Heavy Metals as Status Indicators for the Ob River

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Abstract

This paper presents a comparative analysis of the contents of heavy metals (HM) in the water, suspended matter (SM), and bottom sediments (BS) of the Ob river based on the results of long-term monitoring. The spatial distribution of metals along the middle and lower course of the Ob river is affected by its large tributaries such as Tom', Chulym, and Irtysh and by the Novosibirsk reservoir, as well as by large cities (Novosibirsk, Tomsk, and Nizhnevartovsk). The metal distribution at the interface between water and bottom sediments depends on the hydrological and hydrochemical conditions and is the major factor that is responsible for the higher contents of heavy metals in the bottom sediments of the Novosibirsk reservoir compared with the channel and still water lines of the river. The calculated concentration factors depend on the nature of the metal; the least factors were obtained for lead and zinc. Previous data obtained for the Ob reservoir and the upper Ob in general are discussed.

INTRODUCTION

The Ob-Irtysh basin occupies vast territories in western Siberia, western Kazakhstan region, and northeastern provinces of China. The drainage area of the Ob river is ~3000 thousand km^2, of which 49 % is forest. In the forest zone, the surface slope is insignificant, the runoff is hindered, and the interfluves are strongly waterlogged; especially vast marsh regions are found between the Ob and Irtysh, and also in the Vasyugan region.

Forest-steppe covers vast plains in southwestern Siberia and partly in northern Kazakhstan, amounting to ~404.6 thousand km^2 and including the Tobol, Ishim, Barabinsk, and Ob forest-steppes. The overland runoff is insignificant because many small rivers nearly dry up in summer and freeze up in winter.

The steppe zone occupies about 707.2 thousand km^2 and includes the southwestern part of the Ob Plateau and Kustanay, Ishim, Irtysh, and Kulunda steppes. The steppe plains form a system of flat and generally cutoff interfluves with numerous lakes of which Chany and the Kulunda lake are the largest.

The mountainous regions (222.1 thousand km^2) and tundra (199.6 thousand km^2) are comparable in the drainage area, but the water resources of the tundra territories are approximately five times larger. The mountainous and tundra zones have differences in precipitations, but their common feature is large contributions from inlet glaciers and flood flows resulting from fast melting of snow and snowfields in the former case and spring flood in the latter [1].

PROBLEM FORMULATION

The quality of natural waters is often dictated by the presence of hazardous substances in river waters, but not by the amount of sludge liquor or sediment runoff. Most water

