, perlite, sand), Portugal	Up to 1000	100% removal of MCPA in small LECA, less for clofibric acid	Dordio *et al*. (2007)
Triazophos	Hydroponic wetlands + Plants *canna indica*	0.7–3.6 mg/L	Half-life 20–358 d	Cheng *et al*. (2007)
α og β – endosulfan	Soil degradation/Pakistan	14–88% deg.	Max. at $T = 30^{\circ}$ pH = 8 Organic acids stimulating, amino acids inhibitory effect	Arshad *et al*. (2007)

[a]See Table 2. All concentrations in μg/L unless otherwise stated.

were significantly different, and for removal of chlorfenvinphos four treatments were significantly different. For the three compounds none of the relative treatments were significantly different from L4. Chemical properties of the pesticides could explain some of the behaviour in the watershed and in the wetland.

Light expanded clay aggregates (LECA) has a high sorption capacity for phenoxy compounds (MCPA and clofibric acid), a pH buffering capability and a suited hydraulic permeability. Two other media, expanded perlite and sand, do not exhibit a significant sorption capacity for the studied phenoxy compounds (Dordio *et al*. 2007).

A bacterial consortium capable of simultaneously degrading the fungicide, carbendazim, and the herbicide, 2,4-dichlorophenoxyacetic acid (2,4-D) was obtained by enrichment of soil samples collected from paddy fields, to completely degrade up to 100 mM carbendazim and 3 mM 2,4-D within 36 and 24 h, respectively, in batch culture, but a lag time was observed after precultivation in a rich medium (Pattanasupong *et al*. 2004).

Table 5 | Characteristics of the detected pesticides

Pesticide	K_{oc} Log	$T_{1/2}$ Days	Sol.[a] μg/L	pK_a –	PEC μg/L	Type[b]	Aerob Deg.	Anaerob Deg.
2,4-D	1.68	14	900,000	2.87	2.2	h		
Alachlor					20.00	h		
Atrazine	1.98	45	33,000	1.74	0.40	h	Low	
Chlorfenvinphos	1.72	33	145,000		0.00025	i		Bio.
DDT	5.60	2,000	1		0.050	i		
Endosulfan	4.09	150	330		0.050	i	Mod.	
MCPA	2.04	25	825,000	3.09	13	h		Not an.
Mecoprop		21	860,000	3.11	44	h		Not an.
Metalaxyl	2.23	80	7,100,000		120	f	Mod.	Low an.
Metolachlor					10	h		
Propachlor	1.90	12	613,000		0.29	h		
Simazine	2.10	89	6,200	1.60	0.42	h	Low	Mod. An
Terbutylazine			9,000		0.20	h		

[a]At pH = 7. [b]h = herbicide, f = fungicide, i = insecticide. Deg. = degradation. Mod. = moderate.

