

Distribution of Heavy Metals in Soils of Shihwa Tidal Freshwater Marshes

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ABSTRACT : Shihwa tidal freshwater marsh was constructed recently to treat pollutants entering Shihwa lake. In this study, we examined the spatial and temporal patterns of heavy metal accumulation in soils of Shihwa marsh and sought correlations between several soil variables (pH, electrical conductivity, organic matter, and acid ammonium oxalate-extractable Fe and Al contents) and the heavy metal concentration of soils. Surface soil samples (0~20 cm) were collected in June 2000, November 2000, and July 2001, and were analyzed for heavy metals (Zn, Cd, Pb, Cu, Cr, As, and Hg) and soil chemical properties. The neutral pH and water-saturated conditions of Shihwa marsh appeared to favor immobilization of heavy metal through adsorption onto soils. The concentrations of heavy metal (especially Zn, Cu, and Cr) in soils of Shihwa marsh increased along the sampling occasions, suggesting that soils of Shihwa marsh serve as a sink of heavy metal. Among the sub-marshes, metal concentrations were highest in Banweol high marshes and lowest in Samhwa marshes. The temporal and spatial variations in the heavy metal concentrations of soils were correlated positively with organic matter and oxalate extractable Fe and Al contents, but negatively with electrical conductivity. These results suggest that organic matter and hydrous oxide of Fe/Al may play a key role in removing heavy metals in soils of Shihwa marsh, and that heavy metal removing capacity would increase with desalinization. However, the removal patterns of heavy metal by reeds warrant further studies to evaluate the total removal capacity of heavy metals by Shihwa marsh.

Key words: Fe and Al oxides, Heavy metal adsorption, Organic matter, Salinity, Shihwa tidal freshwater marsh.

INTRODUCTION

Since constructed wetlands are an inexpensive system for wastewater treatment, they have been used to remove organic substances and nutrients from municipal sewage, storm water, and agricultural runoff (Rodgers and Dunn 1992, Lakatos *et al.* 1997). In Korea, Shihwa tidal freshwater marsh was constructed recently on a tideland located in Ansan (126° 50 'N, 37° 17 'E) to treat pollutants, such as organic pollutants, nitrogen, phosphorous, and heavy metal, before entering into newly constructed Shihwa lake. Several studies have been conducted to examine removal pattern of N and P by reeds planted in the marsh (Ro *et al.* 2002), N removal by denitrification (Kim 2002), and P removal by adsorption onto soils (Ryu 2002). However, research on the removal of heavy metal by Shihwa marsh system is very few. Since stream water polluted with heavy metal from industrial activities such as chemical manufacturing, oil refining, and metal processing and plating flows into Shihwa marsh (Hyun *et al.*

1999), removal of heavy metal by the marsh system would play an important role in preventing Shihwa lake from heavy metal contamination.

The removal of heavy metal by plant uptake has been studied for landfill sites (Seo and Cho 2002) and wetlands (Karpiscak *et al.* 2001, Stoltz and Greger 2002). Mays and Edwards (2001) and Cheng *et al.* (2002) reported that wetland soils also contributed to the removal of heavy metal in part. Heavy metals are removed from wetland water through adsorption onto soil by ion exchange, precipitation or co-precipitation, and chelation. The process of adsorption/desorption is controlled by physical (texture and type of clay mineral) and chemical (pH, redox potential, salinity, organic matter, carbonates and sulfides) properties (Gambrell 1994). Therefore, information on the effects of these variables on the adsorption/desorption of heavy metals in soils would be of value for the optimal management of Shihwa marsh as a sink of heavy metals.

In this study, we examined the spatial and temporal patterns of

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heavy metal accumulation in soils of Shihwa marsh and evaluated the effects of several soil variables (pH, electrical conductivity, organic matter, and acid ammonium oxalate-extractable Fe and Al) on the heavy metal concentrations of soils by a correlation analysis.