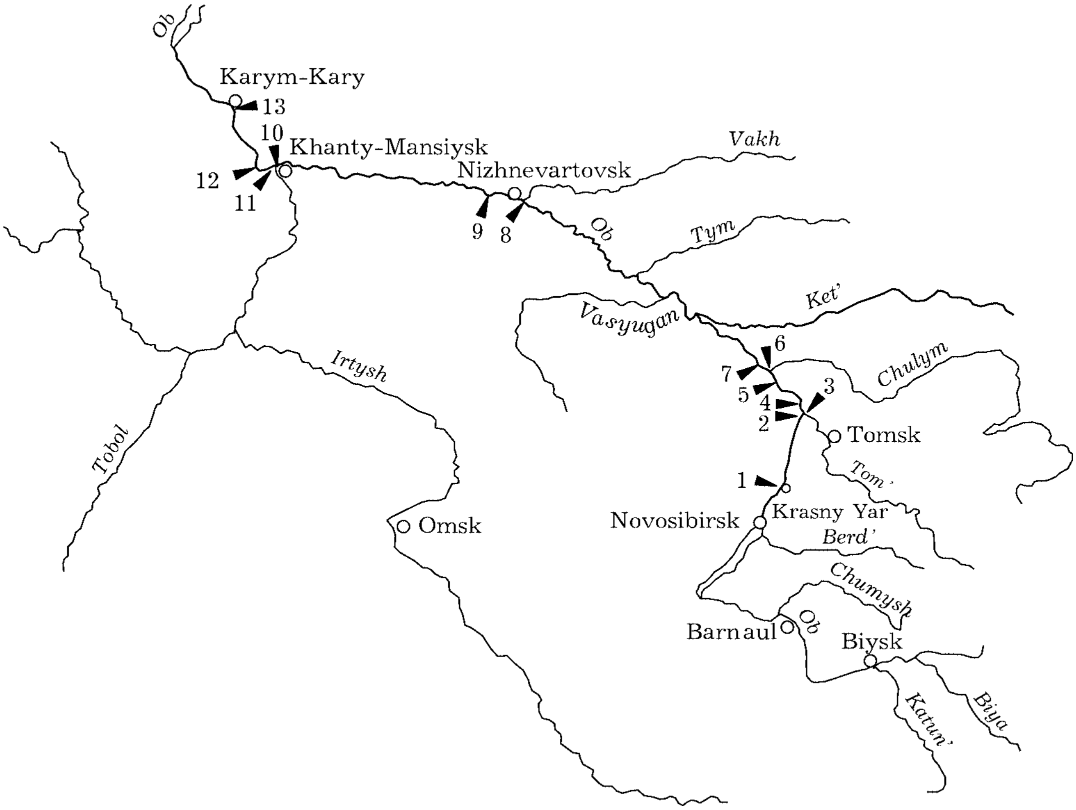


Fig. 1. Skeleton map: *1–13* – test section lines for hydrochemical monitoring.

intake systems in large cities of Siberia (Novosibirsk, Kemerovo, Tomsk, Barnaul) use river (sometimes, artesian) water for water preparation. The quality of water in a large river such as the Ob is determined by the natural and anthropogenic factors. One of the natural factors is the large drainage area with a diversity of natural complexes. Among anthropogenic factors are sources of pollution at enterprises of coal and oil producers and refineries, and large coal and oil production centers: Nizhnevartovsk, Surgut, and Khanty-Mansiysk situated downstream [1]. There are metallurgical works of the Ore Altay (East Kazakhstan region) located along the upper course of the Irtysh, and ore manifestations of copper, silver, and complex ores are found along the upper reaches of the Aley nd Charysh rivers.

In the upper Ob, the quality of water is strongly affected by the Novosibirsk reservoir. In the middle course of the river, the Tom' and Chulym tributaries change the quality of water. Large tributaries such as Ket', Vasyugan, Tym, and Irtysh carry chemical wastewaters in the lower portions of the Ob river.

Small tributaries such as Ul'ba (Ust'-Kamenogorsk), Aley, Barnaulka (Barnaul), Inya (Novosibirsk), and Ushayka (Tomsk) join the Ob and Irtysh Rivers and their large tributaries at different points; although their water content is low, they are subject to a considerable anthropogenic load, especially during spring flood.

The aim of the present work is comparative analysis of water monitoring data for the Ob river within the main groups of chemical pollutants (heavy metals and organic substances) using the results of our own studies (1992–2002, the middle and lower Ob') and literature data.

PROCEDURE

The river bottom was echosounded at test section lines (Fig. 1), and hydrometric investigations were carried out using three velocity verticals and three horizons with concomitant sampling of water, suspension, and bottom sediments (BS). The sludge liquor and sediment runoff were calculated from the flow rate. The integrated hydrochemical characteristics: pH, Eh, conductivity, oxygen solute (biochemical oxygen demand BOD_5) and the total content of organic matter were monitored concurrently. In the water samples, the total hydrochemical characteristics were determined directly at the sampling site; pH, Eh, and O_2 were measured using a Multilane 4 field multimeter equipped with CellOx 325 and SenTix 41-3 (Germany) immersion sensors. At test river stations in still-water riverside regions of the Ob', additional samples of bottom sediments were taken in 1999 and 2001 from depths of 40–100 cm.

The Cu, Pb, Cd, and Zn contents in natural water were determined under laboratory conditions by atomic absorption spectrometry with electrothermal atomization (AAS 30 ETA) or by inversion voltammetry (TA-2 computerized volt-ampere analyzer, Tekhnoanalit, Tomsk Polytechnical University, Tomsk). Analysis for heavy metals (HM: Fe, Mn, Cu, Pb, Cd, Zn) in bottom sediments, core samples (sectioning), and suspensions was carried out after wet air oxidation by atomic absorption spectrometry (AAS 1N). Organic matter, phenols, and oil hydrocarbons were determined by the standard procedures [2-4].

