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Uptake of heavy metals, arsenic, and antimony by aquatic plants in the vicinity of ore mining and processing industries

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Abstract

The uptake of heavy metals, As, and Sb by aquatic plants — fluvial horsetail, platyphyllous cattail, etc. — growing in industrial collection ponds of metal mining industry in the Kemerovo region, Russia, was studied. Cu, Pb, Cd, Zn, As, and Sb are the major pollutants in these plant habitats. The elemental concentrations in plants, their acid extracts, and the bulk water chemistry were determined by flame and electrothermal atomic absorption spectrophotometry. Sediments were analyzed by X-ray fluorescence. The analytical data obtained demonstrate high pollutant concentrations in sediments, water, and plants, especially in their roots. Submerged hornwort and pond scum were shown to be hyperaccumulators. The heavy metal concentrations in the plants exceed the corresponding concentrations in the habitat (water) by tens of thousand times. Analysis of fresh platyphyllous cattail acid extracts has demonstrated that As and Sb present a serious threat to human and animal health, as they are capable of entering food chains in large amounts. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: heavy metals; arsenic; antimony; sediment; water; aquatic plants; foodwebs

1. Introduction

The aquatic plants growing in collection ponds of ore refining plants (ORPs) take up large amounts of elements such as Cu, Pb, Zn, As, Hg, etc. For example, macrophytes in certain Canadian lakes receiving mine drains contain As in amounts of 150–3700 μg/g dry wt (Wagemann et al., 1979). Zn content in aquatic macrophytes growing in mining regions of eastern and northern Canada reaches 12 300 μg/g dry wt (Franzin and McFarlane, 1980). Metal concentration in plants depends on the total soil concentrations, the chemical speciation of metals in soils and soil solutions (Sposito, 1983; Sposito and Page, 1984; Kabata-Pendias and Pendias, 1992) and the involvement of the metal in biological functions (Thornton, 1983; Bowie and Thornton, 1985; Adriano, 1986; Kabata-Pendias and Pendias, 1992). The feature of hydrophytes to take up large amounts of trace elements has a dual manifestation. On the one hand, these plants represent a biological barrier for migration of toxic elements into the environment. Jung and Thornton (1996) demonstrated quantitatively that revegetation decreased the heavy metals flow from tailing impoundments. Ernst (1988, 1996) considers that revegetation and/or recultivation at a later development stage is a reasonable solution in controlling heavy metals-contaminated sites, since soil decontamination by biological agents allows less than 1% of the metal to be removed over a century.

On the other hand, aquatic plants enhance the

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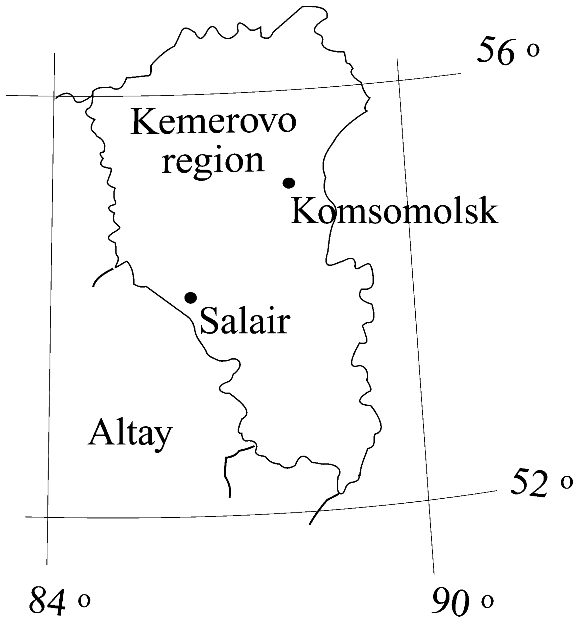

Fig. 1. Location of the Salair and Komsomolsk towns.

penetration of adverse chemical elements into both the environment and food chain constituents. It is known that consumption of grass growing on metal-contaminated soils presents a great danger to man and cattle (MAGS, 1975). A change in pH from 8.4 to 6.9 due to higher plants introduced in a post-flotation waste pond of the Trzebionka Mine (Poland) was reported (Tafas, 1996). The related increase in heavy metals mobility allowed these pollutants to easily penetrate into the environment. According to Kuiters and Mulder (1993), organic acids produced by higher plants are capable of changing the species in which metals occur.

