

TRACE METALS ASSIMILATION IN TREATMENT WETLAND SEDIMENTS

Michael B. Anderson, Glenn D. Dombeck, P.E.* , Mark W. Perry, P.E.* ,

ABSTRACT

Sediment metals concentration data from the Sacramento Regional County Sanitation District's Constructed Wetland Demonstration Project have been analyzed for spatial and temporal trends. The sediment monitoring program covers a five-year period, starting with baseline sampling in December 1993 and extending through 1998. The principal sampling studies include core sampling in vegetated areas, a sediment partitioning study and an accretion study. Of the nine trace metals measured, six metals (silver, cadmium, copper, mercury, lead and zinc) consistently exceed apparent mass removal rates of 60 percent. Apparent concentration and mass removal rates for three trace metals (arsenic, chromium and nickel) have steadily declined over the monitoring period and currently display negative removal rates. Analysis of the sediment trace metals data indicates an apparent trend towards increasing sediment concentrations over time for arsenic, chromium, copper, nickel, and zinc. However, for the other trace metals, no consistent observable trend towards increasing sediment concentrations is observed. Analysis of wetland cell sediment samples indicates an apparent longitudinal trend toward elevated trace metals concentrations within the initial wetland cell quadrant. However, there are no statistically significant differences observed between the first and second quadrants, or the second quadrant and the second half-cell. Detrital layer trace metals concentrations are on average four times greater than the underlying mat and clay layers, a trend that is statistically significant for all metals but chromium and nickel. Of the five trace metals demonstrating an increasing trend over time, only chromium and nickel are observed in concentrations approaching or exceeding the freshwater sediment Probable Effect Levels (PEL). Chromium and nickel are identified as metals for which removal rates are low or even negative, and are the only metals for which accumulated sediment metals concentrations increase over distance. Water column speciation indicates that nickel enters the wetland in a predominantly dissolved and strongly organic-complexed form.

KEYWORDS

Sediments, trace metals, constructed wetland, metals removal, metals assimilation, mass balance, bioaccumulation, surface-flow wetland.

INTRODUCTION

The presence of trace metals in the sediments of a surface-flow treatment wetland is investigated in this paper. Sediment monitoring data observed over a five-year period beginning with baseline sampling in December 1993 prior to application of treated effluent in March 1994 are analyzed for spatial and temporal trends, and compared to available freshwater sediment guidelines to identify potential bioaccumulation hazards.

Much is known about the performance of surface-flow constructed wetlands for the treatment of municipal wastewaters (Crites *et al.*, 1998; Gearheart *et al.*, 1996; Kadlec *et al.*, 1996; Reed *et al.*, 1995); however, less is known about the long-term fate and toxicity of trace metals in the sediments of surface-flow treatment wetlands. There is a good deal of information available in the literature on the interaction of trace metals in sediments (Mitsch *et al.*, 1993; Yong, 1995). The National Oceanic and Atmospheric Administration (NOAA) publishes unofficial screening levels for inorganic and organic contaminants in freshwater sediments (NOAA, 1997). A threshold and probable effects level (TEL and PEL, respectively) are identified for each metal based on *Hyallela azteca* bioassays and the level at which adverse impacts are generally observed in the benthic community. Shacklette *et al.* (1971) report mean background concentrations of metals in soils in the Western United States lower than the NOAA PELs; however, they

*Summit Engineering, Inc.

*County of Sacramento

identify a range of background concentrations for arsenic, chromium and nickel where the upper end of the range approaches two times the PEL. Sorenson *et al.* (1991) report similar conclusions for typical bottom sediments, with mean concentrations for all metals lower than the PEL and the upper end of the range for chromium and nickel averaging 2.3 times the PEL. Kadlecik (1996) noted a trend toward decreasing volatile organic carbon concentrations with distance from the inlet of a surface flow wetland.