RESULTS AND DISCUSSION

Water

The upper reaches of the Katun' and Chuya rivers were explored in detail by the institutes of the Siberian Branch, Russian Academy of Sciences (1989–1994), in efforts to investigate the distribution of mercury from the Aktash and Chagan-Uzun deposits and ore manifestations [5]. The total content of mercury in water was approximately 1.0 μg/l near the Aktash deposit and 0.01–0.21 μg/l in the estuary of the Chuya. This figure decreased away from the deposits to 0.08–0.17 μg/l (section line near the Inya village) and to background values (0.005–0.09 μg/l) at the closing section lines (Elikmonar village, Anos). According to the main ion contents, the water of the Katun' river basin probably belongs to the hydrocarbonate-calcium-magnesium type [6, 7]. According to the data (1989–1991) of the Institute of Water and Environmental Problems, SB RAS (IWEP SB RAS), permanganate oxidizability (PO) in the rivers of the Katun' basin was 0.5–1.3 mg of O/l, bichromate oxidizability (chemical oxygen minimum, COM) was 10–15 mg of O/l, and the biochemical oxygen demand (BOD_5) was 0.3–1.8 mg of O/l. The hydrochemical characteristics were liable to hydrological seasonal fluctuations. During the summer floods, overall mineralization increased consistently with the fluid and solid runoffs; low water runoff in summer and autumn led to increased concentrations of organic substances. Based on O. A. Alekin's concept [8] that PO is the concentration of "readily oxidizable" organic substances and that COM is the content of "hardly oxidizable" organic matter, one can state that the headwaters of the Ob have low or medium contents of organic substances.

Systematic hydrochemical studies on the river segment from Biysk to Kamen'-na-Obi have not been conducted. The data available include scanty information about the impact of the city of Barnaul (Altay State University, IWEP, Sanitary and Epidemiological Inspection). These data indicate that the general hydrochemical characteristics are stable throughout all hydrological seasons of the year and that the total contents of the soluble forms of heavy metals (Hg, Ag, Pb) are lower than those in the basin of the Katun' and Chuya rivers. The left tributaries Aley and Barnaulka produce minor effects on the chemical flow of metals, inflowing with copper and zinc (Aley) and iron (Barnaulka), the effects being especially pronounced during spring flood [9].

Distribution of heavy metals in water along the river

The results of HM determination in the natural waters of the middle and lower Ob (Fig. 2, *a*–*f*) show that the dissolved Fe, Mn (see Fig. 2, *e*, *f*) and other HM forms in the surface waters approximate the upper bound of background contents [10, 11] for unpolluted waters. The spatial distribution of the solute Fe forms in the surface waters of the middle and lower Ob reflect the influence of the tributaries. Thus pronounced concentrations

