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Heavy metals in sediments from constructed wetlands treating municipal wastewater

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Abstract Constructed wetlands are commonly used for treatment of municipal sewage. The treatment is usually aimed at removal of organics, suspended solids, nutrients and microbial pollution. The information on removal and fate of heavy metals is very limited. The purpose of this study was to evaluate the amount of sediments and heavy metal concentration in the sediments in filtration beds of seven constructed wetlands with horizontal subsurface flow treating municipal sewage with various length of operation. The results revealed that concentrations of Cd, Ni, Pb, Cu, Cr and Zn in the sediment are mostly comparable with concentrations occurred in natural unpolluted or slightly polluted wetlands. The concentrations are much lower than those found in wetlands impacted with mine drainage waters or wastewater from industrial operations. Concentrations of studied heavy metals exceeded only occasionally limits set by the Czech legislation. However, when heavy metal concentrations are evaluated within the filtration material the concentrations are well below the limits set for soils in the Czech Republic. The results also revealed that concentrations of heavy metals in the sediment do not reflect the time of operation probably due to build-up of sediments from suspended solids contained in wastewaters. However, the sediment mass increases during the course of operation and consequently the metal mass increases as well.

Keywords Constructed wetlands · Gravel · Heavy metals · Horizontal flow · Wastewater treatment

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Introduction

During last two decades the constructed wetlands with horizontal subsurface flow (HF CWs) have increasingly been used in the Czech Republic to treat municipal wastewater (Vymazal 1995, 1996, 2002, 2009; Vymazal and Kröpfelová 2005). The first constructed wetland was put in operation in 1989 and at present, there are about 300 systems in operation. Trace elements are usually not the target of the treatment of municipal wastewater but their concentrations in the sediments within the filtration bed may be the concern when the filtration bed would need to be excavated and disposed. So far only few investigations have been aimed at the heavy metals concentrations in the filtration beds of constructed wetlands with subsurface flow treating sewage (e.g. Obarska-Pempkowiak and Klimkowska 1999; Obarska-Pempkowiak 2001; Vymazal 2003; Vymazal and Krása 2003; Lesage et al. 2007a, b).

Redox potential and pH of the sediment–water system are the major factors known to influence the mobility of trace elements in wetlands (DeLaune et al. 1998; Koretsky et al. 2008). However, in most municipal sewage the pH is around neutral and, therefore, this parameter does not affect mobility and retention of heavy metals in constructed wetland too much. Particularly in wetlands, oxidation and reduction reactions are of prime importance (Du Laing et al. 2008). Under aerobic conditions, the most important process affecting accumulation of heavy metals is precipitation of Fe/Mn hydrous oxides (Singer and Stumm 1970). The most important processes affecting heavy metals accumulation/mobilization under anoxic and anaerobic conditions are creation of hydrogen sulfide via sulfate reduction and dissolution of Fe/Mn hydrous oxides (Khalid et al. 1978; Green et al. 2003; De Volder et al. 2003; Mansfeldt 2004).

Sedimentation has long been recognized as the principle process in removal of heavy metals from wastewater in constructed wetlands. However, it is not a simple straightforward physical reaction and other chemical processes such as precipitation and co-precipitation have to occur first (Yao and Gao 2007). Iron, manganese and also aluminum can form under aerobic conditions insoluble compounds through hydrolysis and/or oxidation. This leads to formation of variety of oxides, oxyhydroxides and hydroxides (Wieder 1989; Batty et al. 2002; Woulds and Ngwenya 2004; Sheoran and Sheoran 2006). Once associated with the particulate phase, these elements become subject to removal from the water via sedimentation. The stability of these inorganic compounds is controlled primarily by the system pH, the solubility of the product, and concentrations of the metals and relevant anions (Gambrell 1994; Sheoran and Sheoran 2006). At near-neutral to slightly alkaline pH levels, metals tend to be effectively immobilized (Gambrell 1994). Co-precipitation is an adsorptive phenomenon in wetland sediments. The concentration and distribution of many elements, such as Ni, Cu, Zn or Cd, in sediments and overlying waters are strongly influenced by adsorption and/or co-precipitation with Fe and Mn oxides (Krauskopf 1956; Jenne 1968; Feely et al. 1983; Ferris et al. 1989). Copper, nickel, zinc and manganese are co-precipitated in Fe oxides and cobalt, iron, nickel and zinc are co-precipitated in manganese oxides (Stumm and Morgan 1981). In addition, zinc is reported to be retained on iron plaques at the surface of plant roots (Otte et al. 1995). However, in filtration beds of HF CWs anoxic/anaerobic conditions prevail (e.g., Dušek et al. 2008) and therefore precipitation of Fe/Mn compounds is not the major retention mechanism as Mn and Fe precipitates dissolute under these conditions (Laanbroek 1990; Jacobson 1994; Lovley 1995; Green et al. 2003; Cooper et al. 2006).

Under reducing conditions, dissimilatory sulfate reduction transforms SO_4^{2-} to H_2S during respiration by several genera of strictly anaerobic bacteria by reaction with a variety of organic substrates (Gambrell and Patrick 1978; Laanbroek and Veldkamp 1982; Mandernack et al. 2000; Megonikal et al. 2004). Most of the heavy metals react with hydrogen sulfide to form highly insoluble metal sulfides (Krauskopf 1956; Stumm and Morgan 1981; Kosolapov et al. 2004):

$$M^{2+} + H_2S \rightarrow MS \downarrow + 2H^+$$

where M^{2+} represents a divalent metal ion such as Fe^{2+} (pyrite, FeS_2; pyrrhotite, FeS), Pb^{2+} (galena, PbS), Cd^{2+} (CdS), Cu^{2+} (covellite, CuS; chalcocite, CuS_2; chalcopyrite, $CuFeS_2$), Ni^{2+} (NiS) or Zn^{2+} (sphalerite, ZnS). These compounds are very stable and insoluble under anaerobic conditions. However, under oxidized conditions sulfides dissolute and release metals. This may occur, for example, as a consequence of oxygen release from plant roots in the rhizosphere (Engler and Patrick 1975; Gambrell et al. 1980; Jacob and Otte 2003).

Heavy metals may also form carbonates when the bicarbonate concentration in water is high. Although carbonates are less stable than sulfides, they can still perform a significant role in initial trapping of metals (Ramos et al. 1994; Sobolewski 1996; Sheoran and Sheoran 2006; Du Laing et al. 2008). Carbonate precipitation is especially effective for the accumulation of lead and nickel in wetlands (Lin 1995).

Metal complexes with large molecular weight organics tend to be effectively immobilized. There is some evidence that at least some metals are more tightly bound by organics under anoxic or reducing conditions compared with upland conditions because humic material may become structurally less complex under oxic conditions (Gambrell and Patrick 1978; Gambrell et al. 1980; Guo et al. 1997). However, complex formation with soluble and insoluble organic matter under all conditions of pH and oxidation intensity occurs (Verloo and Cottenie 1972).

The purpose of this study was to evaluate the amount of sediments and heavy metal concentration in the sediments in filtration beds of constructed wetlands with horizontal subsurface flow treating municipal sewage with various time of operation.