The five-year demonstration wetlands program was motivated by the interest in the use of constructed wetlands to polish secondary effluent for trace metals removal to meet strict effluent discharge limits proposed in the California Inland Surface Waters Plan (SWRCB, 1991). In addition to development of a comprehensive monitoring program that investigated water column, sediment, vegetation and biota, a constructed wetlands fate and transport evaluation model (CWFATE) has been developed (Jones and Stokes, 1993; Perry *et al.*, 1996). Kallin (1999) adopted the two-dimensional CWFATE model to an existing vertical one-dimensional sediment model that calculates equilibrium speciation, kinetic reaction and transport of metals in the presence of biodegradation of organic substances in saturated porous media. Kallin provides interesting insights into the role of scavenging by algae and phytoplankton, rhizosphere oxygenation and transpiration-induced flow in the fate of trace metals in wetlands.

The configuration of the wetland, operated by the Sacramento Regional County Sanitation District in Elk Grove, California, is presented in Figure 1. The project consists of ten wetland treatment cells, each with dimensions of approximately 384 m by 15 m, and an identically sized groundwater-supplied control cell. The wetland cells are constructed in two halves, with the inlet located in the B-half and the wetland effluent collected in the A-half on the same end of the cell as the inlet. A 0.8 ha habitat wetland handles the wetland cell effluent prior to discharge back to the treatment plant. The wetland cells are numbered sequentially from one to eleven from west to east and consist of five distinct process types, including batch discharge, recycle, combination overland flow/plug flow, plug flow and combination subsurface flow/plug flow. A more detailed description of the wetland system can be found in the five-year summary report (Nolte, 1999).

The wetland cells were constructed of native soils, with soil from a regional wildlife refuge imported to establish the vegetation. The dominant soil types identified at the site (USDA, 1993; Kleinfelder, 1992) are Galt clay, Clear Lake clay, Madera loam and San Joaquin silt loam. Soil permeability for the restricting layer of these soils is characterized as "very slow," less than 0.06 in/hr.

METHODOLOGY

Beginning in 1993, periodic measurements of sediment metals concentrations along the vegetated portion of the wetland profile of several treatment cells have been performed. The control cell was added to the sediment sampling program in 1996 to provide additional background reference information. Initially, sediment samples were collected by driving a 3-in PVC pipe into the wetland bottom and removing and compositing the collected material. This technique failed to provide consistent sample volumes and neglected the layer of suspended detrital material at the surface of the wetland sediments. Failure to include sediment moisture content in the analyses also made comparison of dry weight concentrations difficult.

A sediment metals partitioning study was initiated in October 1996 to establish criteria for the collection of samples from defined sample layers. Three sediment layers were identified; a top layer of suspended detritus (A layer), a more consolidated layer of vegetative mat (B layer), and the clay layer underlying the vegetative mat (C layer). Sediment samples were consistently collected at identical depths so that the volume of material collected in the C layer was relatively consistent. In a separate study, the accretion over time of material within 250-mL glass jars placed in vegetated areas of a treatment cell was investigated. Sediment collection jars were placed at the bottom of the cell at locations 91 m and 277 m from the wetland cell inlet.

RESULTS

Apparent concentration and mass removal rates observed within the water column from 1994 to 1998 are presented in Table 1 (Nolte, 1999). Of the nine trace metals measured, four metals (silver, mercury, lead and zinc) consistently exceed apparent mass removal rates of 70 percent, while two more (cadmium and copper) generally exceed apparent mass removal rates of 60 percent. Apparent concentration and mass removal rates for three trace metals, arsenic, chromium and nickel, have steadily declined over the monitoring period, and currently display negative removal rates. Analysis of hydraulic conditions to factor out the effects of evapoconcentration, infiltration and vegetation harvest has been performed elsewhere (Dombeck *et al.*, 1998). Seasonal effects of evaporation, precipitation and infiltration on trace metals mass removal can be significant and combined may contribute to as much as a 30 percent deviation from the average annual removal rate. Annual vegetation harvest accounts for less than five percent of annual trace metals removal.