This work focuses on the uptake patterns of heavy metals, As, and Sb by aquatic plants in the system 'sediment–water–aquatic macrophytes'. The detection of plants suitable for remediation of collection ponds will also be discussed.

1.1. Study area

The work was carried out on two ore refining plants in the Kemerovo region (southwestern Siberia) — Salair and Komsomolsk (Fig. 1).

The collection ponds of the Salair ORP consist of a set of natural hollows filled with wastes of flotation and cyanidation of complex barite ores. It consists of three interconnected, presently inactive collection ponds of Dyukov ravine (northern, southern, and the well) and presently active settling and collection ponds of Salagaev ravine (Fig. 2). Cu, Sb, Zn, and Cd are the major pollutants in the Salair ORP ponds. As a result of waste storage, small manmade lakes are formed. They are solid-matter wastes (tens of thousands of tons) covered by water. A part of drainage waters enters a river of M. Talmovaya. Unlike collection ponds, an artificial pond called 'Settler' on M. Talmovaya River is designed to receive emergency effluents of the plant. The upper flow of M. Talmovaya River upstream of the plant, was chosen as a control area.

Komsomolsk tailing pond comprises collection and settling ponds (Fig. 3). Since 1964, the collection pond is filled with wastes of gold–arsenopyrite quartz ore cyanidation. Typical of these wastes are high contents of pollutants such as As and Sb. After treatment with iron vitriol, the water of the collection pond is conveyed to a settling pond. From the settling pond, water comes to the Voskresenka River. The Berchikul Lake, located in the city of Komsomolsk, 10 km from the plant, was chosen as a control area.

The most representative plant species at the two locations were sampled: platyphyllous cattail (*Typha latifolia*), sylvan bulrush (*Scirpus sylvaticus*), and fluvial horsetail (*Equisetum fluviatile*) for the Salair ORP collection ponds; platyphyllous cattail, sylvan bulrush, and common reed (*Phragmites australis*) for the Komsomolsk collection ponds. These plant species display a wide ecological tolerance. The plants displayed morphological changes such as growth suppression. The height of several adult plants of common reed found in the Salagaev collection pond did not exceed 50 cm, whereas its height under normal conditions reaches 4 m (Komarnitskii et al., 1975). These plants were found growing in technogenic sediments whose layer was considerably thicker than the root length. Low-height plants of sylvan bulrush were also found within both the sites.