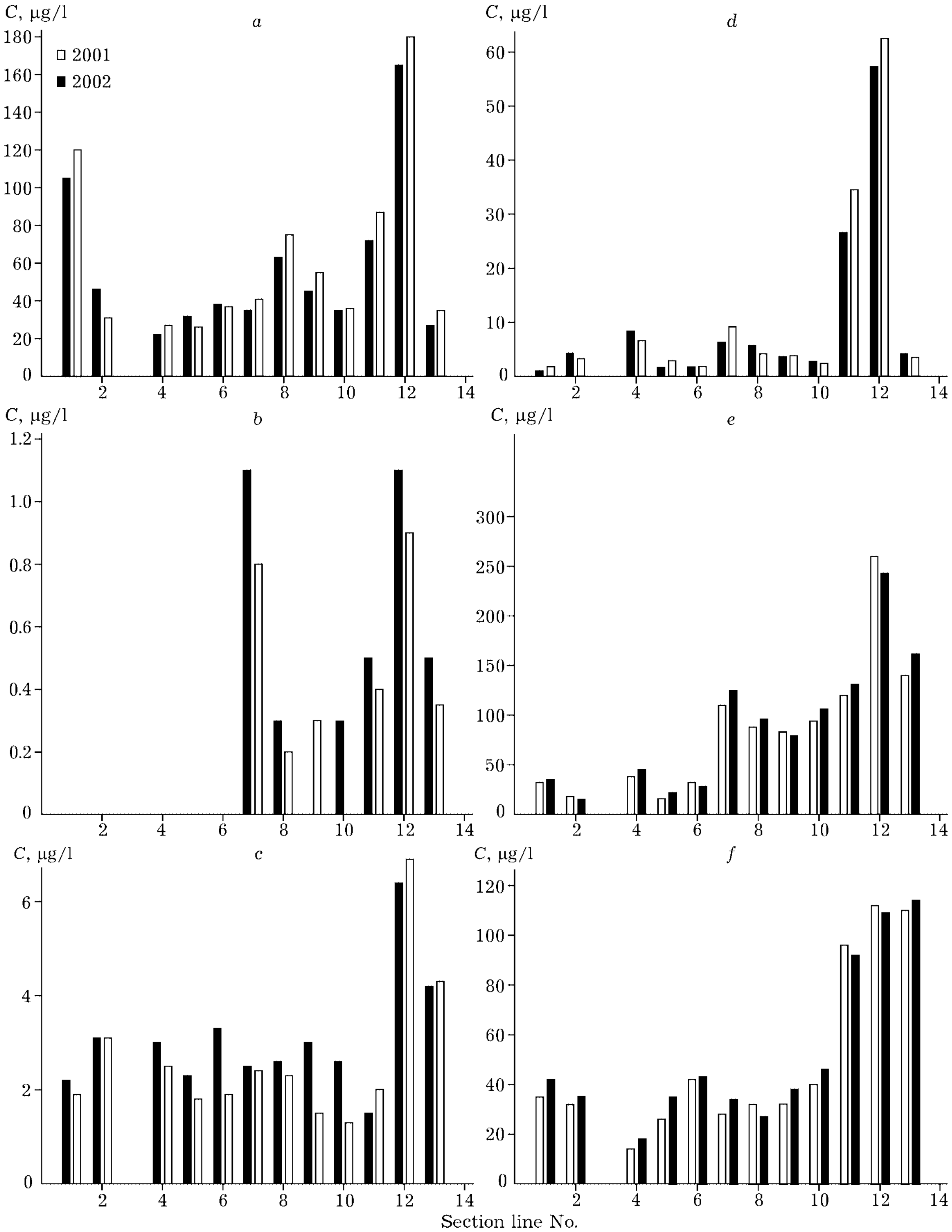


Fig. 2. Distribution of heavy metals along the river.

were revealed on section line *8* (below the junction with the Chulym) and on section lines 7 and *12* (below the delta of the Irtysh) (see Fig. 2, *e*). The concentration of the solute manganese on section line *11* in the Ob is approximately doubled (see Fig. 2, *f*), then its level remains constant.

The largest concentrations of the solute forms of ion (manganese) were found below the junction with the Irtysh (see Fig. 2, *e*, *f*). They correspond to the MPCw for industrial and household waters and exceed the existing fish industry standard by a factor of 6 and 10, respectively [3].

The concentration of the solute forms of Cu and Zn (see Fig. 2, *a*, *d*) was 2–63 and 26–180 μg/l, respectively, and steadily exceeded the background values for world's surface waters by a factor of ~10. For the test section line below the delta of the Irtysh, the maximal figures were 180 μg/l (Zn) and 63 μg/l (Cu), which exceeded the MPCw.f. (for fish ponds) by a factor of 18 and 63, respectively [3].

The highest concentrations of cadmium were found below the junctions with the Chulym (0.8–1.1 μg/l) and Irtysh (0.9–1.1 μg/l) on the middle and lower Ob (see Fig. 2, *b*). These values are close to the MPCw and are two times higher than the background indices for the world's river waters [10, 11].

The content of the solute lead in the Ob was determined in the low water in autumn 2001 and 2002 (see Fig. 2, *c*). It was within the limits of the existing norms and corresponded to the background level for river waters. The maximal quantity of the solute forms of lead was found below the junction with the Irtysh. The HM concentrations found in the Ob water in 2001 generally agree in magnitude with the results of monitoring studies of 2002 and earlier (1999, 1997 [12]) and reflect the impact of the lateral tributaries (the Tom', Chulym, and Irtysh rivers).

Suspended matter

Mercury is transported together with suspended matter (SM) from the estuary of the Chuya (0.3–18.9 mg/kg) in the upper Ob, its content decreasing to background values of 0.04–5.6 mg/kg away from the mercury deposits. In Katun', suspended matter accumulates elements that form natural associations related to the deposits and ore manifestations of the mercury (Hg, Ag, Zn, Cu) and lead-zinc (Pb, Zn, Cu, Ag) formations. In Katun's solid runoff, mercury is found mostly in its sorption form. In the suspension of the Katun' river, the specific concentrations of other chalcophile elements (Pb, Zn, Cu, Sn) exceed several fold those in world's clays and shales [5]. These elements mostly come to the channel net from the deposits of the mercury, polymetallic, and other ore formations and from the deep break zone of the Mountainous Altay region.

The Katun' and Chuya rivers are fed by glaciers (70–80 %). In summer, actively melting glaciers of the North-Chuya and Katun' ridges of the Altay mountains can bring with them not only HM of natural origin, but also sorbed forms of HM accumulated in them and coming to the channel net of these rivers from anthropogenic sources. In the glaciers of the basin of the Aktru river (North-Chuya ridge), the Pb, Zn, and Cu contents were much higher than the background values [13]. According to the observations of 1998–2000, seasonal snow cover contained up to 10–15 μg/l Pb. Isotope analysis of core samples from Altay's glaciers [14] showed that the lead isotope ratio was identical to that in the urban snow samples collected near the lead and zinc industrial complex in Ust'-Kamenogorsk.