Average trace metals concentrations in wetland sediment from 1993 to 1998 are presented in Table 2. Out of the nine metals tested, analysis of the sediment trace metals data indicates an apparent trend towards increasing sediment concentrations over time for arsenic, chromium, copper, nickel and zinc. The other trace metals (silver, cadmium, lead and mercury) demonstrate no consistent observable trend towards increasing sediment concentrations. The general trend observed for the average trace metals concentrations presented in Table 2 is an initial decrease in concentration, followed by a steady rise, ending with a final concentration decrease.

The NOAA freshwater sediment probable effects levels (PELs) also are presented in Table 2, along with the ratio of the most recent (1998) sediment concentrations to the PELs. For all but two metals, the 1998/PEL ratio is below 0.3, signifying that the sediment metals concentration is less than one-third of the PEL. However, the 1998 average chromium concentration approaches the PEL, and the 1998 nickel concentration exceeds the PEL. Average sediment chromium concentrations also exceeded the PEL in 1997 and average sediment nickel concentrations have done so since 1995. Interestingly, chromium and nickel are the only two metals for which exceedences of the PEL have been observed in the control cell, a trend that was observed to occur almost simultaneously to the exceedences of the PEL observed in the treatment cells. The 1998 PEL ratios have also been calculated separately for the control cell. For all of the metals except arsenic, chromium, and nickel, the control cell PEL ratio is lower than the PEL ratio calculated for all cells.

The results of the sediment accretion study are presented in Table 3. Accumulated sediment at both the upstream and downstream sediment collection jar locations was analyzed for five trace metals. Of the five metals tested, a decrease in longitudinal sediment trace metals concentrations was consistently observed for three metals (copper, lead and zinc). However, for chromium and nickel, a slight increase in longitudinal sediment metals concentrations was generally observed. Trace metals concentrations for accreted sediment (samples taken from sediment collection jars) lie within the range of concentrations observed for the partition study sediment (samples collected from the wetland cell bottom).

DISCUSSION

Analysis of 1997 wetland cell sediment samples indicates an apparent longitudinal trend toward elevated trace metals concentrations within the initial wetland cell quadrant. However, there are no statistically significant differences observed between the first and second quadrants, or the second quadrant and the second half-cell, using a single-factor ANOVA at the 95 percent confidence level. Detrital layer trace metals concentrations are on average four times greater than the underlying mat and clay layers, a trend that is statistically significant for all metals but chromium and nickel.

Additional insight into the nature of chromium and nickel behavior within wetland sediments is provided by water column speciation investigations conducted in fall 1997 (Nolte *et al.*, 1998). Nickel enters the wetland in the secondary effluent in a predominantly dissolved form (5-year average of 83 percent dissolved fraction as reported in Nolte, 1999). Approximately 55 percent of the dissolved fraction was identified in three speciation study sampling events to be chelated by an organic ligand, with the remainder

as either free metal ion or inorganically complexed. The percentage of organically complexed nickel did not change from influent to effluent, suggesting that the nickel is strongly bound to the organic ligand. The presence of a dissolved strong organic nickel complex, likely a synthetic complex, has been observed in other wastewaters (Sedlak *et al.*, 1997) and may contribute to the poor water-column removal of this substance, as the rate of dissociation of nickel from ligands is slow, on the order of years (Phinney, 1997). The observation during the speciation study of effluent particulate nickel concentrations greater than the influent concentrations may suggest that resuspension of nickel from the sediments into the water column is occurring, which could also contribute to the low nickel removal.

Chromium also enters the wetland in a predominantly dissolved form (5-year average of 52 percent dissolved fraction as reported in Nolte, 1999, 88 percent dissolved fraction in speciation study sample). Of this dissolved fraction, approximately 55 percent was determined to be Cr (VI). Cr (VI) is soluble in oxygenated water and is considered to be the form that is most bioavailable. The chromium undergoes reduction in the anoxic environment from Cr (VI) to Cr (III), which readily binds to organic particles in the wetland sediments. The authors speculate that metal-binding adsorption sites become limited in the wetland over time, resulting in competition for sorption sites. Hence, chromium is likely displaced from available sediment binding sites by metals with stronger binding affinity, such as zinc, copper, lead or mercury (Dzombak *et al.*, 1990 for ferric oxides). This would help explain the decreasing ability to remove these metals from the water column, as evidenced by the declining mass removal rates. Analysis of cation exchange capacity (CEC) supports this assumption, as average treatment cell CECs appear to decline from approximately 30 to 20 meq/100 g during the initial three project years (Table 4). This theory may also address the sediment release and poor removal of the available nickel (free ion, inorganically and weakly complexed nickel), which has an even lower level of binding affinity to oxides than chromium.