Materials and methods

Seven constructed wetlands with horizontal subsurface flow (Fig. 1) with time of operation varying

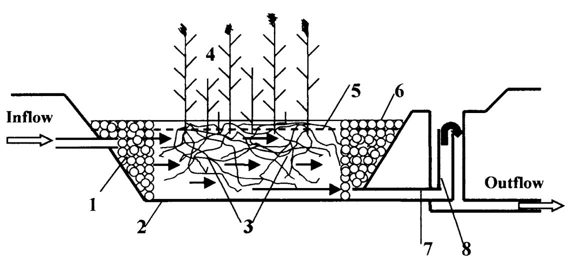

Fig. 1 Schematic representation of a constructed wetland with horizontal sub-surface flow. 1—distribution zone filled with large stones, 2—surface of the bed, 3—water level in the bed, 4—impermeable liner, 5—medium (e.g., gravel, crushed stones), 6—collection zone filled with large stones, 7—collection drainage pipe, 8—outlet structure for maintaining of water level in the bed. The *arrows* indicate only a general flow pattern (Vymazal 2001)

between 2 and 16 years (Table 1) were sampled in 2008. In each constructed wetland samples were taken in the inflow, middle and outflow zones (three samples in each zone). Gravel or crushed rock samples were taken using the reinforced stainless steel soil sampler (so called "Russian corer") which was driven into the filtration substrate to a depth of 60 cm. Samples were divided into surface (0–20 cm) and bottom sections (20–60 cm) in order to evaluate vertical distribution of sediments. In the laboratory, samples were cleaned from roots and freeze-dried under low pressure and temperature. Dried sediment material was weighed, homogenized and passed through a 0.5 mesh sieve. After drying both sediment and filtration material volume were determined in order to calculate the volume ratio between sediment and filtration material. 500 mg of the dry sediment was digested in reverted (Löfelt) aqua-regia (4.5 ml HNO_3 and 1.5 ml HCl) under high pressure and temperature in microwave apparatus (MARS-5, CEM, USA) according to the modified U.S. EPA method 3052 (U.S. EPA 1995). After digestion, the sample was filtered in order to obtain a clear sample. Heavy metals were analyzed by ICP-MS (PQ-ExCell, VG-Thermo Elemental, Winsford, Cheshire, UK) according to U.S. EPA method 200.8 (U.S. EPA 1994). For statistical analyses of heavy metal concentrations along the filtration beds paired-sample t-test ($P < 0.05$) was used. Differences between sediment concentrations in the filtration beds were statistically evaluated through the two-way ANOVA ($P < 0.05$) for vertical (top and bottom) and horizontal (inflow, middle and outflow) profiles.

Table 1 Major design parameters of monitored constructed wetlands

	Čejkovice	Libníč	Břehov	Slavošovice	Mořina	Příbraz	Sp. Poříčí
Start of operation	2006	2006	2003	2001	2000	1999	1992
Length of operation (Years)	2	2	5	7	8	9	16
Designed PE	500	400	100	150	700	300	700
Type of sewer system	Combined	Combined	Combined	Combined	Separate	Combined	Combined
Area (m^2)	1,900	1,280	504	830	3,520	1,400	2,500
Filtration material	CR	CR	G	G	CR	G	G
Fraction (mm)	4–16	4–8	4–8	4–32	4–8	4–8	0–16
Vegetation	PHA	PHA	PHA + PG	PG	PHA + PG	PHA + PG	PHA + PG

PE population equivalent; *CR* crushed rock, *G* gravel; *PHA Phalaris arundinacea*, *PG Phragmites australis*

Results and discussion

Concentrations of heavy metals in the sediments

Cadmium

Under anoxic conditions, cadmium forms very insoluble compounds with sulfide (CdS) and under slightly reduced to oxidized conditions solid carbonate ($CdCO_3$) is a major control mechanism for cadmium solubility (Khalid (1980). Precipitation of carbonate can be microbially mediated, for example, by *Alcaligenes denitrificans* (Remacle et al. 1992). Under aerobic conditions, cadmium could be adsorbed or co-precipitated with oxides, hydroxides, and hydrous oxides of Fe, Mn and possible Al (Khalid 1980). Cadmium complexed with the organic fraction may be divided into chelated and organic bound. Chelated Cd is the fraction that is loosely attached to immediately mobile and easily decomposable organic material while organic-bound Cd is the fraction incorporated into the insoluble organic material and can be solubilized only after intense oxidation of the organic matter (Khalid 1980).

Fig. 2). In all systems, the concentrations in the top layer did not significantly differed from those found in bottom layers with the exception of Příbraz and Spálené Poříčí where the Cd concentration was

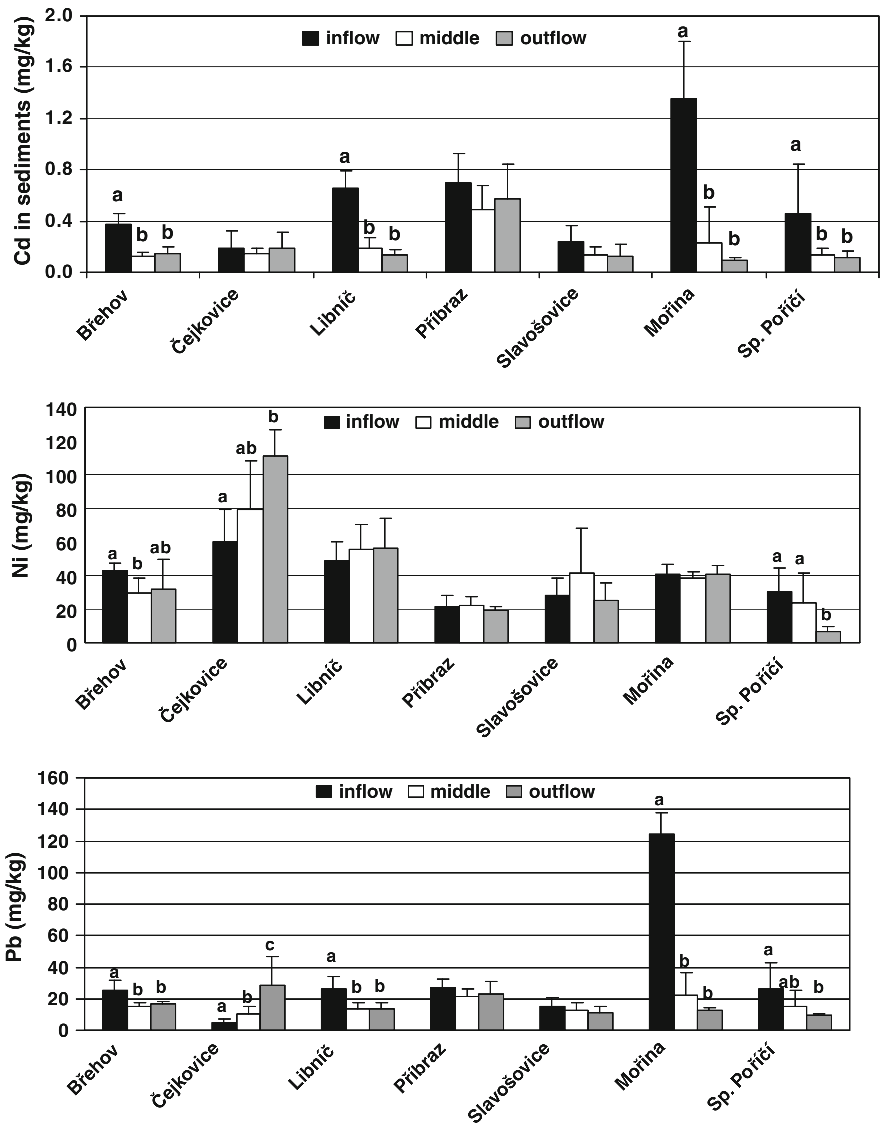

Fig. 2 Concentration of cadmium, nickel and lead in seven horizontal sub-surface flow constructed wetlands. *Different letters* indicate significant difference at $\alpha = 0.05$ between the means. *Bars without letters* are not significantly different

significantly higher at the outflow zone in the top layer. The results shown in Fig. 2 indicate that in Břehov, Libníč, Mořina and Spálené Poříčí the concentration of Cd near the inflow was significantly higher than in the middle of the bed and near the outflow. Similar observations were also reported by Lesage et al. (2007a, b) from HF CWs in Zemst and Zevergem, De Pinte, Flanders, Belgium and by Vymazal (2003) from HF CW Nučice in the Czech Republic. Also, the Cd concentrations found in our study were comparable with those reported by Lesage et al. (2007b). The values are slightly lower than those reported by Lesage et al. (2007a), Haberl and Perfler (1990), Samecka-Cymerman et al. (2004), Gschlössl and Stuible (2000) or Zuidervaart (1996) from HF CWs in Belgium, Austria, Poland, Germany and the Czech Republic, respectively. On the other hand, the concentrations were lower than concentrations reported from constructed wetlands for road runoff or mine wastewater treatment (Table 2). The data on sediment concentration in various wetlands (Table 2) indicate that concentrations found in our study are

Table 2 Concentration of cadmium (mg/kg) in sediments of natural and constructed wetlands