2. Materials and methods

2.1. Sampling

Averaged samples of aboveground and underground

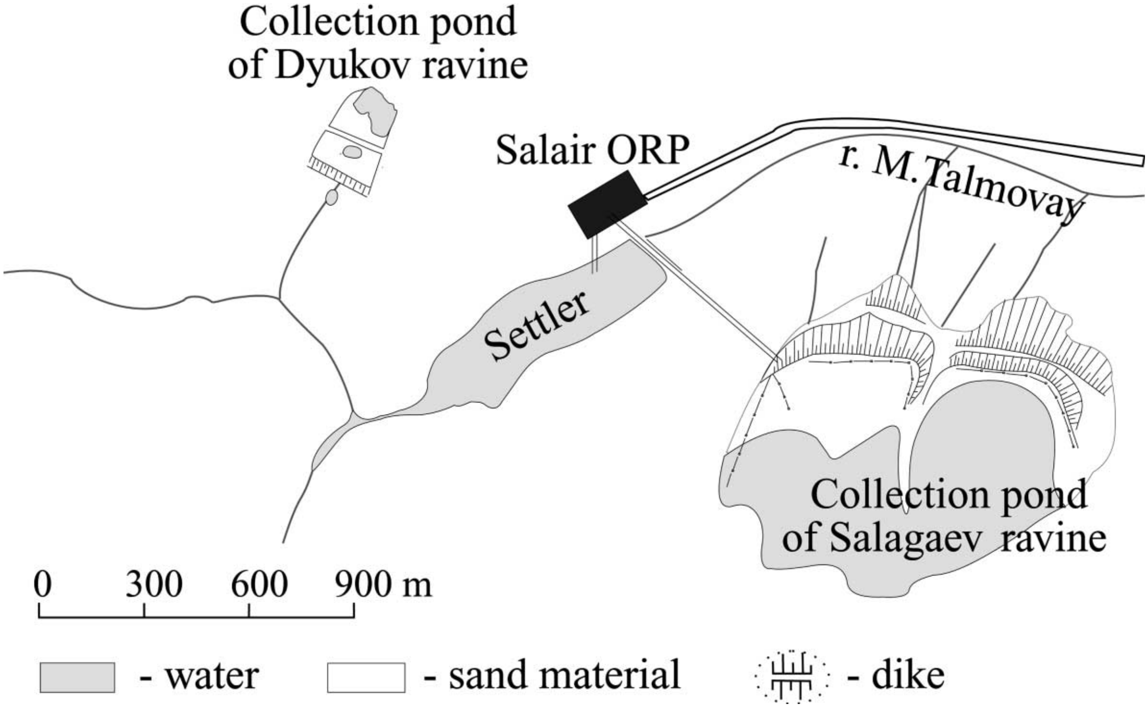

Fig. 2. Scheme of the Salair ORP.

parts of these aquatic plants, their bleeding sap (flow of nutrients supplied by the root system to the aboveground organs), and samples of sediment and water on the site of their growth were collected during the field studies at both locations.

Since the area occupied by each plant species studied was small enough (20–30 m^2) in each collection pond, two averaged samples were taken from each such area for each species — one sample of the aboveground and one sample of underground parts. For this purpose, five sampling points equidistant from one another were selected in the area and three plants were collected from each point. The plants, especially their roots, were thoroughly washed with the lake water, and their above- and underground parts were separated. The aboveground parts of 15 plants were united and mixed to form one averaged aboveground sample. Similarly, an averaged plant underground sample was produced. Then, these averaged samples were dried to an air-dry state and reduced to powder.

Averaged bleeding sap samples were collected from three plants of each species equidistant from one another according to Sabinin's technique (Agrokhimicheskie metody, 1975). The essence of this technique is as follows. The plant stem is cut at a certain distance from the soil surface (1.5–2.0 cm). A narrow rubber tube is put on the cut end. A stopper with a hole for air outlet is inserted into the tube's free end. The accumulated plant sap was carried over from rubber tube into glass vial, containing 2–3 drops of toluene. The samples collected were kept in a refrigerator until analyzed. In case the bleeding sap amount was less, it was collected from the tube walls with pieces of filter paper (1×2 cm^2), each absorbing about 0.05 ml liquid, and transferred into solution by extraction with distilled water.

Acid extracts were obtained from averaged aboveground and underground samples. For this purpose, 5 g of a raw plant material from an averaged sample were ground and supplemented with 50 ml HCl with the acidity (pH 2) corresponding to that of gastric juice of cattle, consuming actively aquatic plants of manmade lakes. The solutions were filtered after 1–1.5 h and stored in a refrigerator until the analysis.

Samples of bulk water were taken in 5 points of a tailing impoundment and mixed. Averaged water