The relatively low levels of arsenic removal from the water column (Table 1) may be explained in part by Kallin's theory of the role of wetland plant rhizomes in controlling the redox potential of the sediments through diffusion of oxygen in the root zone (Kallin, 1999). It is speculated that oxygen diffusion in microenvironments within vegetated areas results in higher levels of As (V) sorbed to manganese and ferric oxides in the upper oxic sediment layer which precipitate out of the water column. As these iron oxide-metal complexes are buried within lower anoxic sediment layers they become reduced and release As (III) to the overlying soil-water column. Although no data on sediment pE profiles, oxygen diffusion rates, or sediment porewater arsenic or iron concentrations are available to confirm this theory, high concentrations of arsenic are observed in the detrital A layer and a trend towards both decreasing water column removal and sediment concentrations over time is observed.

Benthic invertebrates were sampled from the wetland cells in fall 1993, fall 1996 and spring 1997. The 1993 baseline data indicated that the constructed wetlands had been colonized by a diverse invertebrate community typical of a wetland environment (*Cladocera*, *Copepoda*, *Podocopa*, *Oligochaeta*, *Chironomidae* and *Gastropoda*). The 1996 and 1997 sampling indicated dissimilar invertebrate population abundance and diversity between the treatment cells and the control cell, but also indicated a similar pollution tolerance index among the identified taxa, including the presence of species with low pollution tolerance in both communities. Invertebrate tissue samples analyzed for trace metals in 1997 and 1998 indicated no significant differences between the inlet and outlet tissue concentrations and lower tissue concentrations for five of seven metals in the wetland cells in comparison with an adjacent natural wetland that receives urban runoff (Nolte, 1999).

CONCLUSIONS

Interpretation of the sediment trace metals data over the entire sampling period is complicated by inconsistencies in sample depths, lack of percent moisture analysis for early samples, lack of detrital (A layer) collection prior to 1996 and high variance in the data.

Out of the nine metals tested, analysis of the sediment trace metals data indicates an apparent trend towards increasing sediment concentrations over time for arsenic, chromium, copper, nickel and zinc. The other trace metals (silver, cadmium, lead and mercury) demonstrate no consistent observable trend towards increasing sediment concentrations. Of the five trace metals demonstrating an increasing trend, only

chromium and nickel are observed in concentrations approaching or exceeding the freshwater sediment PELs (Table 2). Chromium and nickel are identified as metals for which water column removal rates are low or even negative (Table 1), and as the only metals for which accreted sediment metals concentrations increase over distance (Table 3). Because the trend towards relatively high chromium and nickel sediment concentrations is mirrored in the control cell (groundwater) sediments, it is possible that elevated sediment concentrations result from background water column concentrations of chromium and nickel and are of little concern with respect to biohazards at the current levels. Invertebrate analysis does not indicate trace metals concentrations in treatment wetland benthic populations greater than that observed in surrounding natural wetlands.

Over the five-year program, the surface-flow treatment wetland cells have demonstrated consistently high removal rates for most trace metals in secondary municipal effluent, although declining removal rates are observed for arsenic, chromium and nickel. Based on the observations of sediment trace metals concentrations, benthic invertebrate data, observations of vegetation and biota, and comparisons with other wetlands, the treatment wetlands appear to remove trace metals effectively from the water column, where they become assimilated into the bottom sediments at levels below those that could contribute to adverse effects among wetland biota.