MATERIALS AND METHODS

Study site

Shihwa marsh is one of the largest wetlands in Korea, encompassing about 750,000 m². This marsh is a kind of surface flow one, and receives waters mainly from three streams: Banweol, Donghwa, and Samhwa streams (Fig. 1). The soil texture of the basin was silty clay loam, and *Phragmites australis* were transplanted for water decontamination. In Banweol and Donghwa stream areas, the marshes are divided into low and high marshes based on the height above sea level. As such, Shihwa marsh is consisted of five sub-marshes: Banweol high (B_H, 190,063 m²) and low (B_L, 225, 889 m²), Donghwa high (D_H, 152,500 m²) and low (D_L, 113,499 m²), and Samhwa (S, 68,672 m²) marshes (Ro *et al.* 2002).

Sampling and analysis

At each sub-marsh, two (for S) or four (for B_H, B_L, D_H, and D_L) stations were established for the collection of soil (Fig. 1). Five surface soil samples (0–20 cm) were collected from sampling plots (2.0 × 2.0 m²) established at each station in June 2000, November 2000, and July 2001. A composite soil sample was made using five sub-samples taken individually. At each sampling occasion, a new sampling plot was established within a cir-

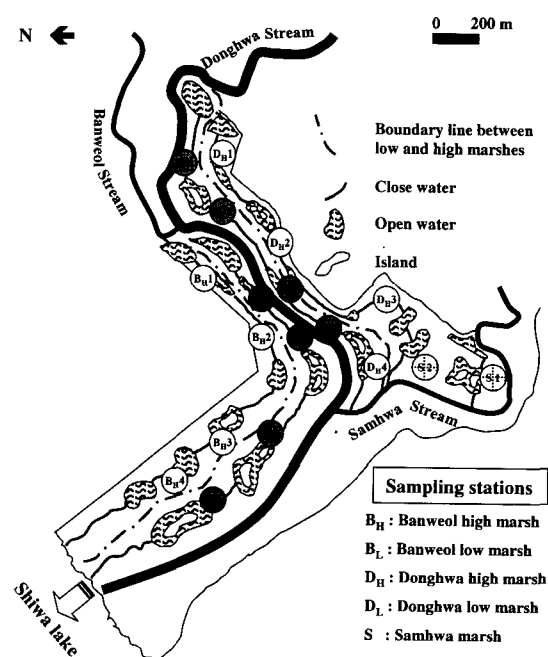


Fig. 1. Study site map.

cle of 30 m radius. Soil samples were air-dried, passed through a 2-mm sieve, and used for chemical analyses.

Soil pH (1:5) and electrical conductivity (1:5) were determined using a pH meter (DMP2000, MDS, Korea) and a conductivity meter (PW9505, Philips, UK). Soil organic matter was determined using a CN analyzer (IsoPrime-EA, Micromas, UK). Acid ammonium oxalate-extractable Al and Fe, which represent the amount of active Al and Fe oxides, were determined by the

Table 1. Variations in pH, electrical conductivity, organic matter, and oxalate extractable Fe and Al contents of soils