After the confluence of the Biya and Katun' rivers away from the mountain province, the flow rate decreases, and saprogenic organic matter appears in the river along with fish and hydrobionts. In the region of Kamen'-na-Obi in the upper part of the Novosibirsk reservoir, where the flow is regulated, sedimentation of suspended matter and active formation of bottom sediments take place. In the reservoir, the river water is cleared from sus-

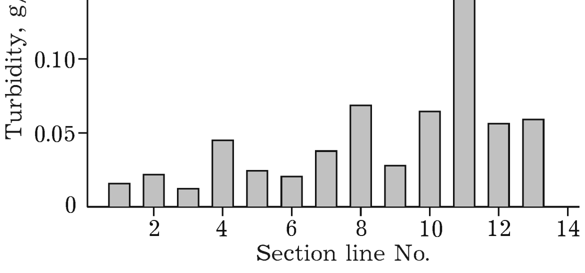


Fig. 3. Distribution of turbidity along the river in 2002 (suspension).

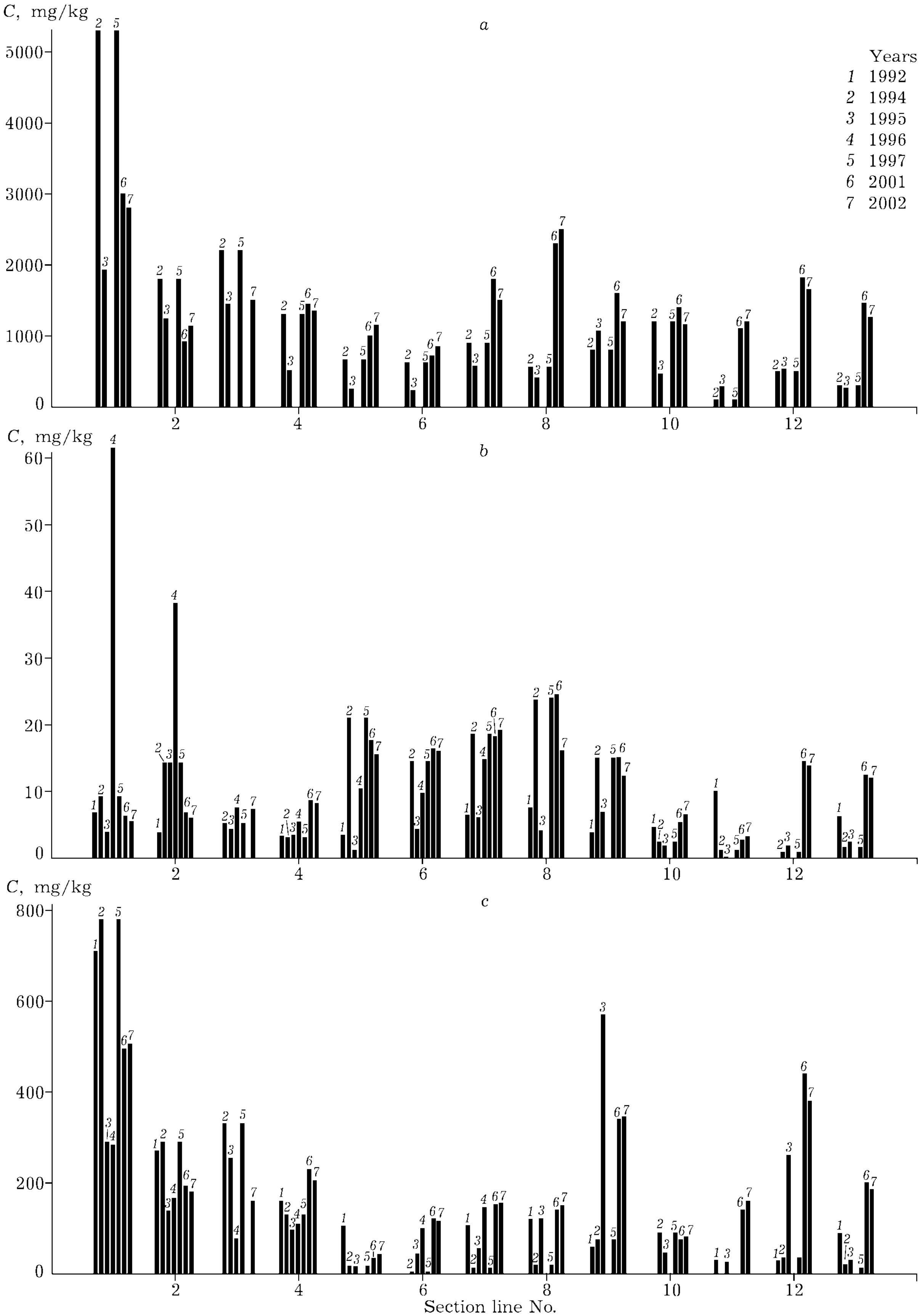
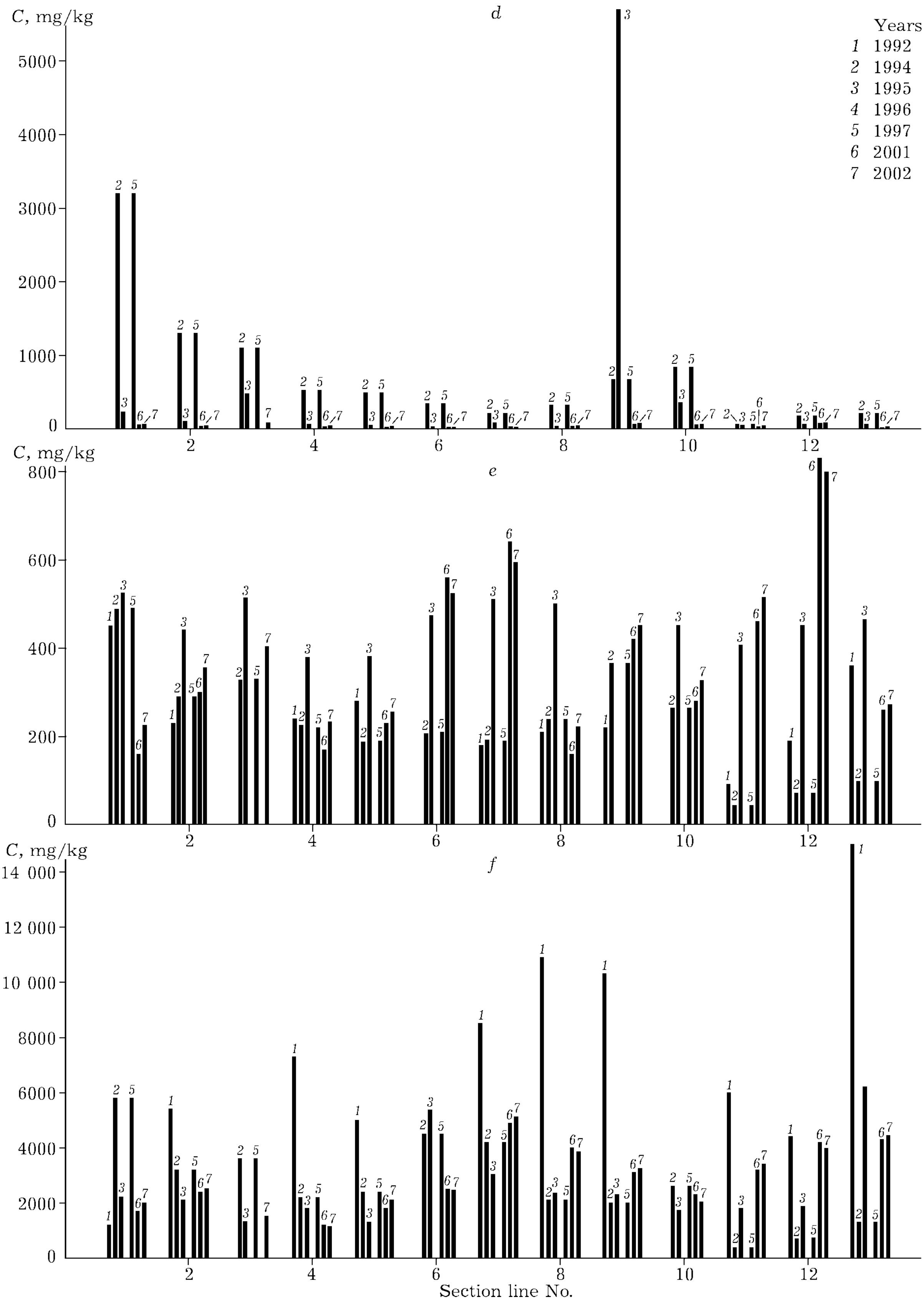