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Glenn Dombeck is a Senior Engineer with Summit Engineering, Inc. in Santa Rosa and was formerly project manager of the wetlands project at Nolte and Associates. Michael Anderson is a project engineer at Nolte and Associates. Mark Perry is currently an Associate Engineer with the Sacramento County Regional Sanitation District's Wastewater Collections section of the Water Quality Division and was formerly project engineer responsible for coordination of the Sacramento Demonstration Constructed Wetlands.

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Figure 1 - Layout of Sacramento Constructed Wetlands Demonstration Project

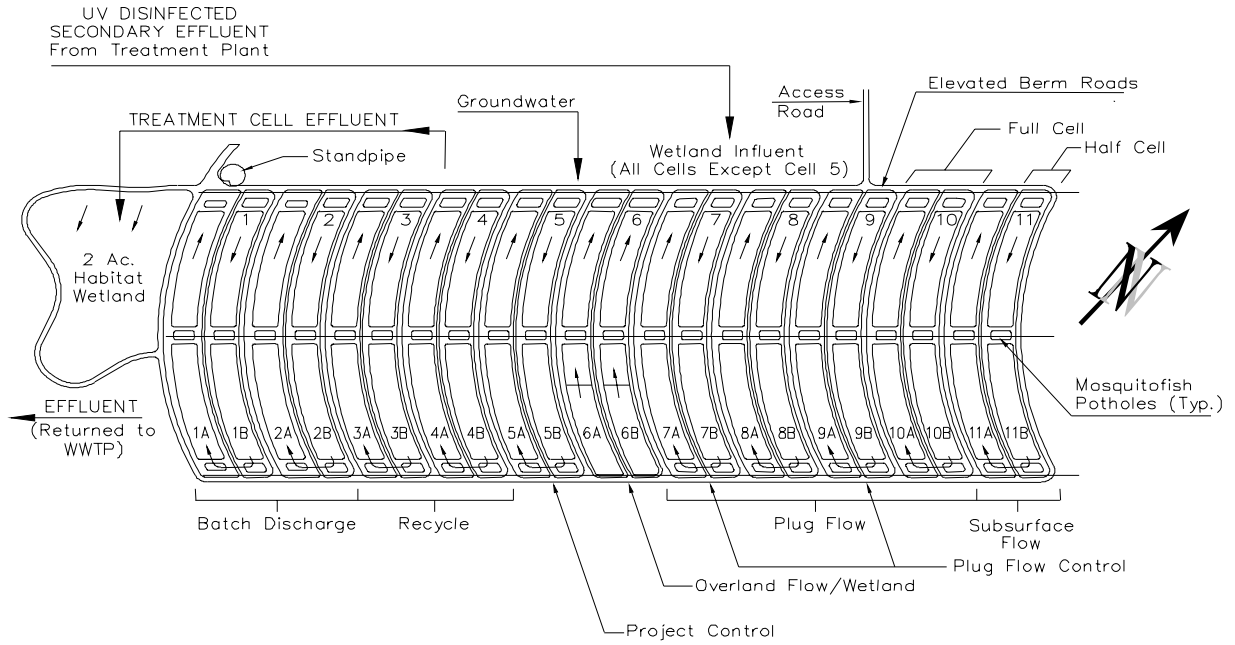


Table 1 – Apparent Concentrations and Mass Removals for Wetland Trace Metals for 1994 to 1998^a

Metal	Apparent Concentration Removal, percent^b					Apparent Mass Removal, percent^c				
	1994	1995	1996	1997	1998	1994	1995	1996	1997	1998
Silver	58	85	88	96	91	73	91	92	97	93
Arsenic	18	-7	-44	-19	-62	47	31	0	23	-32
Cadmium	40	60	56	76	48	61	74	69	84	58
Chromium	42	53	-37	-8	-5	63	70	5	30	15
Copper	64	52	51	58	65	77	69	66	73	72
Mercury	62	65	60	61	66	75	78	72	75	73
Nickel	-5	5	-23	-32	-32	32	39	15	15	-7
Lead	84	86	75	67	72	89	91	82	78	77
Zinc	78	91	80	82	76	86	94	86	89	81

^a Source: Nolte and Associates, Inc., *et al.* June 1999, Sacramento Constructed Wetlands Demonstration Project Five-Year Summary Report, 1994-1998.