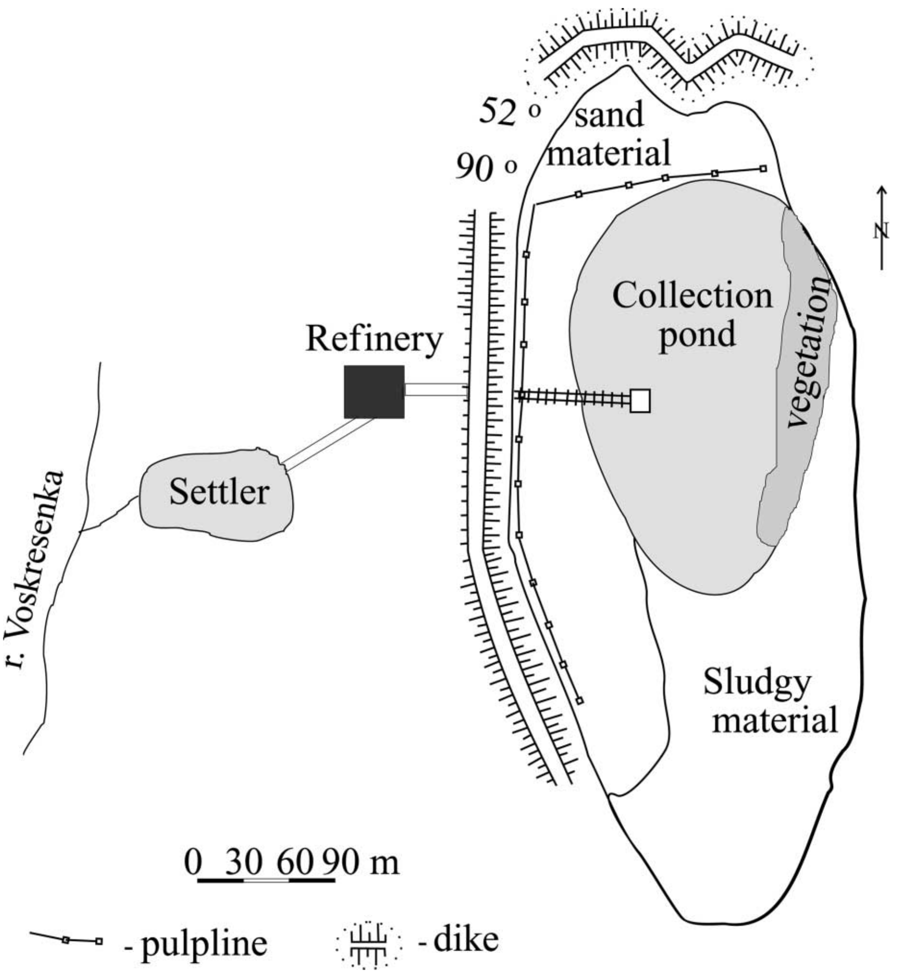

Fig. 3. Scheme of the Komsomolsk ORP.

samples were filtered through Nuclepore filters (0.45 μm), collected in clean polyethylene bottles, acidified with 1% HNO_3, and stored at 10°C until assayed.

Sediment samples were collected from each 5 points of the sampling areas at a depth of 10–15 cm, where the main root mass of aquatic macrophytes such as horsetail, cattail, and sylvan bulrush are located. In laboratory sediment samples were dried, ground, and mixed in one vessel to obtain an average sample.

2.2. Analytical procedures

Samples (1 g) of dried and ground plant material were decomposed according to Millon's technique with an acid mixture (1: 4 v/v conc. H_2SO_4:HNO_3) by heating for 15 min (Bock, 1979). Each solution, containing a sample, is heated until the solution becomes transparent.

The contents of heavy metals, arsenic, and antimony in the solutions obtained, bleeding sap, acid extracts, and water were determined by (1) atomic absorption spectrometry (AAS) using an air–acetylene flame with deuterium background correction (PV-Unikam SP-9) and (2) flameless AAS with Zeeman background correction (Perkin–Elmer 3030z and graphite furnace HGA-600). Metal concentrations in sediments were determined by X-ray fluorescence (XRF).

Table 1
Heavy metals in sediment and bulk water of the Salair ORP collection ponds

		Pb	Cu	Cd	Zn
Sediment (%)					
Control area (upper flow of M. Talmovaya River)		0.0025	0.0015	0.000030	0.0040
Dyukov ravine	Well	0.54/216[a]	0.26/173	0.0084/280	1.9/475
	Southern lake	0.49/196	0.22/147	0.0019/63	0.78/195
	Northern lake	0.54/216	0.16/107	0.00060/20	0.27/68
Settling		0.090/36	0.26/173	0.0005/17	0.80/200
Salagaev ravine		0.30/120	0.21/140	0.0015/50	0.54/135
Bulk water (μg/l)					
Control area (upper flow of M. Talmovaya River)		0.60	1.9	0.14	10
Dyukov ravine	Well	3.0/5	28/15	10/71	6300/630
	Southern lake	9.0/15	19/10	3.0/21	4800/480
	Northern lake	15/25	8.6/4.5	0.1/0.7	31/3
Settling		100/167	30/16	10/71	500/50
Salagaev ravine		0.70/1.2	25/13	1.7/12	310/31

[a] CC values are shown as denominators.