Location	Type of sediment	Concentration	Reference
Natural wetlands, unpolluted			
Zambia	Unpolluted wetland	<0.2	Von der Heyden and New (2004)
South Carolina, USA	Unpolluted sediments	0.03	Babcock et al. (1983)
Czech Republic	Fishponds	0.03–1.0	Švehla et al. (2002)
Estonia	64 mires	0.12	Orru and Orru (2006)
Czech Republic	Fishponds	0.13–2.34	Pokorný et al. (1999)
Russia	Unimpacted West Siberia tundra wetlands	0.24	Zhulidov et al. (1997)
UK	Unpolluted pond	1.4	Ye et al. (1997)
Canada	Man-made lake marsh	1.0–1.9	Murdoch and Capobianco (1978)
Spain	Unaffected marsh	1.45	Madejón et al. (2006)
UK	Unpolluted wetland	0.1–2.0	Scholes et al. (1998)
Hong Kong	Natural marsh	ND–3.46	Liang and Wong (2003)
UK	Unpolluted wetland	4.2	Shutes et al. (1993)
Worldwide	Estuaries and deltas	0.13–7.4	Accornero et al. (2008)
Denmark	Oligotrophic lake	11.0	Schierup and Larsen (1981)
Natural wetlands, polluted			
Turkey	Polluted marsh	0.22	Aksoy et al. (2005)
Gaza Strip	Slightly polluted wetland	ND–0.4	Shomar et al. (2005)
Turkey	Lake sediments impacted by urbanization	0.14–0.57	Duman et al. (2007)
Vietnam	Water spinach cultivation sediments amended with wastewater	0.33–0.67	Marcussen et al. (2008)
Zambia	Wetland affected by mine waters	<0.1–0.8	Von der Heyden and New (2004)
Poland	Three lakes receiving wastewater	1.2–1.8	Szymanowska et al. (1999)
Tanzania	Natural wetland receiving various wastewaters	0.6–2.5	Ojo and Mashauri (1996)
Spain	Marsh affected by tailing spill	3.29	Madejón et al. (2006)
Poland	Anthropogenic lakes (former open cut brown coal mines)	<0.08–3.3	Samecka-Cymerman and Kempers (2001)
Canada	Infiltration field for petroleum refinery wastes	<0.5–4.0	Higgins and Brown (1999)
New Jersey, USA	Freshwater tidal marsh receiving urban runoff	4.8–6.1	Simpson et al. (1983)
China	Coastal wetlands with various degree of pollution	1.84–6.43	Li et al. (2007)
UK	Natural wetland receiving highway runoff	2.9–8.7	Mungur et al. (1994)
Russia	Impacted West Siberia tundra wetlands	9.0	Zhulidov et al. (1997)
UK	Natural wetland receiving highway runoff	9.7	Shutes et al. (1993)

Table 2 continued

Location	Type of sediment	Concentration	Reference
China	River sediments contaminated with e-wastes	ND–10.3	Wong et al. (2007)
China	Polluted lakes sediment	0.11–10.4	Yang et al. (2007)
Poland	Two lakes near copper smelter/ore processing plant	9.4–12.9	Samecka-Cymerman and Kempers (2004)
Mexico	Lagoon affected by industrial wastes	5.3–13.5	Carranza-Álvarez et al. (2008)
China	Pond receiving mine waters	20	Ye et al. (1997)
USA	Contaminated urban lake	4–22	DeLaune et al. (1989)
UK	Pond receiving metal-enriched drainage	9–26	Ye et al. (1997, 1998)
Belgium	Freshwater tidal marshes	7.3–34.1	Teuchies et al. (2008)
India	Anthropogenic lake affected by coal mine drainage	29.9–40.2	Mishra et al. (2008)
Michigan, Indiana, Wisconsin	Contaminated sediments	0.01–45.6	Folsom and Lee (1981)
China	Wetlands receiving wastewaters	17–46	Deng et al. (2004)
China	Stream near Pb/Zn mine	10–48	Deng et al. 2008)
Denmark	Eutrophic lake	65.4	Schierup and Larsen (1981)
China	Impacted wetland	26 (2.6–71)	Bi et al. (2007)
Spain	Saltmarsh affected by mining wastes	77	Jiménez-Cárceles et al. (2008)
China	Sediments contaminated with municipal and industrial wastewaters	1.2–642	Peng et al. (2008)
Constructed wetlands[a]			
New York, USA	HF CW for landfill leachate, 1 year in operation	<0.1–0.3	Eckhardt et al. (1999)
Belgium	HF CW, 4 years in operation	0.5–0.9	Lesage et al. (2007a)
Belgium	VF CW, 4 years in operation	1.2	Lesage et al. (2007a)
Poland	FWS CW	1.0–1.9	Samecka-Cymerman et al. (2004)
Belgium	HF CW, 3 years in operation	0.2–2.5	Lesage et al. (2007b)
Austria	HF CW, 6 years in operation	2.0–2.3	Haberl and Perfler (1990)
Germany	HF CW, 10 years of operation	0.3–4.0	Gschlössl and Stuible (2000)
Poland	VF CW, 5 years in operation	4.8–5.2	Obarska-Pempkowiak and Klimkowska (1999)
Czech Republic	4 HF CWs, 2–5 years in operation	0.24–5.75	Zuidervaart (1996)
Belgium	FWS CW, 16 years in operation	0.5–7.7	Lesage (2006)
UK	2 FWS CWs treating urban runoff	3.0–9.6	Scholes et al. (1998)
Poland	HF CW	7.6–11.4	Samecka-Cymerman et al. (2004)
UK	FWS CW for road runoff	7.4–11.5	Carapeto and Purchase (2002)
China	FWS CW for Pb/Zn mine	17–24.8	Lan et al. (1990)
Czech Republic	HF CW, 3 years in operation	27.5	Vymazal (2003)
India	HF CW	92–129	Oke and Juwarkar (1996)

ND not determined, *HF* horizontal sub-surface flow, *VF* vertical sub-surface flow, *FWS* free water surface

[a] For municipal sewage unless specified

also comparable with those found in natural unpolluted wetlands. It is obvious that the highest Cd concentrations are found in wetlands receiving industrial wastewaters or mine drainage waters.

Nickel

Under oxic or suboxic conditions, Ni sorbs to Mn oxides and can substitute for Ni in the lattice of some Mn oxides

Table 3 Concentration of nickel (mg/kg) in sediments of natural and constructed wetlands

Location	Type of sediment	Concentration	Reference
Natural wetlands, unpolluted			
South Carolina, USA	Unpolluted sediments	1.5	Babcock et al. (1983)
Estonia	64 mires	3.7	Orru and Orru (2006)
Italy	Volcanic lake	3.1–7.5	Baldantoni et al. (2009)
Portugal	Three reed stands	11.7	Santos-Oliveira et al. (1999)
Texas, USA	Three marshes	3.1–14.1	Lindau and Hossner (1982)
UK	Unpolluted wetland	2–23	Scholes et al. (1998)
Canada	Man-made lake marsh	9.2–24.1	Murdoch and Capobianco (1978)
Zambia	Unpolluted wetland	8–38	Von der Heyden and New (2004)
Worldwide	Estuaries and deltas	10–51	Accornero et al. (2008)
Hong Kong	Natural marsh	17.3–51.7	Liang and Wong (2003)
Czech Republic	Fishponds	11.5–52.4	Pokorný et al. 1999)
Czech Republic	Fishponds	8–72	Švehla et al. (2002)
Natural wetlands, polluted			
Poland	Anthropogenic lakes (former open cut brown coal mines)	0.3–31	Samecka-Cymerman and Kempers (2001)
Tanzania	Natural wetland receiving various wastewaters	0.2–33.5	Ojo and Mashauri (1996)
Turkey	Lake sediments impacted by urbanization	20.1–34.3	Duman et al. (2007)
Poland	Two lakes near copper smelter/ore processing plant	12–38	Samecka-Cymerman and Kempers (2004)
Uganda	Lake shore affected by mining	28.2–42.8	Lwanga et al. (2003)
Turkey	Polluted marsh	44.4	Aksoy et al. (2005)
Slovenia	Anthropogenic lake contaminated with ash from lignite coal	14.6–48.1	Mazej and Germ (2009)
Vietnam	Water spinach cultivation sediments amended with wastewater	29.9–52.8	Marcussen et al. (2008)
Gaza Strip	Slightly polluted wetland	4.5–60	Shomar et al. (2005)
New Jersey, USA	Freshwater tidal marsh receiving urban runoff	59–70	Simpson et al. (1983)
USA	Contaminated urban lake	20–87	DeLaune et al. (1989)
Zambia	Wetland affected by mine waters	3–220	Von der Heyden and New (2004)
China	Coastal wetlands with various degree of pollution	13.8–318	Li et al. (2007)
China	River sediments contaminated with e-wastes	12.4–543	Wong et al. (2007)
Canada	Silver mine vicinity	590	Moore and Sutherland (1981)
Sudbury, Canada	Near copper smelter	9,372	Taylor and Crowder (1983)
Constructed wetlands			
Belgium	VF CW, 4 years in operation	24	Lesage et al. (2007a)
UK	FWS CW for road runoff	6–35	Carapeto and Purchase (2002)
Belgium	FWS CW, 16 years in operation	7–35	Lesage (2006)
Belgium	HF CW, 3 years in operation	32–42	Lesage et al. (2007b)
Czech Republic	HF CW, 3 years in operation	45.4	Vymazal (2003)
Belgium	HF CW, 4 years in operation	40–46	Lesage et al. (2007a)
Italy	FWS CW. non-point pollution	36.1–57.2	Bragato et al. (2006)
Germany	HF CW, 10 years in operation	7–71	Gschlössl and Stuible (2000)
UK	2 FWS CWs treating urban runoff	17–187	Scholes et al. (1998)