Table 6 | PPPCs measured in wetlands

Pollutants Pharmaceuticals and personal care products (PPCP) (µg/L) italic = wetland effluent, bold = >limit[a]	What/where	Concentration	Comment/removal	Reference
6 pharmaceuticals, 1 veterinary 2 PPCP	Surface flow CW, Spain. 100 m^3/d WWTP effluent	Pharm: 0.04–2.1, vet: 1.06, PPCP: 0.9–2.9	Rem. %: pharm: 16–99, vet. 64, 85–90	Matamoros *et al.* (2008c)
8 emerging pollutants	4 not degraded: atrazine, DEET, picloram, clofibric acid	Duckweed incr. rem.; fluoxetine, ibuprofen, 2,4-d, triclosan	Uptake and sorption	Reinholdt *et al.* (2010)
Antibiotics	Wetlands and rivers Canada	4,000/**1,000** Tetracycline 80% ads. In wetlands	Decrease pH increase ads.	Verma *et al.* (2007)
Chlofibric acid, ibuprofen, carbamazepine	SSF CW, lab.	12.5 mg/L. No difference between org. C sources	Chlo.acid as tracer, little rem. of Carbamazepine, Ibuprofen by aerobic deg. (51%)	Matamoros *et al.* (2008a, b)
Chlofibric acid, ibuprofen, carbamazepine	2 pilot SSF CW, Spain 55 m^2	12.5 mg/L	Chlofibric acid behave as Br-tracer Ibuprofen rem. 48% in deep, 81% in shallow CW (aerobic), Carbamazepine 16–26	Matamoros *et al.* (2005)
EDC (endocrine disrupting chemicals)	FWS	9 EDCs	Completely or significantly removed	Chapman (2003)
Fluoroquinolone antibiotics Wetland soil sorption experiment	Sorption K_d-values	20 & 60 ppm	Ciprofloxacin, Ofloxacin, Norfloxacin	Conkle *et al.* (2010)
HOC (hydrophobic organic carbon) 20 comp. + 6 pests. +46 trace elements in water	Constructed wetlands, fish USA Summer + winter	*ng/L* *Bisphenol A: 25–**104**;* *caffeine: 87–181;* *cholesterol: 300–870; 17-α-estradiol: <5–425;* *triclosan: 86–92*	40–99% removal	Barber *et al.* (2006) Very good overview
Model wetland	1 anaerobic reactor, 2 parallel wetlands and 1 in series		Ibuprofen 99% in B3, Naproxen & Diclofenac 93% in B1& B2, anaerobic: tonalide & BpA 94% & 83% in 1&2	Avila *et al.* (2010)

[a]See Table 2. All concentrations in micro g/l unless otherwise stated.

The factors controlling the distribution of organochlorine pesticides (OCP) in creek sediments were the distance from the sources and the grain size composition. The total OCP concentrations ranged from 6 to 25 ng/g dry weight (Ribeiro *et al.* 2005). However, when OCPs were expressed in ng/g TOC one creek presented 4-fold higher total levels as a consequence of a higher OCP input during the recent past in that watershed. These results indicate that anthropogenic activity such as urbanization and agricultural practices have affected the sediment quality. Endosulfans, DDTs and chlordanes were the dominant compounds in the selected creeks. Endosulfan sulphate was the most abundant pesticide in most of sampling stations. The predominance of metabolite with respect to parent compounds suggests a contamination mainly by runoff from aged and weathered agricultural soils. Although total OCP concentrations in creek sediments from the coastal zone are considered high, they do not represent a potential hazard for wildlife.

Pharmaceuticals, personal care products PPCs and hormone disrupters

The findings of PPCPs are listed in Table 6. There are few listings of these compounds in the commonly used water quality limit values, except for some well-known endocrine disrupters such as nonylphenol, phtalates etc.

A major source of endocrine disrupting chemicals (EDC) has been identified as sewage effluent. More recently,

technologies such as coronation, UV treatment and advanced filtration have improved the quality of effluent discharged to the environment but there are still unresolved issues relating to poorly understood chemistries relating to EDCs. Compounds detected in the raw effluent included pesticides, herbicides, some heavy metals and the human hormones 17-β-estradiol and estrone. Most of these were removed by the advanced treatments at the water reclamation plant, with only trace concentrations of some compounds present in the final effluent.

The mass emission rate of 12 pollutants from a wastewater treatment plant (WWTP) secondary effluent into a small tributary was determined. The pollutants tested included pharmaceutical and personal care products (PPCPs) and herbicides (Matamoros *et al*. 2008c). Furthermore, a 1-hectare surface flow constructed wetland (SFCW) was evaluated for pollution removal. Whereas the low concentration values (ng/L) of PPCP discharge into the tributary was comparable to inter- and intra-campaigns, herbicides and a veterinary drug (flunixin) exhibited a high variability in concentrations (mg/L). Moreover, removal efficiencies were often higher than 90% for all compounds, with the exception of carbamazepine and clofibric acid (ca. 30–47%). As expected, a seasonal trend of pollutant removal in the wetland was observed for compounds with low biodegradation and moderate photodegradation rates (i.e. naproxen and diclofenac).