2.3. Data treatment section

The ratio between a trace element concentration in sediment, water or plant of the tailing impoundment and that of the corresponding control area was used to estimate the value of technogenic load in tailing impoundments (Moore and Ramamoorthy, 1987):

$$CC = C_{ti}/C_{ca},$$

where CC is concentration coefficient; C_{ti} is trace element concentration in sediment, water, or plant of a tailing impoundment and C_{ca}, its concentration in the same components of the control area.

The ratio between element concentration in underground and aboveground parts of a plant also called acropetal coefficient (AC) was used for the same purpose:

$$AC = C_u/C_a,$$

where C_u corresponds to the trace element in the underground part and C_a, its concentration in the aboveground part. According to Brooks (1986), the lower the AC is, the lower is the technogenic load on the plants.

The ratio between a trace element concentration in the aboveground part of plant and the bleeding sap — the so-called xylem coefficient (XC), was used to estimate the element mobility in the sap:

$$XC = C_a/C_{bs},$$

where C_a is the trace element concentration in the aboveground part and C_{bs}, its concentration in the bleeding sap. The lower is XC, the smaller the fraction is of the element that remains in the plant and the larger the fraction is that can be released into the environment.

Concentration factor (CF), as a ratio of a chemical element content in live matter to its content in the corresponding habitat, was used to estimate the extent of heavy metals, As, and Sb concentration by plants.

$$CF = C_p/C_{ph},$$

where C_p is the element concentration in the plant and C_{ph}, their concentration in the plant habitat. The habitat for unrooted aquatic plants (submerged hornwort and pond scum) is water (Moore and Ramamoorthy, 1987).

3. Results

3.1. Salair

According to the data obtained, the plant habitat contains high quantities of elements such as Pb, Cu, Cd, and Zn. The average Pb, Cu, and Zn concentrations in sediments varies between 0.20 and 0.50%. The Cd concentration is between 6.0 and 80 ppm. Their CCs in sediments vary from 17 to 475 (Table 1). The concentration of Pb, Cu, and Cd in bulk water varies from 0.10 to 100 μg/ml. The Zn concentration plots between 31 and 6300 μg/ml. Their CCs in bulk water is situated in the range of 0.7–630 (Table 1).

Fluvial horsetail exemplifies best the interactions in the water–sediments–aquatic plants system, although the concentrations of the elements found in this plant are at the same level of magnitude as in the other two plants studied, namely platyphyllous cattail and sylvan bulrush. The concentrations of Pb, Zn, and Cd in horsetail plants exceed that in the control area by a factor of ten and more. The Cu concentration is twice as high, twofold (Table 2). The concentrations of Pb, Cu, and Zn in platyphyllous cattail plants differ for each particular lake. The Pb concentration varies from 7.5 to 2400 μg/g dry wt; Cu and Zn concentrations vary from 33 to 4300 μg/g dry wt, exceeding the corresponding concentrations in the control area by a factor of ten. The Cd concentration varies from 1.0 to 19 μg/g dry wt. It exceeds the concentration in the control area by a factor of hundred (Table 3). Pb, Cu, Zn, and Cd concentrations in sylvan bulrush exceed the corresponding concentrations in the control area by many times (Table 3).

The heavy metals are mainly accumulated in the roots (Tables 2 and 3). On the one hand, this indicates a high content of mobile species in sediments; on the other it suggests that protective barriers exist, preventing the toxicant to penetrate from the roots into the aboveground plant parts.