For details see Table 2

(Green-Pedersen et al. 1997; Tonkin et al. 2004). Under anoxic/anaerobic conditions nickel forms insoluble sulfides (Sobolewski 1999) and is incorporated into pyrite (Morse and Luther 1999). Also carbonates could be an effective sink for nickel (Lin 1995).

The concentration of nickel in sediments of monitored constructed wetlands varied between 7.0 and 111 mg/kg (Fig. 2). In all systems, the concentrations in the top layer did not significantly differed from those found in bottom layers. The results shown in Fig. 2 indicate that in Břehov, and Spálené Poříčí the concentration of Ni near the inflow was significantly higher than in the middle of the bed and near the outflow. In Libníč, Příbraz, Slavošovice and Mořina, the Ni concentration in the sediments did not vary too much. In Čejkovice, the highest Ni concentration was measured at the outflow. The literature results on nickel distribution along the filtration bed also vary. Vymazal (2003) found significantly higher Ni concentration in the inflow zone while Lesage et al. (2007b) observed only a slight decrease along the bed and Lesage et al. (2007a) a slight increase in Ni concentration in the sediments along the bed. Nickel concentrations were comparable with Ni concentrations reported from constructed wetlands treating municipal sewage (Table 3). Also, the Ni concentrations are within the range of Ni concentrations reported from both unpolluted and polluted wetlands. Results presented in Table 3 revealed that by far the highest Ni concentrations are found in wetlands receiving industrial wastewater, mine drainage waters and also road runoff.

Lead

Koretsky et al. (2008) pointed out that lead, like Zn and Cu, is a chalcophile that forms discrete sulfide phases and may also bind strongly to organic matter. Also carbonates could be an effective sink for lead (Lin 1995). It has been shown that lead also strongly adsorbs to Fe/Mn oxides and it has been found in association with rhizosphere Fe(III) plaques (Dzombak and Morel 1990). However, it has been concluded that the Pb is not trapped by Fe oxides, but rather is complexed to organic matter either in the rhizosphere solution or on the root surface (Sundby et al. 2005).

The concentration of lead in sediments of monitored constructed wetlands varied between 9.3 and 125 mg/kg but most values were lower than 30 mg/kg (Fig. 2). In all systems, the concentrations in the top layer did not significantly differed from those found in bottom layers. The results shown in Fig. 2 indicate that in Břehov, Libníč, Mořina and Spálené Poříčí the concentration of Pb near the inflow was significantly higher than in the middle of the bed and near the outflow. In Příbraz and Slavošovice the Pb concentration in the sediments did not vary too much and decreased slightly along the bed. In Čejkovice, similarly to Ni, the concentrations gradually increase along the bed. Lesage et al. (2007a, b) reported that lead concentration in the sediment decreased along the bed. Vymazal (2003) found a significant decrease after 16 m of the bed but than the concentration increased again and after 48 m the Pb concentration was only slightly lower as compared to the concentration near the inflow. Lead concentrations found in our study were comparable with Pb concentrations reported from natural unpolluted and lightly polluted wetlands (Table 4) In comparison with the results reported from various constructed wetlands the measured concentrations are slightly lower. The data in Table 4 also clearly indicate that Pb concentrations in sediments of wetlands receiving mining drainage waters and waters affected by smelters are up to two orders of magnitude higher.

Copper

Copper forms under anoxic conditions very insoluble compounds with sulfur, including both cupric and cuprous sulfides (Sobolewski 1999; Morse and Luther 1999) and may also associate with pyrite (Huerta-Diaz et al. 1993). Copper also forms insoluble hydroxides and carbonates (Morel and Hering 1993) but those are important in presence of sulfides. Copper also forms strong complexes with organic matter and can be bound to Fe/Mn oxides under oxic conditions via formation of ternary complexes with organic matter (Achterberg et al. 1997).

The concentration of copper in sediments of monitored constructed wetlands varied between 6.3 and 139 mg/kg but most values were lower than 75 mg/kg (Fig. 3). The concentrations of Cu in the top layer did not differ from those found in the bottom layers in all seven systems. With the exception of Čejkovice and Příbraz, in all other system the Cu concentration was significantly higher in the

Table 4 Concentration of lead (mg/kg) in sediments of natural and constructed wetlands

Location	Type of sediment	Concentration	Reference
Natural wetlands, unpolluted			
Estonia	64 mires	3.3	Orru and Orru (2006)
Russia	Unimpacted West Siberia tundra wetlands	5.7	Zhulidov et al. (1997)
Zambia	Unpolluted wetland	7–10	Von der Heyden and New (2004)
UK	Unpolluted pond	26	Ye et al. (1997)
Spain	Unaffected marsh	38	Madejón et al. (2006)
UK	Unpolluted wetland	4–40	Scholes et al. (1998)
Czech Republic	Fishponds	12–47	Švehla et al. (2002)
Canada	Man-made lake marsh	18.2–63.7	Murdoch and Capobianco (1978)
Czech Republic	Fishponds	28.4–67.6	Pokorný et al. (1999)
UK	Unpolluted wetland	71	Shutes et al. (1993)
Hong Kong	Natural marsh	17.1–90.7	Liang and Wong (2003)
Italy	Volcanic lake	47–93	Baldantoni et al. (2009)
Denmark	Oligotrophic lake	361	Schierup and Larsen (1981)
Worldwide	Estuaries and deltas	20–372	Accornero et al. (2008)
Natural wetlands, polluted			
Turkey	Polluted marsh	7.91	Aksoy et al. (2005)
Poland	Three lakes receiving wastewater	9.7–12.9	Szymanowska et al. (1999)
Turkey	Lake sediments impacted by urbanization	12.9–17.8	Duman et al. (2007)
Russia	Impacted West Siberia tundra wetlands	34	Zhulidov et al. (1997)
Slovenia	Anthropogenic lake contaminated with ash from lignite coal	10.4–36.4	Mazej and Germ (2009)
Spain	Marsh affected by tailing spill	45	Madejón et al. (2006)
Mexico	Lagoon affected by industrial wastes	20.3–55.2	Carranza-Álvarez et al. (2008)
Poland	Anthropogenic lakes (former open cut brown coal mines)	1–64	Samecka-Cymerman and Kempers (2001)
Vietnam	Water spinach cultivation sediments amended with wastewater	32.5–67.4	Marcussen et al. (2008)
Czech Republic	Eutrophic pond	1–68	Zuidervaart (1996)
India	Anthropogenic lake affected by coal mine drainage	45–74	Mishra et al. (2008)
Zambia	Wetland affected by mine waters	9–75	von der Heyden and New (2004)
Tanzania	Natural wetland receiving various wastewaters	9.1–85	Ojo and Mashauri (1996)
China	Polluted lakes sediment	27–86	Yang et al. (2007)
China	Coastal wetlands with various degree of pollution	12.3–86	Li et al. (2007)
China	Impacted wetland	66–160	Bi et al. (2007)
Australia	Contaminated urban streams	9.7–182	Cardwell et al. (2002)
Gaza Strip	Slightly polluted wetland	2.5–193	Shomar et al. (2005)
China	Paddy fields	33–501	Deng et al. (2006)
UK	Natural wetland receiving highway runoff	506	Shutes et al. (1993)
China	River sediments contaminated with e-wastes	28.6–590	Wong et al. (2007)
USA	Contaminated urban lake	102–1,130	DeLaune et al. (1989)
Denmark	Eutrophic lake	1,298	Schierup and Larsen (1981)
UK	Natural wetland receiving highway runoff	929–1,329	Mungur et al. (1994)
Canada	Silver mine vicinity	1,800	Moore and Sutherland (1981)
New Jersey, USA	Freshwater tidal marsh receiving urban runoff	237–2,161	Simpson et al. (1983)
Poland	Two lakes near copper smelter/ore processing plant	1,460–2,220	Samecka-Cymerman and Kempers (2004)
China	Stream near Pb/Zn mine	400–4800	Deng et al. (2008)
China	Pond receiving mine waters	5,686	Ye et al. (1997)