Subsurface flow constructed wetlands (SSFs) constitute a wastewater treatment alternative to small communities due to the low operational cost, reduced energy consumption, and no sewage sludge production. Although much information is available about conventional water quality parameters in SSF constructed wetlands, few data are available regarding specific contaminants (Matamoros *et al*. 2008a, b). Focus on the behaviour of three widely used pharmaceuticals (clofibric acid, ibuprofen, and carbamazepine) in two pilot SSF constructed wetlands planted with *Phragmites australis* and characterized by different water depths (i.e., 0.3 and 0.5 m). These SSFs partially treat the urban wastewater from a housing development (ca. 200 inhabitants). The behaviour of clofibric acid was similar to that of bromide, and no sorption into the gravel bed occurred. On the other hand, carbamazepine showed a higher sorption than bromide and clofibric acid, which is attributable to its interaction on the gravel bed. Accordingly, the use of clofibric acid as a hydraulic tracer is proposed, taking into account its low residence time. Ibuprofen removal was 81% in the shallow SSF and 48% in the deep one. The less anaerobic environment of the shallow wetland could explain differences in removal efficiency.

A discrete injection experiment was carried out in a horizontal subsurface flow pilot plant constructed wetland to evaluate the behaviour of selected priority pollutants. A total of eight European Priority Pollutants listed in the Water Framework Directive were considered, including a commonly used herbicide (mecoprop). The pollutants encompassed a variety of chemical classes and physicochemical properties. They included organochlorine, organophosphorus, phenols, chloroacetanilides, triazine, phenoxycarboxylic acid and phenylurea pesticides (Matamoros *et al*. 2005). A time series of composite effluent samples and discrete gravel bed samples from the wetland were analysed. Response curves for all the pollutants injected from effluent concentrations were obtained and compared with the tracer (clofibric acid). On the basis of an analysis of the samples taken 21 days after the injection, priority pollutants were classified into four groups according to their removal efficiency. These groups were: (i) the highly efficiently removed (>90%), namely lindane, pentachlorophenol, endosulfan and pentachlorobenzene; (ii) the efficiently removed (80–90%), namely alachlor and chlorpyriphos; (iii) poorly removed (20%), namely mecoprop and simazine; and (iv) recalcitrant to elimination, namely clofibric acid and diuron. Taking into account the poor accumulation of the injected contaminants in the gravel bed (0–20%), biodegradation and plant uptake are postulated as the most likely elimination pathways for the pollutants.

In wetland water, the addition of EDTA generally promoted the release of tetracycline indicating that a portion of the tetracycline was bound to metal ions (Verma *et al*. 2007). Decreasing the pH of the wetland water led to increased adsorption of tetracycline suggesting either that tetracycline epimerizes or binds, by hydrogen bonding, to acidic portions of organic material in the water. The combination of the effects of matrixes of the water, light and UV radiation therefore play a significant role in catalysing the removal of tetracycline from different waters. In deep waters and in systems where sunlight is highly attenuated, the effects of light on tetracycline may be considerably reduced.

CONCLUSIONS

A literature review shows the occurrence of more than 500 organic and metallic pollutants in wetlands. The removal of

heavy metals is reported typically in the order of 30 to 60%, but can reach 80 to 90%. An effective removal of hydrocarbons requires aerobic conditions. Typical removal of hydrophobic organic compounds is 50 to 100%. The removal of explosives is more efficient when microbiological processes and plants are included in the wetland, and inefficient if only adsorption processes are used. Pesticide removal is typically in the order of 40 to 99%, but some compounds show much less removal. The removal of PPCPs seems to be good in wetlands. The performance of wetland systems of removing pharmaceuticals and personal care products (PPCPs) are similar to that obtained in conventional activated sludge wastewater treatment plants.

DISCLAIMER

The content of this paper is based on the available literature. There might be errors due to incorrect reproduction of the data.

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First received 14 April 2011; accepted in revised form 23 August 2011