There is deficiency of biogenic elements such as Fe and Mn in the fluvial horsetail as compared with the values in the control area. The Fe concentration in the aboveground part is 32-fold lower than the control one; in the underground part, 1.3-fold lower. Mn concentrations are 2-fold and 1.3-fold lower in the aboveground and underground parts, respectively (Table 2). However, the horsetail plants from Dyukov ravine and the control area displayed no exceptional differences.

CC of As, Pb, Zn, and Sb in the aboveground part of horsetail plants amount to 1.7, 25, 46, and 30, respectively, exceeding the underground CC in case of As by a factor of 9. For Pb, Zn and Sb the factor is respectively 13, 6 and 63 (Table 2). Thus, these elements penetrate into the aboveground parts of horsetail plants growing in the well of Dyukov ravine more

Table 2
Heavy metals, As, and Sb concentrations in fluvial horsetail plants growing in the well of Dyukov ravine

		Pb	Cu	Cd	Zn	As	Sb	Fe	Mn
Fluvial horsetail (*Equisetum fluviatile*)									
Control area	Aboveground	0.89	39	0.084	26	0.9	1.3	18000	490
(upper flow of M. Talmovaya River)	Underground	100	250	0.67	670	110	98	13000	660
Dyukov ravine, well	Aboveground	22	68	3.0	1200	1.5	39	560	240
	Underground	190	560	28	4800	22	47	10000	520
CC of fluvial horsetail									
Dyukov ravine, well	Aboveground	25	1.7	36	46	1.7	30	0.03	0.5
	Underground	1.9	2.2	42	7.2	0.2	0.48	0.78	0.79
AC of fluvial horsetail									
Control area (upper flow of M. Talmovaya River)		112	6.4	7.8	26	122	75	0.72	1.3
Dyukov ravine, well		8.6	8.2	9.3	4	14.7	1.2	18	2.2
Bleeding sap of fluvial horsetail (μg/l)									
Dyukov ravine, well		220	89	7.7	7200	55	11	2800	3200
XC of fluvial horsetail									
Dyukov ravine, well		110	764	390	167	27	3545	200	75

Table 3
Heavy metals concentrations in aquatic macrophytes of the Salair ORP, μg/g dry wt

			Pb	Cu	Cd	Zn
Platyphyllous cattail (*Typha latifolia*)						
Control area		Aboveground	23	4.7	0.023	51
(upper flow of M. Talmovaya River)		Underground	20	20	0.14	31
Dyukov ravine	Well	Aboveground	46	28	1.0	340
		Underground	100	150	19	4300
	Southern lake	Aboveground	820	99	1.0	40
		Underground	2400	580	4.0	450
	Northern lake	Aboveground	7.5	7.0	1.6	33
		Underground	600	39	11	430
Settling		Aboveground	48	34	2.3	400
		Underground	46	130	7.2	1400
Salagaev ravine		Aboveground	18	19	1.3	210
		Underground	80	81	11	510
Sylvan bulrush (*Scirpus sylvaticus*)						
Control area		Aboveground	–	10	1.8	84
(upper flow of M. Talmovaya River)		Underground	23	12	1.3	40
Salagaev ravine		Aboveground	31	28	1.0	410
		Underground	37	49	1.7	150

efficiently than into their underground parts. This is likely to relate to a failure of the protective mechanisms on the underground—aboveground boundary due to a larger amount of pollutants penetrating into the plant roots because of the high technogenic load. Consequently, these elements commence penetrating into the vegetative part with a higher efficiency compared with that in the control area. However, this may be explained by the fact that the horsetail plants in the well are growing deeper into water, where the elemental concentrations are higher than those in the control area. This is most likely the reason why the amounts of As, Pb, Zn, and Sb that can enter directly the aboveground part of these plants in the well of Dyukov ravine is higher than in the control area.

The AC of Cu and Cd for horsetail plants from the well of Dyukov ravine and the corresponding control area exceed 1, indicating a gradient of pollutant concentrations on the underground–aboveground boundary in plants (Table 2). This indicates the presence of protective mechanisms. Note that the AC for Cu for the horsetail plants from Dyukov ravine is equal to that from the control area. Consequently, the efficiencies of protective mechanisms with respect to copper are similar in both areas.