Fig. 4. Distribution of Zn (*a*), Cd (*b*), Pb (*c*), Cu (*d*), Fe (*e*), Mn (*f*) along the river (suspension).


d
C, mg/kg
5000
4000
3000
2000
1000
0
Years
1 1992
2 1994
3 1995
4 1996
5 1997
6 2001
7 2002
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C, mg/kg
800
600
400
200
0
f
C, mg/kg
14 000
12 000
10 000
8000
6000
4000
2000
0
2
4
6
8
10
12
Section line No.

pensions. Figure 3 shows the typical distribution of SM in the Ob downstream of the reservoir, where decreased turbidity of water due to silt sedimentation in the reservoir was also reported previously [15].

It can be seen that the spatial distribution of suspension is affected by the lateral tributaries (section lines: *4* – Tom', *8* – Vakh, *11* – Irtysh). The concentrations of SM increase in the lower course, but stay within the limits of the existing norms for the low water of summer and autumn. SM in general is a natural indicator to the HM contents in the river's ecosystem because it sorbs the maximal quantities of HM, while at the same time this is the mobile part of the sediment load of the Ob. These results may be represented as diagrams of HM distribution along the river (Fig. 4, *a–f*). Apart of HM, the suspension has organic matter sorbed on it, whose spatial distribution is depicted in Fig. 5. For iron (manganese), everywhere (section lines *7–10*) the concentrations were two or three times higher than the abundance ratios for clays [8], especially on those segments where the river flows through marshlands with intense supply of HM both from the drainage area and from the large tributaries (Ket', Vasyugan, Tym, Vakh). The sediment runoff on this segment also probably includes a contribution from the oil and gas industrial complex.

Figure 4 (*b*, *e*, *g*) reflects the impact of the marshland segment of the river on the distribution of iron, manganese, and organic carbon (section lines *7–10*). In the Fe and Mn contents the impact of marshland territories is comparable to the effect of the Irtysh on the water of the Ob. The spatial distribution of Pb is largely anthropogenic in character (see Fig. 4, *c*). The increased concentrations of Pb on section lines *1–4*, *9* reflect the impact of the cities (Novosibirsk, Tomsk, Nizhnevartovsk), which is comparable to the effects of the Irtysh (section line *12*).

The spatial distribution of Cd is shown in Fig. 4 (*b*). One can observe a relative increase in the metal concentrations on section lines *7–9* due to the suspended forms of cadmium which the Chulym, Ket', Vasyugan, Tym, and Vakh tributaries bring to the waterlogged segment of the river. The increased concentration of Cd is more pronounced for the water-soluble forms (see Fig. 2, *b*). The distribution of Cu and Zn presented in Fig. 4, *a*, *d* differs from that of Fe and Mn in the increased content. The increased contents of these metals on section lines *1*, *3*, *4*, *8*, and *12* are also associated with hydrobionts that ended their year's life cycle and were brought to the river together with SM with autumn low water. The anthropogenic influence of large cities, *e. g.*, Nizhnevartovsk, cannot be excluded either (see Fig. 4, *d*; section line *9*).

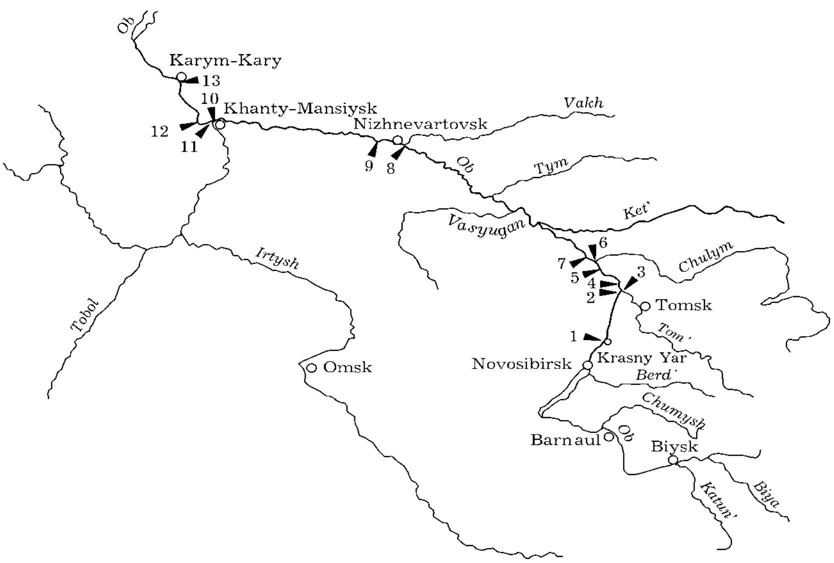

Fig. 5. Distribution of organic matter along the river (suspension).