Table 4 continued

Location	Type of sediment	Concentration	Reference
China	Wetlands receiving wastewaters	112–11,161	Deng et al. (2004)
China	Wetlands affected by mining activity and smelters	649–11,185	Deng et al. (2006)
China	Sediments contaminated with municipal and industrial wastewaters	74–13,900	Peng et al. (2008)
Spain	Saltmarsh affected by mining wastes	16,845	Jiménez-Cárceles et al. (2008)
UK	Pond receiving metal-enriched drainage	430–18,894	Ye et al. (1997, 1998)
Constructed wetlands			
Italy	FWS CW, non-point pollution	17.3	Mattiuzzo et al. (2007)
New York, USA	HF CW for landfill leachate	1.1–19.6	Eckhardt et al. (1999)
Poland	VF CW, 5 years in operation	40–42	Obarska-Pempkowiak and Klimkowska (1999)
Czech Republic	4 HF CWs, 2–5 years in operation	1.3–56.5	Zuidervaart (1996)
Germany	HF CW, 10 years of operation	4–90	Gschlössl and Stuible (2000)
Belgium	VF CW, 4 years in operation	98	Lesage et al. (2007a)
Belgium	HF CW, 4 years in operation	44–95	Lesage et al. (2007a)
Poland	HF CW	97–114	Samecka-Cymerman et al.(2004)
UK	FWS CW for road runoff	55–126	Carapeto and Purchase (2002)
Poland	FWS CW	114–147	Samecka-Cymerman et al.(2004)
Belgium	HF CW, 3 years in operation	14–162	Lesage et al. (2007b)
UK	2 FWS CWs treating urban runoff	38–350	Scholes et al. (1998)
Belgium	FWS CW, 16 years in operation	50–364	Lesage (2006)
Czech Republic	HF CW, 3 years in operation	155 (52–409)	Vymazal (2003)
China	FWS CW for Pb/Zn mine	4,941–7,019	Lan et al. (1990)

For details see Table 2

inflow zone as compared to the middle and outflow zones. Lesage et al. (2007a, b) observed a steep decrease in Cu sediment concentration in two HF CWS in Belgium. Vymazal and Krása (2003) reported slight decrease in Cu concentration along the filtration bed of a HF CW in the Czech Republic. The copper concentrations found in our study were comparable with higher values found in unpolluted wetlands and with lower end of the range reported for polluted wetlands (Table 5). The copper concentrations shown in Fig. 3 were similar to the concentrations reported from Poland and Italy and also by Zuidervaart (1996) who studied heavy metals in the Czech constructed wetlands more than 10 years ago (Table 5). On the other hand, copper concentrations found in our study were lower than concentrations reported from Belgium (Table 5). The data in Table 5 also clearly indicate that Cu concentrations in sediments of wetlands receiving mining drainage waters and waters affected by smelters are up to two orders of magnitude higher.

Chromium

Contrary to most heavy metals such as Zn, Cd, Pb or Ni, chromium undergoes a change in oxidation state as a consequence of soil oxidation–reduction conditions (Gambrell 1994). These conditions play a major role in chromium speciation, solubility and mobility with reduction transformations being microbially mediated (Masscheleyn et al. 1992; Cervantes et al. 2001). DeLaune et al. (1998) reported that reduction of Cr(VI) occurs at approximately same redox levels as nitrate reduction. Under oxic and suboxic conditions chromium typically sorbs to Fe, and especially Mn, oxides (Davison 1993; Guo et al. 1997; Achterberg et al. 1997). Under anoxic sediments, reduced chromium is not readily incorporated into sulfides (Huerta-Diaz et al. 1998) but instead tends to associate with organic matter (Otero and Macias 2002). Also Guo et al. (1997) reported that under reducing conditions, the behavior of Cr is controlled primarily by insoluble large molecular humic materials.

Table 5 Concentration of copper (mg/kg) in sediments of natural and constructed wetlands

Location	Type of sediment	Concentration	Reference
Natural wetlands, unpolluted			
Estonia	64 mires	4.4	Orru and Orru (2006)
Russia	Unimpacted West Siberia tundra wetlands	16	Zhulidov et al. (1997)
Louisiana, USA	Freshwater marsh	16.4	Fejitel et al. (1988)
UK	Unpolluted wetland	4–20	Scholes et al. (1998)
Czech Republic	Fishponds	32.4	Pokorný et al. (1999)
Italy	Lake shore	10.3–43	Baudo et al. (1985)
Canada	Man-made lake marsh	7.0–43.7	Murdoch and Capobianco (1978)
Spain	Unaffected marsh	47	Madejón et al. (2006)
Italy	Volcanic lake	6.1–76	Baldantoni et al. (2009)
Hong Kong	Natural marsh	18.8–86.6	Liang and Wong (2003)
Belgium	Freshwater tidal marshes	79–198	Teuchies et al. (2008)
Zambia	Unpolluted wetland	53–463	Von der Heyden and New (2004)
Natural wetlands, polluted			
Turkey	Polluted marsh	6.72	Aksoy et al. (2005)
Poland	Anthropogenic lakes (former open cut brown coal mines)	0.4–18.6	Samecka-Cymerman and Kempers (2001)
Slovenia	Anthropogenic lake contaminated with ash from lignite coal	6.1–24.5	Mazej and Germ (2009)
China	Impacted wetland	27 (17–33)	Bi et al. (2007)
Turkey	Lake sediments impacted by urbanization	17.8–35.5	Duman et al. (2007)
Spain	Marsh affected by tailing spill	61	Madejón et al. (2006)
Vietnam	Water spinach cultivation sediments amended with wastewater	34–62.1	Marcussen et al. (2008)
China	Polluted lakes sediment	12.1–72.9	Yang et al. (2007)
Russia	Impacted West Siberia tundra wetlands	134	Zhulidov et al. (1997)
USA	Contaminated urban lake	19–140	DeLaune et al. (1989)
China	Stream near Pb/Zn mine	160–220	Deng et al. 2008)
Ontario, Canada	Near Cu smelter	3,738	Taylor and Crowder (1983)
New Jersey, USA	Freshwater tidal marsh receiving urban runoff	111–129	Simpson et al. (1983)
Uganda	Lake shore affected by mining	270–273	Lwanga et al. (2003)
China	Sediments contaminated with municipal and industrial wastewaters	27.9–452	Peng et al. (2008)
China	Coastal wetlands with various degree of pollution	ND–351	Li et al. (2007)
Spain	Saltmarsh affected by mining wastes	418	Jiménez-Cárceles et al. (2008)
Gaza Strip	Slightly polluted wetland	4.7–566	Shomar et al. (2005)
Worldwide	Estuaries and deltas	7–648	Accornero et al. (2008)
UK	Natural wetland receiving highway runoff	323–1,441	Mungur et al. (1994)
China	River sediments contaminated with e-wastes	17–4,540	Wong et al. (2007)
Poland	Two lakes near copper smelter/ore processing plant	4,600–5,620	Samecka-Cymerman and Kempers (2004)
China	Wetlands receiving wastewaters	95–5770	Deng et al. (2004)
Zambia	Wetland affected by mine waters	109–12,112	Von der Heyden and New (2004)