Certain amount of each trace element enters the vegetative organs via bleeding sap (the flow of nutrients) (Table 2). A certain amount of trace elements stays in the plant, whereas the rest is released outside. XC characterizes the fraction of an element remaining in the vegetative organs. XC decreases in the line $Sb > Cu > Cd > Fe > Zn > Pb > Mn > As$ (Table 2). The lesser the XC is, the smaller is the fraction of an element remaining in the plant and the greater is its fraction released. Arsenic excretion is the most efficient ($XC \leq 27$); Mn is the next ($XC = 75$); then goes Pb ($XC = 100$), etc. Toxicants like Sb, Cu, and Cd are poorly excreted from the aboveground parts of horsetail plants. Therefore, these elements are accumulated in the aboveground organs. It then can pass into food chains.

Hyperaccumulator plants (Brooks, 1986), were discovered in the study locations. Submerged hornwort and pond scum are such plants (Table 4). Hornwort plants hyperaccumulate Cd, Cu, and Zn: concentrations of these metals in plants are 500-, 20 000-, and 4200-fold higher than in water, respectively. Pond scum as hyperaccumulator behaves strikingly. Its contents of Pb, Cu, Cd, and Zn are by 2–5 orders of magnitude higher than in water.

Table 4
Heavy metals concentrations in hyperaccumulators (ppm = μg/g dry wt) growing in collection ponds of the Salair ORP and their concentrations in bulk water (ppm = 10^{-3} μg/l); numerator to denominator, respectively

	Submerged hornwort (*Ceratophyllum demersum*)	Pond scum (*Algae*)		
	Salagaev ravine	Dyukov ravine, well	Dyukov ravine, well	Salagaev ravine
Pb	–/0.10	390/0.0030	3.0/0.015	120/0.10
Cu	600/0.030	3900/0.028	43/0.0086	360/0.030
Cd	5.0/0.010	130/0.010	2.0/0.00010	4.0/0.010
Zn	2100/0.50	30000/6.3	370/0.031	4800/0.50

Pond scum and hornwort absorb essential amounts of pollutants, grow in bulk water, and have no roots. Therefore, they can be placed in bulk water around the orifice of a drain pipe for decontaminating the water that goes throw the pipe, so-called drainage water of collection ponds. One part of the drainage water is returned to the plant for technical use and the other, discharged into M. Talmovaya River. The plant layer saturated with pollutants can be easily removed and substituted with fresh plants. Such a purification system would decrease essentially the technogenic load of the local rivers.

3.2. Komsomolsk

Unlike near the Salair plant, the hydrophyte habitats of Komsomolsk ORP contains high amounts of As and Sb. Their concentrations in water and sediments are by 2–4 orders of magnitude higher than that compared to the concentrations recorded in the control area. CCs vary from 100 to 4500 (Table 5). Arsenic and antimony are accumulated predominantly in plant roots, exceeding the contents in aboveground parts by 1–2 orders of magnitude. Correspondingly, their ACs are greater than 1 (Table 6). This indicates high As and Sb concentrations in the habitat. At such concentrations (As — 0.21% and Sb — 0.080% (Bortnikova et al., 1998), these elements are toxic for plants. The protective mechanisms at the aboveground–underground boundary are activated and prevent toxicants from penetrating into vegetative organs. Thus, pronounced gradients of As and Sb concentrations between the roots and vegetative organs occur.

The As and Sb concentrations in platyphyllous cattail and sylvan bulrush roots are equal to or higher than in the sediment (Fig. 4). Therefore, these plants can be used to decrease the pollutant migrations from the sediment into the soil around the collection pond. The roots of hornwort and bulrush planted on manmade lakefronts will accumulate As and Sb from the upper soil layer, thus decontaminating the latter. Possibly, it will be necessary to remove and utilize the plants having accumulated these metals to prevent secondary soil pollution.