^b Apparent concentration removal based on the difference between inlet and outlet concentrations (does not account for effects of infiltration, evapoconcentration, or harvest).

^c Apparent mass removal based on measured constituent concentrations, average inflow of approximately 15.9 m³/hr (70 gal/min), and average outflow of approximately 10.2 m³/hr (45 gal/min). (Average inflow and outflow values vary slightly from year to year.)

Table 2 – Average Concentrations and PEL Ratios for Trace Metals in Sediment From Wetland Treatment Cells for 1993 to 1998

Metal	Average sediment concentration, mg/kg dry weight							NOAA PEL ^h	1998 PEL Ratio ⁱ	
	1993 ^a	1994 ^b	1995 ^c	1996 ^d	1996 ^e	1997 ^f	1998 ^g		All Cells	Control
Silver	2.18	0.01	NA	0.06	NA	0.84	0.45	--	NA	NA
Arsenic	1.16	0.13	2.98	2.98	NA	6.35	4.60	17	0.27	0.68
Cadmium	1.02	0.05	0.08	0.97	NA	0.27	0.14	3.53	0.04	0.03
Chromium	20.8	16.2	55.0	55.5	59.9	179.6	65.5	90	0.73	0.85
Copper	19.6	14.9	23.4	30.1	35.7	51.8	34.9	197	0.18	0.14
Mercury	0.030	NA	NA	0.020	NA	0.051	0.024	0.486	0.05	0.04
Nickel	27.4	19.9	49.9	44.4	50.4	116.3	53.4	35.9	1.49	1.53
Lead	12.10	7.70	7.70	6.20	12.60	12.90	9.20	91.3	0.10	0.10
Zinc	23.8	16.2	40.5	38.9	114.9	134.9	87.5	315	0.28	0.15

^a Average of 12/9/93 bulk samples from cells 1, 3, 6, 7, and 9 adjusted to dry weight with estimated moisture content of 0.29.

^b Average of 9/6/94 bulk samples from cells 1, 3, 7, and 9 adjusted to dry weight with estimated moisture content of 0.29.

^c Average of 7/24/95 bulk samples from cells 1, 3, 6, 7, 9, and 11.

^d Average of 7/18/96 bulk samples from cells 3, 6, 7, 9, and 11.

^e Average of 10/24/96 partitioning study samples (three layer average) from cells 4 and 7.

^f Average of 8/7/97 partitioning study samples (three layer average) from cells 4, 6, and 7.

^g Average of 8/10/98 partitioning study samples (three layer average) from cells 4, 6, and 7.

^h National Oceanic and Atmospheric Administration compilation of probable effects levels for benthic organisms within freshwater sediments.

ⁱ PEL ratio equals average sediment metals concentration divided by Probable Effects Limit. Control wetland cell supplied by groundwater.

Table 3 – Average Concentrations for Trace Metals in Wetland Sediment Accumulation Study

Metal	Avg. concentration, mg/kg dry weight^a	
	1997	1998
Chromium	32	69
	57	59
Copper	18	47
	17	28
Nickel	27	39
	48	45
Lead	9	23
	6	13
Zinc	151	313
	76	87

^a For each metal, upstream (91 m from inlet) and downstream (277 m from inlet) concentrations are provided in the upper and lower row, respectively.

Table 4 – Average CEC Values in Wetland Sediment Samples for 1994 to 1996^a

Cell	Average CEC concentration, meq/100 g		
	1994	1995	1996
1	9	15	-
3	31	23	18
5	-	30	23
6	26	14	21
7	49	31	20
9	35	22	18
11	-	17	20
Avg.	30	22	20
Max.	49	31	23
Min.	9	14	18

^a A total of 33, 62, and 94 samples were collected in 1994, 1995, and 1996, respectively.