Soil characteristics	Sampling time	Sites ^a																	
		B _H 1	B _H 2	B _H 3	B _H 4	B _L 1	B _L 2	B _L 3	B _L 4	D _H 1	D _H 2	D _H 3	D _H 4	D _L 1	D _L 2	D _L 3	D _L 4	S1	S2
pH (1:5)	June-00	7.3	6.7	7.6	7.7	7.8	7.5	7.6	7.8	6.6	7.0	7.3	7.2	6.7	7.2	6.6	6.9	7.3	6.9
	Nov-00	7.2	6.6	7.9	6.8	7.4	8.0	5.6	7.3	7.6	7.5	7.7	7.3	6.6	6.5	6.9	6.1	7.6	6.9
	Jul-01	7.6	7.5	6.7	7.0	6.4	7.4	6.0	7.3	6.6	6.2	7.3	7.2	5.5	6.1	6.9	6.0	6.0	6.8
Electrical conductivity (1:5, dS m ⁻¹)	June-00	1.0	1.7	1.7	1.7	1.5	1.5	1.4	1.4	2.2	3.1	1.9	2.3	1.4	1.7	2.4	2.0	2.1	2.1
	Nov-00	0.3	1.4	0.6	0.4	1.1	0.8	1.0	0.8	0.9	0.7	0.5	0.5	0.9	1.0	0.1	1.4	1.1	1.4
	Jul-01	0.2	0.2	0.3	0.3	0.7	0.3	1.1	0.9	0.5	0.7	0.6	0.7	0.4	0.4	0.3	0.7	0.7	1.4
Organic matter (g kg ⁻¹)	June-00	17.1	10.9	10.9	10.0	11.7	12.6	9.9	8.6	12.3	8.9	5.9	1.7	9.6	8.9	9.1	9.5	12.0	9.3
	Nov-00	10.5	11.3	10.0	10.7	12.2	9.8	11.2	9.3	9.3	9.2	6.0	6.5	10.0	11.6	9.4	11.9	12.7	12.2
	Jul-01	16.4	12.5	13.0	12.4	15.3	11.4	14.4	11.7	13.9	10.4	8.3	7.0	13.6	14.0	9.9	13.5	13.1	7.1
Oxalate extractable Fe (mg kg ⁻¹)	June-00	4846	4690	5610	3209	5294	3848	3284	3286	4328	3843	2820	2793	3150	2907	3404	3635	3307	2374
	Nov-00	4975	4725	5589	3317	4967	3927	3521	3348	4281	3979	2933	2649	3167	2879	3384	3573	3419	2198
	Jul-01	5324	4974	5681	3599	5328	3996	3308	3299	4497	3924	2756	2741	3328	3107	3549	3769	3518	2485
Oxalate extractable Al (mg kg ⁻¹)	June-00	1695	1668	2113	1150	1864	1426	1338	1162	1260	1394	1134	1060	1162	1168	1173	1314	1211	1003
	Nov-00	1933	1524	1625	1328	1577	1019	1957	1208	1420	1156	989	1197	1092	1479	1287	1170	1208	1097
	Jul-01	1824	1827	1893	1427	1792	1548	1401	1284	1550	1297	1161	1202	1211	1397	1407	1146	1182	1175

^a B_H, Banweol high marsh; B_L, Banweol low marsh; D_H, Donghwa high marsh; D_L, Donghwa low marsh; S, Samhwa marsh.

methods of McKeague *et al.* (1971) with an inductively coupled plasma-atomic emission spectrophotometer (ICPS-1000IV, Shimadzu, Japan). The concentrations of heavy metals were determined by the standard methods for the examination of soil pollution by the Ministry of Environment, Korea (1999). Soil was extracted with 0.1 N HCl for Zn, Cu, Cd, Pb, and Cr and with 1 N HCl for As. For Hg determination, soil was digested with concentrated nitric and perchloric acid. The concentrations of the heavy metals in the extracts were analyzed with the atomic absorption spectrophotometer (AA-6510F, Shimadzu, Japan).

RESULTS AND DISCUSSION

Characteristics of soils

Wetland soils experience water-saturated conditions for a substantial part of the year. In general, reducing conditions cause increases in pH, while oxidation decreases pH. Gambrell (1994) reported that pH of water-saturated soils in wetland tends to converge toward 7, regardless of whether the soil was acidic or alkaline initially. In this study, soil samples had pH values between 6.5 and 7.5 with a few exceptions, reflecting the anaerobic condi-

tions (Table 1). In particular, the neutral soil pH could also be attributed to properties of tidal marshes. Low redox potential and high pH favor heavy metal removal by adsorption (Harter 1983, Patrick and Verloo 1998). For example, heavy metals are subject to change into insoluble hydroxy species at high pH (Farrah and Pickering 1977, Holmgren *et al.* 1993). Under anaerobic conditions, sulfate ions (SO_4^{2-}) are reduced to sulfide (S^{2-}) which, then, leads to the precipitation of metal sulfides such as CdS, HgS, CuS, and ZnS (Sposito and Page 1985). Therefore, this result suggests that soil of Shihwa marsh can serve as an effective sink of heavy metal. Electrical conductivity of soil ranged from 0.2 to 3.1 dS m^{-1} , and decreased with time. Organic matter content of marshes soil did not show large variation: 0.2~1.7 (average 1.0 \pm 0.3) at June 2000, and 0.7~1.6 mg kg^{-1} (average 1.2 \pm 0.3 mg kg^{-1}) at July 2001, and the content was lower than that of a paddy soil in Korea (25 mg kg^{-1} , n=1168) reported by Jung *et al.* (1998). Oxalate extractable Fe and Al contents varied spatially, but did not show any significant change temporally; the sum of oxalate extractable Fe and Al content was 3377~7723 (5051 \pm 1197) at June 2000, and 3660~7574 mg kg^{-1} (5256 \pm 1186 mg kg^{-1}) at July 2001.