The acid extracts obtained from fresh plants at pH 2 reflect the danger of trace element transfer from hydrophytes into food chain constituents. Acidity of the extraction solution is similar to that of ruminant gastric juice. However, the solution lacks the microorganisms and plant tissue-digesting enzymes present in the gastric juice. Therefore, plant acid extracts

Table 5
Concentrations of chemical elements in sediments and bulk water of the Komsomolsk ORP collection pond

	As	Sb	Fe	Mn	As	Sb
	Sediment (%)				Bulk water (μg/l)	
Control area (Berchikul Lake)	10	7.0	3.0	57	1.9	0.8
Collection pond	1000/100[a]	780/111	340/113	38000/667	200/105	3600/4500

[a] CC values are shown as denominators.

Table 6
Heavy metals in plants growing in the Komsomolsk ORP collection pond (μg/g dry wt)

		As	Sb	Fe	Mn
Platyphyllous cattail (*Typha latifolia*)	Aboveground	5.8	15	390	630
	Underground	850	1300	13000	300
	AC	147	87	33.3	0.48
Sylvan bulrush (*Scirpus sylvaticus*)	Aboveground	13	19	1400	980
	Underground	580	100	9800	350
	AC	45	53	7	0.36
Common reed (*Phragmites australis*)	Aboveground	11	15	690	150
	Underground	270	320	6800	230
	AC	25	21	9.6	1.5

characterize the most mobile fraction of trace elements, which, once the plants reach the stomach, will enter the digestive system of ruminant animals with a high probability. However, it is likely that a larger amount of toxicants enter the digestive system due to the effects of enzymes and microorganisms. Therefore, acid extracts help to assess the transfer of toxicants from hydrophytes into the food chain. The analysis performed demonstrates that the concentrations of As, Sb, and Fe in the aboveground part of the horsetail plants are in orders of magnitude lower than in the underground part (Table 7). However, these amounts present a serious danger for human and animal health. The contents of As and Sb are 0.15 and 1.5 μg/g fresh cattail, respectively. Thus, an animal that consumes 1 kg of fresh cattail will receive at least 0.15 g As and 1.5 g Sb. The normal concentration of As in goods of plant origin does not exceed 1 μg/g (Raily, 1985). But the normal concentration of Sb in goods does not exceed 0.5 μg/g (Rosival et al., 1982). This is 3 times lower than that in the acid extracts of cattail aboveground part.

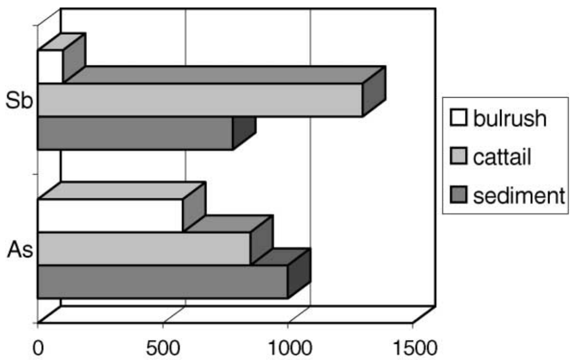

Fig. 4. Concentrations of As and Sb in sediments of the Komsomolsk ORP collection pond (ppm) and in underground part of the cattail and the bulrush plants (ppm).

Table 7
Concentrations of chemical elements in acid extracts (pH 2) of fresh platyphyllous cattail plants (μg/g fresh wt) growing in Komsomolsk ORP collection pond

	As	Sb	Fe	Mn
Aboveground	0.15	1.5	11	4.9
Underground	0.60	7.9	63	5.8

4. Conclusions

Water and sediments of collection ponds contain large amounts of ecologically adverse chemical elements. They are predominantly accumulated in roots of aquatic plants, indicating high contents of mobile elemental forms in sediments and occurrence of protective mechanisms, preventing toxicants from penetrating into the aboveground plant part. However, the toxicants that reached the aboveground plant part can enter the food chains, presenting a potential danger for human and animal health. The plants studied can be used to decrease the heavy metals, As, and Sb discharges into the environment: hornwort and pond scum as hyperaccumulators for decontaminating drainage water and cattail and bulrush for decontaminating soil around collection ponds.

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