Table 5 continued

Location	Type of sediment	Concentration	Reference
Constructed wetlands			
Poland	FWS CW	16.2–31.9	Samecka-Cymerman et al. (2004)
Italy	FWS CW, non-point pollution	21.6–33.4	Bragato et al. (2006)
Italy	FWS CW, non-point pollution	11.9–43.4	Mattiuzzo et al. (2007)
Poland	VF CW, 5 years in operation	57–65	Obarska-Pempkowiak and Klimkowska (1999)
Poland	HF CW	90–99	Samecka-Cymerman et al. (2004)
Czech Republic	HF CW, 3 years in operation	110	Vymazal and Krása (2003)
Czech Republic	4 HF CWs, 2–5 years in operation	1.3–117	Zuidervaart (1996)
Belgium	HF CW, 4 years in operation	52–139	Lesage et al. (2007a)
Germany	HF CW, 10 years of operation	4–143	Gschlössl and Stuible (2000)
UK	2 FWS CWs treating urban runoff	17–178	Scholes et al. (1998)
Belgium	VF CW, 4 years in operation	201	Lesage et al. (2007a)
Belgium	HF CW, 3 years in operation	15–288	Lesage et al. (2007b)
Belgium	FWS CW, 16 years in operation	19–308	Lesage (2006)

For details see Table 2

The concentration of chromium in sediments of monitored constructed wetlands varied between 13 and 163 mg/kg but in Příbraz, Slavošovice, Mořina and Spálené Poříčí the average Cr concentrations in sediments did not exceed 45 mg/kg (Fig. 3). The concentrations of Cu in the top layer did not differ from those found in the bottom layers in all seven systems. In Břehov and Spálené Poříčí the highest Cr concentrations were recorded in the inflow zone while in Čejkovice and Příbraz the highest concentrations were recorded in the outflow zone. Lesage et al. (2007a, b) observed a slight decrease in Cr sediment concentration in Belgium. The chromium concentrations found in our study were higher as compared to values found in natural unpolluted wetlands and are similar to concentrations found in polluted wetlands (Table 6). The data in Table 6 indicate that Cr concentration in sediments in studied HF CWs was slightly higher than most data reported in the literature from constructed wetlands. The range of concentrations found in our study is comparable with concentrations found in a constructed wetland treating road runoff (Scholes et al. 1998). However, the highest values found in our study were lower than concentrations reported by Gschlössl and Stuible (2000) from Germany.

Zinc

Under aerobic conditions zinc is commonly associated with Fe and Mn oxides, hydroxides and oxyhydroxides (Krauskopf 1956; Jenne 1968; Ferris et al. 1989; Bostick et al. 2001). Zinc is also retained in iron plaques on plant root surface (Otte et al. 1995). Under anoxic conditions zinc forms very insoluble sulfides (Huerta-Diaz et al. 1993; Achterberg et al. 1997; Stumm and Morgan 1981; Kosolapov et al. 2004) and carbonates Hansel et al. 2001; Bostick et al. 2001).

The concentration of zinc in sediments of monitored constructed wetlands varied widely between 1.0 and 1,768 mg/kg (Fig. 3). In Příbraz, Slavošovice and Spálené Poříčí significantly more zinc was found in the top layer. In most surveyed constructed wetlands significantly more Zn was found in the inflow zone. The extremely high concentrations of Zn in sediments in Mořina are influenced by naturally high inflow Zn concentrations (Kröpfelová et al. 2009). Very high accumulation of zinc in the inflow zone of HF CWs was also reported by Lesage et al. (2007a, b) from systems in Zemst and Zevergem in Belgium and by Vymazal and Krása (2003) from the HF CW in the Czech Republic. Zinc concentrations

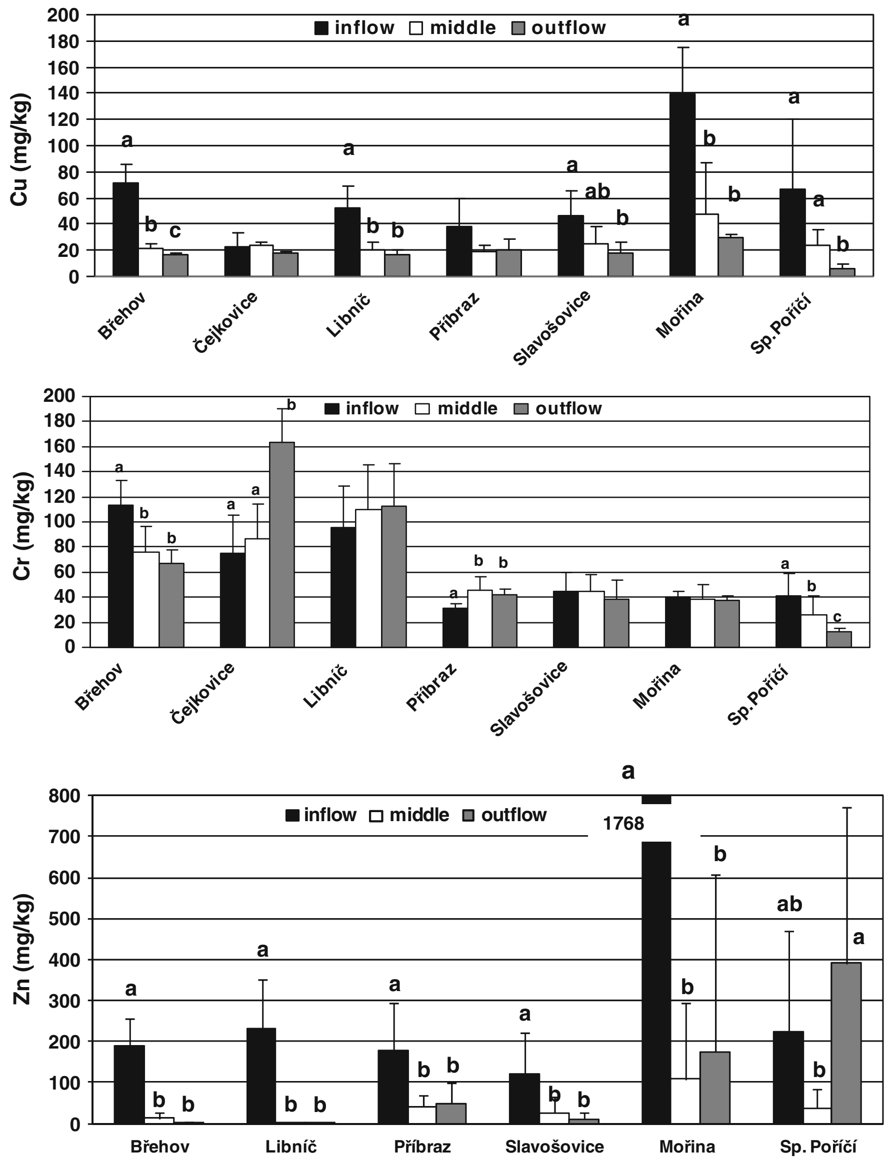

Fig. 3 Concentration of copper, chromium and zinc in seven horizontal subsurface flow constructed wetlands. *Different letters* indicate significant difference at $\alpha = 0.05$ between the means. *Bars without letters* are not significantly different

found in our study were comparable with higher Zn concentrations reported from natural unpolluted wetlands and with lower range of concentrations reported from polluted wetlands (Table 7). Zinc concentrations found in our study has never reached concentrations reported from wetlands impacted by mining activity (Table 7). In comparison with the results reported from various constructed wetlands the measured Zn concentrations are within the same range with the exception of Zn concentrations reported from a constructed wetland treating mining waters from Pb/Zn mine in China (Table 7).