Table 2. Variations in heavy metal concentrations of soils (unit: mg kg^{-1})

Metals	Sampling time	Sites ^a																	
		B _{H1}	B _{H2}	B _{H3}	B _{H4}	B _{L1}	B _{L2}	B _{L3}	B _{L4}	D _{H1}	D _{H2}	D _{H3}	D _{H4}	D _{L1}	D _{L2}	D _{L3}	D _{L4}	S1	S2
Zn	Jun-00	18.2	18.3	11.9	16.8	10.0	13.6	7.9	5.3	18.1	9.9	11.9	4.7	14.4	7.3	8.4	9.1	5.3	4.5
	Nov-00	36.9	21.3	13.1	20.3	10.1	7.8	46.1	11.4	23.2	8.6	6.8	8.7	10.9	15.8	10.0	9.9	7.2	7.3
	Jul-01	66.4	33.4	27.8	41.0	19.9	16.9	31.8	18.5	47.8	16.3	10.2	12.5	26.5	23.5	17.3	24.5	12.2	11.2
Cu	Jun-00	42.9	33.1	10.6	26.0	8.0	11.7	9.2	4.9	31.2	7.6	9.2	3.7	14.3	6.1	5.3	5.8	2.7	2.0
	Nov-00	50.1	31.5	17.0	30.0	10.2	6.5	80.5	14.5	36.2	9.4	6.9	9.2	10.3	20.2	8.5	8.6	6.2	4.5
	Jul-01	97.5	34.2	31.6	49.9	9.7	13.5	38.7	14.2	63.3	11.3	4.9	7.2	16.1	19.4	7.4	11.6	5.1	7.5
Cd	Jun-00	0.1	0.1	0.1	0.1	0.1	0.1	0.8	0.1	1.5	0.1	0.1	0.1	0.2	0.1	0.1	0.1	0.2	0.1
	Nov-00	0.3	0.2	0.2	0.2	0.2	0.2	0.3	0.3	0.1	0.2	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2
	Jul-01	0.2	0.1	0.1	0.1	0.3	0.2	0.3	0.3	0.1	0.2	0.1	0.1	0.3	0.2	0.2	0.2	0.3	0.2
Pb	Jun-00	12.3	10.5	6.6	10.1	6.5	7.2	7.6	6.6	9.6	7.4	4.6	3.8	7.8	7.7	7.1	7.4	8.3	6.6
	Nov-00	15.0	9.5	7.4	7.2	6.0	5.9	14.0	6.2	7.8	5.9	2.2	3.2	5.0	5.8	4.6	6.0	5.6	5.1
	Jul-01	15.9	9.9	10.6	12.1	7.5	7.4	11.2	7.2	12.1	7.7	5.0	5.9	9.0	9.4	7.5	7.5	8.3	5.1
Cr	Jun-00	1.4	1.3	0.7	1.1	0.7	0.7	0.7	0.6	1.0	0.7	0.5	0.6	0.7	0.6	0.3	0.3	1.9	0.3
	Nov-00	0.1	0.3	1.4	1.0	1.8	2.1	1.2	1.8	1.6	1.1	2.3	2.3	2.3	1.7	2.3	2.8	1.8	2.7
	Jul-01	2.8	1.6	3.8	2.4	1.5	1.4	2.6	2.9	6.0	2.1	1.5	1.1	2.3	1.5	1.7	1.3	1.7	1.6
As	Jun-00	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.3
	Nov-00	0.3	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.2	0.2	0.2	0.2	0.2	0.3	0.2	0.3	0.3
	Jul-01	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.4	0.3	0.4	0.4	0.5	0.3
Hg	Jun-00	0.2	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.1	0.2	0.2	0.2	0.2	0.1	0.2	0.1	0.1
	Nov-00	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
	Jul-01	0.1	0.2	0.1	0.1	0.2	0.1	0.2	0.2	0.2	0.2	0.2	0.1	0.2	0.2	0.2	0.2	0.2	0.2
Sum	Jun-00	75.6	63.7	30.2	54.4	25.6	33.6	26.6	18.0	61.9	26.0	26.6	13.3	37.7	22.2	21.7	23.1	18.8	14.0
	Nov-00	102.8	63.2	39.4	59.0	28.6	22.8	142.5	34.5	69.4	25.5	18.7	23.7	29.0	44.0	26.0	27.8	21.4	20.1
	Jul-01	183.3	79.6	74.3	105.9	39.4	39.9	85.1	43.5	129.7	38.1	22.2	27.2	54.8	54.5	34.8	45.7	28.2	26.1

^a B_H, Banweol high marsh; B_L, Banweol low marsh; D_H, Donghwa high marsh; D_L, Donghwa low marsh; S, Samhwa marsh.

Variations in heavy metal concentrations

The concentrations of heavy metals showed temporal and spatial variations (Table 2). In general, total concentrations of heavy metals (Sum in Table 2) increased with time from 30.2~54.4 (average 56.0) to 74.3~183.3 mg kg⁻¹ (average 110.8 mg kg⁻¹, LSD=34.2) for Banweol high, 18.0~33.6 (average 25.9) to 39.4~85.1 mg kg⁻¹ (average 52.0 mg kg⁻¹, LSD=50.3) for Banweol low, 13.3~61.9 (average 31.9) to 22.2~129.7 mg kg⁻¹ (average 54.3 mg kg⁻¹, LSD=30.0) for Donghwa high, 21.7~37.7 (average 26.2) to 34.8~54.8 mg kg⁻¹ (average 47.4 mg kg⁻¹, LSD=12.0) for Donghwa low, and 14.0~18.8 (average 16.4) to 26.1~28.2 mg kg⁻¹ (average 27.1 mg kg⁻¹, LSD=5.6) for Samhwa marshes between Jun 2000 and July 2001. These temporal increases indicate the accumulation of heavy metal in soils. In particular, accumulations of Zn, Cu, and Cr were much greater than those of other metal. Compared with average heavy metal concentrations of paddy soils in Korea (3.9, 4.5, and 0.4 mg kg⁻¹ for Zn, Cu, and Cr, respectively, n=1196) reported by Kim *et al.* (1995), the concentrations of Zn, Cu, and Cr in soils of Shihwa marsh were much higher. In contrast, Cd and Pb concentrations were similar or slightly higher than those of paddy soils. These results could be attributed to inflows of industrial wastewater with high Zn, Cu, and Cr concentrations as suggested by Hyun *et al.* (1999). In other words, the higher concentrations of these metal species in Shihwa marsh suggested that this marsh plays an important role in preventing Shihwa lake from heavy metal pollution.

In this survey, Banweol high marsh showed relatively high metal concentrations, while Samhwa marsh showed lower concentrations, compared with other marshes. In particular, in Banweol high and Donghwa high marshes, the metal concentrations were much higher at a point closer to stream water inlet (B_{H1} and D_{H1}), reflecting metal removal from water by adsorption onto soils during transport of water to outlet. However, these spatial variations are not easy to interpret, because several factors,

such as heavy metal concentrations of recharged water, heavy metal uptake by reeds, and soil variables affecting adsorption of heavy metal, should be considered with the metal concentrations in soils (Gambrell 1994). In this study, however, we just considered the effects of soil variables on heavy metal concentrations by a correlation analysis. (Table 3).

Total heavy metal concentrations were correlated positively with organic matter and oxalate extractable Fe and Al contents, while negatively with electrical conductivity. No significant correlation was found with pH (Table 3). However, it was an unexpected result that the concentrations of Zn, Cd, As, and Hg were negatively correlated with pH, because solubility of heavy metal is inversely related to pH as discussed previously. As seen in Table 1, the pH ranged from around 6.5 to 7.5 with a few exceptions. Since the solubility of heavy metal sharply drops at these pH ranges (Farrah and Pickering 1977), it would be difficult to observe a positive correlation between those two parameters.