In Table 8, average concentrations of studied heavy metals in seven HF constructed wetlands are shown. The data could be compared with background values and legal limits (Table 9). The data indicate that concentrations of Cd exceeded the Czech limits for light soils in Mořina and Příbraz but in general the concentrations were only slightly elevated as compared to concentrations found in unpolluted soils and sediments (Bowen 1979). Also concentrations of nickel were only slightly elevated as compared to unpolluted soils and sediments and only in Čejkovice the average Ni concentration exceeded the Czech limit for other soils.

Table 6 Concentration of chromium (mg/kg) in sediments of natural and constructed wetlands

Location	Type of sediment	Concentration	Reference
Natural wetlands, polluted			
Estonia	64 mires	3.1	Orru and Orru (2006)
Italy	Volcanic lake	2.56–5.15	Baldantoni et al. (2009)
South Carolina, USA	Unpolluted sediments	8.7	Babcock et al. (1983)
Turkey	Polluted marsh	15.3	Aksoy et al. (2005)
Zambia	Unpolluted wetland	15–23	Von der Heyden and New (2004)
Canada	Man-made lake marsh	5.0–24.9	Murdoch and Capobianco (1978)
Hong Kong	Natural marsh	10.5–46.1	Liang and Wong (2003)
UK	Unpolluted wetland	7–71	Scholes et al. (1998)
Worldwide	Estuaries and deltas	11–288	Accornero et al. (2008)
Natural wetlands, polluted			
Poland	Two lakes near copper smelter/ore processing plant	10–17	Samecka-Cymerman and Kempers (2004)
Turkey	Lake sediments impacted by urbanization	13.1–22.5	Duman et al. (2007)
Slovenia	Lake contaminated with ash from lignite coal	18.1–82.3	Mazej and Germ (2009)
Gaza Strip	Slightly polluted wetland	32–117	Shomar et al. (2005)
USA	Contaminated urban lake	33–118	DeLaune et al. (1989)
Vietnam	Water spinach cultivation sediments amended with wastewater	68–122	Marcussen et al. (2008)
Zambia	Wetland affected by mine waters	9–130	Von der Heyden and New (2004)
Poland	Anthropogenic lakes (former open cut brown coal mines)	20–165	Samecka-Cymerman and Kempers (2001)
Uganda	Lake shore affected by mining	120–169	Lwanga et al. (2003)
China	Coastal wetlands with various degree of pollution	13.5–191	Li et al. (2007)
Mexico	Lagoon affected by industrial wastes	16.1–1,039	Carranza-Álvarez et al. (2008)
Constructed wetlands			
Belgium	VF CW, 4 years in operation	17	Lesage et al. (2007a)
Belgium	HF CW, 4 years in operation	29–41	Lesage et al. (2007a)
Belgium	HF CW, 3 years in operation	26–41	Lesage et al. (2007b)
Poland	VF CW, 5 years in operation	38–41	Obarska-Pempkowiak and Klimkowska (1999)
Italy	FWS CW	12–43	Mattiuzzo et al. (2007)
Italy	FWS CW	46–71	Bragato et al. (2006)
Belgium	FWS CW, 16 years in operation	20–140	Lesage (2006)
UK	2 FWS CWs treating urban runoff	3–167	Scholes et al. (1998)
Germany	HF CW, 10 years of operation	10–259	Gschlössl and Stuible (2000)

For details see Table 2

Concentrations of lead were very low and comparable with unpolluted soils and sediments (Bowen 1979). Also for Cu, concentrations in the sediments were quite low and only in Mořina the average value exceeded slightly the Czech limit for light soils. Concentrations of Cr exceeded slightly the Czech limit for light soils in Čejkovice and Libníč, otherwise the concentrations were low and comparable with unpolluted soils and sediments. Concentrations of zinc showed the greatest variation among studied constructed wetlands. In Mořina and Spálené Poříčí the average values exceeded the Czech limits for other soils.

Table 7 Concentration of zinc (mg/kg) in sediments of natural and constructed wetlands

Location	Type of sediment	Concentration	Reference
Natural wetlands, unpolluted			
Estonia	64 mires	10	Orru and Orru (2006)
Russia	Unimpacted West Siberia tundra wetlands	24	Zhulidov et al. (1997)
UK	Unpolluted wetland	23–50	Scholes et al. (1998)
Louisiana, USA	Freshwater marsh	54.5	Fejitel et al. (1988)
Italy	Lake shore	50–103	Baudo et al. (1985)
Spain	Unaffected marsh	104	Madejón et al. (2006)
Czech Republic	Fishponds	118	Pokorný et al. (1999)
Ontario, Canada	Lake marsh	29–123	Murdoch and Capobianco (1978)
Zambia	Unpolluted wetland	1–138	Von der Heyden and New (2004)
Italy	Volcanic lake	119–149	Baldantoni et al. (2009)
Hong Kong	Natural marsh	56–328	Liang and Wong (2003)
Worldwide	Estuaries and deltas	47–649	Accornero et al. (2008)
Belgium	Freshwater tidal marshes	531–1144	Teuchies et al. (2008)
Natural wetlands, polluted			
Turkey	Polluted marsh	39.6	Aksoy et al. (2005)
Turkey	Lake sediments impacted by urbanization	39–75	Duman et al. (2007)
Uganda	Lake shore affected by mining	87.8–96.3	Lwanga et al. (2003)
Zambia	Wetland affected by mine waters	7–125	Von der Heyden and New (2004)
Poland	Anthropogenic lakes (former open cut brown coal mines)	10–131	Samecka-Cymerman and Kempers (2001)
Gaza Strip	Slightly polluted wetland	14.7–140	Shomar et al. (2005)
Vietnam	Water spinach cultivation sediments amended with wastewater	99–189	Marcussen et al. (2008)
Slovenia	Anthropogenic lake contaminated with ash from lignite coal	72–197	Mazej and Germ (2009)
China	Polluted lakes sediment	54–236	Yang et al. (2007)
Russia	Impacted West Siberia tundra wetlands	294	Zhulidov et al. (1997)
Ontario, Canada	Near Cu smelter	343	Taylor and Crowder (1983)
China	Coastal wetlands with various degree of pollution	121–478	Li et al. (2007)
China	Impacted wetland	540	Bi et al. (2007)
New Jersey, USA	Freshwater tidal marsh receiving urban runoff	310–669	Simpson et al. (1983)
China	River sediments contaminated with e-wastes	51–628	Wong et al. (2007)
Spain	Marsh affected by tailing spill	718	Madejón et al. (2006)
UK	Natural wetland receiving highway runoff	583–742	Mungur et al. (1994)
China	Paddy fields	182–857	Deng et al. (2006)
China	Sediments contaminated with municipal and industrial wastewaters	23.5–1,080	Peng et al. (2008)
Poland	Three lakes receiving wastewater	475–1,100	Szymanowska et al. (1999)
USA	Contaminated urban lake	113–1,340	DeLaune et al. (1989)
Poland	Two lakes near copper smelter/ore processing plant	1070–1,860	Samecka-Cymerman and Kempers (2004)

Table 7 continued

Location	Type of sediment	Concentration	Reference
China	Wetlands receiving wastewaters	713–4,805	Deng et al. (2004)
China	Wetlands affected by mining activity and smelters	564–8,427	Deng et al. (2006)
China	Stream near Pb/Zn mine	1,500–9,000	Deng et al. (2008)
Spain	Saltmarsh affected by mining wastes	62,280	Jiménez-Cárceles et al. (2008)
Constructed wetlands			
Italy	FWS CW, non-point pollution	26–103	Mattiuzzo et al. (2007)
Italy	FWS CW, non-point pollution	83–108	Bragato et al. (2006)
Poland	VF CW, 5 years in operation	118–130	Obarska-Pempkowiak and Klimkowska (1999)
Poland	FWS CW	71–131	Samecka-Cymerman et al. (2004)
Belgium	HF CW, 4 years in operation	181–355	Lesage et al. (2007a)
Germany	HF CW, 10 years of operation	13–490	Gschlössl and Stuible (2000)
Poland	HF CW	205–510	Samecka-Cymerman et al. (2004)
Belgium	VF CW, 4 years in operation	662	Lesage et al. (2007a)
UK	2 FWS CWs treating urban runoff	21–830	Scholes et al. (1998)
Belgium	HF CW, 3 years in operation	65–934	Lesage et al. (2007b)
Belgium	FWS CW, 16 years in operation	157–1,139	Lesage (2006)
Czech Republic	4 HF CWs, 2–5 years in operation	33.5–1,453	Zuidervaart (1996)
Czech Republic	HF CW	273 (73–1,986)	Vymazal and Krása (2003)
China	FWS CW for Pb/Zn mine	4,729–6,863	Lan et al. (1990)