A negative correlation between metal concentrations and electrical conductivity indicates the effect of competing ions on the adsorption of metal onto soils. Christensen (1984) reported that Cd sorption was decreased by 30% when the Ca concentration was increased by ten-fold, and he attributed this to competition by Ca for adsorption sites. Lim *et al.* (1999) also suggested that adsorption capacity for heavy metal of tidal soils increased with decreasing base saturation. Therefore, our result suggests that metal removal capacity of Shihwa marsh would increase as desalinization proceeds with time. Retention of heavy metal by organic matter, in particular humic and fulvic acid, has been well reported (Harter 1983). Therefore, a positive correlation between metal concentrations and organic matter contents of soils could be attributed to the formation of metal-organic precipitates. According to Brummer and Herms (1983), metals are immobilized by soil organic matter in the order of Cu > Cd > Zn > Pb and the immobilization effect depends on soil pH. They suggested that metal immobilization by organic matter decreased with

Table 3. Correlation coefficients between metal concentrations and selected parameters of soils (n=54)

Metal	Soil parameters					
	pH(1:5)	EC (1:5)	Organic matter	Oxalate-Fe ^b	Oxalate-Al ^c	Oxalate-(Fe+Al) ^d
Zn	-0.28*	-0.46***	+0.51***	+0.41**	+0.42**	+0.42**
Cu	-0.16	-0.33**	+0.42***	+0.42**	+0.40**	+0.42**
Cd ^a	-0.39**	-0.36**	+0.36**	+0.08	+0.09	+0.09
Pb	-0.25	-0.19	+0.63***	+0.49***	+0.48***	+0.50***
Cr	-0.19	-0.48***	+0.37**	+0.08	+0.02	+0.07
As	-0.55***	-0.48***	+0.39**	-0.02	-0.02	-0.02
Hg	-0.27*	+0.10	+0.12	+0.01	-0.02	-0.00
Sum	-0.22	-0.39**	+0.50***	+0.43**	+0.42**	+0.44**

* P<0.05, ** P<0.01, *** P<0.001.

^a Cd contents for B_{L3} and D_{H1} samples in June 2000 were excluded.

^b Oxalate-extractable Fe, ^c Oxalate-extractable Al, ^d Sum of oxalate-extractable Fe and Al.

increasing soil pH through formation of soluble organic-metal complexes. Therefore, it is expected that the correlation between the two parameters would be strong when soil pH decreases.

It is well established that oxides of Fe and Al effectively adsorb or occlude most trace and toxic metal (Gambrell 1994). Our results are in accordance with previous studies in a way that metal concentrations of soils are positively correlated with the concentrations of oxalate extractable Fe and Al (Table 3). However, hydrous oxides of Fe or Al tend to become unstable under typically reduced, flooded soil conditions. Nevertheless, they are effective in controlling metal retention for surface soils of wetland. Giblin *et al.* (1986) has shown Fe oxyhydroxides were important in retaining metals in surface sediment of a salt marsh receiving trace and toxic metals.

In conclusion, Shihwa marsh has favorable conditions (neutral pH and water-saturated conditions) for heavy metal removal by adsorption onto soils. This study shows that the concentrations of heavy metal (especially Zn, Cu, and Cr) in soils of Shihwa marsh are increasing with time, suggesting heavy metal removal from surface water through adsorption onto soils. Among sub-marshes, the metal concentrations were highest in Banweol high marshes and lowest in Samhwa marshes. The heavy metal concentrations of soils were correlated positively with organic matter and oxalate extractable Fe and Al contents, but negatively with electrical conductivity. These correlations allow us to expect that the metal-removing capacity of soils would increase with time. However, because metal uptake by reeds would contribute to metal removal in this marsh (Stoltz and Greger, 2002), the concentrations of metal in reeds should also be investigated through further study to evaluate the metal removing capacity of Shihwa marsh (by both soil and plant).

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