For details see Table 2

Table 8 Average concentrations (mg/kg, SD in parentheses) of studied heavy metals in sediments of monitored constructed wetlands ($n = 18$)

	Cd	Ni	Pb	Cu	Cr	Zn
Čejkovice	0.17(0.10)	**<u>83</u>**(29)	14.4(14)	22(6.9)	**107**(46)	
Libníč	0.33(0.25)	53(14)	17.7(8.0)	30(18)	**107**(32)	78(126)
Břehov	0.21(0.13)	35(12)	19.0(5.7)	36(36)	85(25)	67(92)
Slavošovice	0.17(0.10)	32(17)	12.7(4.8)	29(18)	43(14)	53(74)
Mořina	**0.56**(0.63)	40(4.6)	53.1(51)	**72**(56)	39(7.2)	**<u>684</u>**(827)
Příbraz	**0.59**(0.23)	21(4.6)	23.8(6.1)	26(15)	40(6.7)	90(93)
Sp. Poříčí	0.23(0.26)	20(16)	16.7(13)	32(39)	27(17)	**<u>218</u>**(179)

Values exceeding Czech limits for light soils in bold, values exceeding limits for other soils bold and underlined (for limits see Table 9)

The results did not show any relationship between the concentration of heavy metals and the time of operation. This is probably a consequence of the sediment build-up in the filtration beds where the sediments are also formed by suspended solids. Haberl and Perfler (1990) documented that concentration of Zn, Cu and Cd remained steady during the 7-year study. While the concentrations do not change substantially during the course of constructed wetland operation, due to increase in the sediment biomass the amount of heavy metals increases. This was also observed in our study.

Concentrations of sediment in the filtration beds

For constructed wetlands in the Czech Republic, washed gravel or crushed stones are used. In the beginning of operation, the amount of sediment is zero and its concentration increases during the time of operation. In Table 10, concentrations of sediment expressed in %DM of the filtration bed material are shown. The results indicate the increase of sediment concentration with increasing time of operation. The amount of sediment was usually greater in the inflow zone as compared to outflow zone but the difference was not always statistically significant (Table 10). In Slavošovice, significantly more sediment mass was found at the bottom layer while in Spálené Poříčí significantly more sediment mass was found in the top layer in the inflow and middle zones. Also, in Břehov and Mořina, more sediment was found in the top layer. This variation is probably affected by the placement of the distribution pipes. While in Slavošovice the distribution pipes are buried near the bottom of the bed, in Spálené Poříčí, Břehov and Mořina, the distribution systems is either laid down on the surface of the filtration bed or it is buried only shallowly bellow the bed surface. Taking into consideration the sediment/filtration material mass ratio it was possible to calculate average heavy metal concentrations in the filtration material including sediments (Table 11). As sediment mass varied between 0.42 and 10.55% of the filtration material, the final heavy metal concentrations are much lower than legal limits (Table 9).

Table 9 Limits for heavy metals concentration (mg/kg) in soils and sediments

	Cd	Ni	Pb	Cu	Cr	Zn	Ref
Czech Republic: limit for light soils	0.4	60	100	60	100	130	1
Czech Republic: limit for other soils	1.0	80	140	100	200	200	1
Belgium: soil remediation values	2.0	100	200	200	130	600	2
China: criteria for agricultural soil quality	0.3	50	300	100	200	250	3
Netherlands: remedial intervention should be taken	12	210	530	190	380	720	4
Consensus based probable effect concentrations	4.98	48.6	128	149	111	459	5
Background values	0.8	9	40	17	37	62	2
Average concentrations in unpolluted soils	0.35	40	35	30	70	90	6
Unpolluted freshwater sediments	0.17	52	19	33	72	95	6

1: PAS (1994), 2: VLAREBO (1996), 3: National Standard of PR China (1995), 4: Department of Soil Protection, Netherlands (1994), 5: McDonald et al. (2000), 6: Bowen (1979)

Table 10 Average sediment concentration in inflow, middle and outflow parts of the filtration bed of surveyed CWs (in % DM, SD in parentheses)

	Layer	Čejkovice	Libníč	Břehov	Slavošovice	Mořina	Příbraz	Sp. Poříčí
Years of operation		2	2	5	7	8	9	16
Inflow	Top	1.0(0.5)^{ab}	0.5(0.08)^{a}	4.4(1.2)	1.7(0.5)^{a}	4.0(1.3)^{a,b}	5.9(2.8)^{a}	14.8(1.8)^{a}
	Bottom	1.2(0.02)^{a}	0.6(0.3)^{a,b}	2.7(0.5)	4.2(0.7)^{b}	3.3(1.8)^{a,b}	2.9(0.2)^{b}	7.8(1.3)^{b}
Middle	Top	1.0(0.6)^{ab}	0.3(0.03)^{b}	3.9(2.3)	0.7(0.6)^{a}	3.4(0.4)^{a}	2.0(0.4)^{b}	15.3(3.7)^{a}
	Bottom	0.5(0.2)^{b}	0.4(0.09)^{a,b}	2.5(0.4)	3.8(1.0)^{b}	1.5(0.2)^{b}	3.4(0.6)^{b}	7.7(1.9)^{b}
Outflow	Top	0.1(0.07)^{b}	0.3(0.06)^{b}	4.3(1.1)	0.4(0.1)^{a}	4.7(4.4)^{a,b}	2.4(0.5)^{b}	7.8(3.7)^{b}
	Bottom	0.1(0.06)^{b}	0.4(0.1)^{a,b}	2.1(0.5)	2.7(0.5)^{b}	1.4(0.1)^{a,b}	2.8(0.2$)^{b}$	9.9(0.2)^{b}

Different letters indicate significant difference at $\alpha = 0.05$ between the inflow ($n = 3$), middle ($n = 3$), outflow ($n = 3$), top ($n = 3$) and bottom ($n = 3$) samples

Table 11 Average concentrations (mg/kg DM, SD in parentheses) of studied heavy metals in filtration material (i.e. sediment + filtration substrate) of monitored constructed wetlands ($n = 18$)

	Cd	Ni	Pb	Cu	Cr	Zn
Čejkovice	0.0011(0.0009)	0.54(0.32)	0.09(0.02)	0.14(0.1)	0.70(0.45)	–
Libníč	0.0014(0.001)	0.22(0.11)	0.07(0.02)	0.13(0.09)	0.45(0.27)	0.33(0.71)
Břehov	0.0070(0.005)	1.16(0.55)	0.63(0.38)	1.29(1.0)	2.82(1.16)	2.22(3.42)
Slavošovice	0.0038(0.002)	0.72(0.43)	0.29(0.11)	0.65(0.50)	0.97(0.51)	1.19(1.12)
Mořina	0.0171(0.012)	1.22(0.57)	1.62(0.91)	2.20(1.12)	1.02(0.85)	20.9(22.7)
Příbraz	0.0192(0.008)	0.68(0.63)	0.77(0.55)	0.85(0.65)	1.30(1.62)	2.93(2.80)
Sp. Poříčí	0.0243(0.014)	2.11(1.31)	1.76(1.12)	3.38(2.23)	2.85(1.76)	23.0(28.6)

Conclusions

Concentrations of Cd, Ni, Pb, Cu, Cr and Zn were evaluated in seven constructed wetlands with horizontal subsurface flow treating municipal wastewater in the Czech Republic. The time of operation varied between 2 and 16 years among systems. The results revealed that concentrations of heavy metals were low and comparable with concentrations found in unpolluted or lightly polluted natural wetlands. The concentrations were much lower than concentrations found in wetlands receiving mine drainage or industrial wastewaters. The concentrations of heavy metals did not reflect the length of operation but the amount of sediment mass increases with the length of operation. This will result in greater heavy metal mass in the system. The concentrations of heavy metals in the sediment exceeded occasionally the limits for agricultural soils but when filtration material was taken into consideration, the concentrations were well below the limits.

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