Thus the results of these studies showed that the large tributaries of the Ob such as Tom', Chulym, and especially Irtysh have the most pronounced effect on the formation and distribution of the chemical flow of HM in the middle and lower Ob.

The basin of the Tom' river brings not only HM to the Ob, but also oil products, phenols, polyaromatic and other hydrocarbons, and the mineral forms of nitrogen [16, 17]. The chemical flow of the Tom' generally increases during the spring flood when active snow melting brings chemical substances including metals from the basin to the channel net of the Tom' [18] and Ob. During spring flood, this HM washout effect amounts to 15–20 %. Profound anthropogenic effects of the chemical flow were reported for small rivers near large cities, for example, for the Barnaulka near Barnaul [9]. However, the total inflow of small rivers such as Barnaulka into the Ob is generally marginal (1–2 %).

Regional accumulation of chemical substances was reported for closed drainage territories of the Ob-Irtysh basin. Thus increased concentrations of organic pollutants (phenols, oil products, and pesticides) and HM were observed over years in the Ishim region near Irtysh [19]. Regional excess of mineral substances was observed in closed drainage of the Kulunda steppe [20]. On this territory, the HM contents are insignificant compared with the increased natural background of the mineral salts. An exception is cadmium, which accumulates in the water objects (suspended matter, bottom sediments) in the delta of the Kulunda as a result of farming activities.

Bottom sediments

In the upper reaches of the Katun' river, mercury is unevenly redistributed into bottom sediments. The metal concentration in them decreases by a factor of 5–7 from 0.7–1.5 mg/kg in the delta of the Chuya to 0.01-0.3 mg/kg in the lower Katun', which is lower than the abundance ratio of mercury (0.4 mg/kg) for clays and shales [6].

Bottom sediments on the segment from Biysk to Kamen'-na-Obi of the Ob river were little explored. Table 1 presents comparative data on BS for the Ob. Monitoring studies of HM contents in the bottom sediments of the Novosibirsk reservoir are described in detail in [21]. According to most HM, the bottom sediments of the reservoir proved to be more contaminated than the sediments of lakes in the Altay region. HM accumulate more actively in the lower portion of the reservoir due to sedimentation of smaller fractions of SM [21].

The authors of the present work and members of the Trofimuk United Institute of Geology, Geophysics and Mineralogy, SB RAS, examined the bottom sediments of the middle and lower Ob during the period from 1999 to 2001 (see Table 1).

As can be seen from Table 1, the BS of the Novosibirsk reservoir accumulate nearly all of the stated HM, due to which the concentrations of the latter decrease several fold compared with the BS of the middle and lower Ob. The only exception is cadmium. The accumulation of cadmium in lakes and reservoirs is probably affected by the general mineralization of water and concentration of organic matter. Previously, the BS of ponds in the Kulunda depression were reported to contain up to 1 mg/kg cadmium [20]. The 1999 data indicate that the river flow (flow rate ~0.5 m/s) decreases metal accumulation, on the average, by a factor of two or three irrespective of the nature of the metals. These differences in accumulation of HM are due to the lower rate of sedimentation of small fractions of SM.

Apart from concentration, HM accumulation is affected by mineralization, temperature, the presence of organic matter, pH, Eh, (5–370 mV), and solute oxygen concentration (0.02–1.09 mg/l) in the interstitial waters of the BS of the middle and lower Ob, which is, on the average, 100 times smaller than on the midstream verticals of the test section lines.

The physicochemical distribution of HM on the interface BS/water may be quantitatively expressed by the concentration coefficients, for example, by the molar concentration coefficient. The latter was calculated from analytical data: K_c = HM(BS) [mmol/kg]/HM(W + SM) [mmol/l]. For five samples from the upper layer 5 cm thick for BS core samples collected in 2001, the values of the coefficients K_c change in relation to the nature of the metal: 200–

TABLE 1

Heavy metal contents in the bottom sediments of water objects in the Ob-Irtysh basin

Statistical characteristic		Fe/100	Mn/10	Zn	Cu	Pb	Cd
			Lower part of the Novosibirsk reservoir [21]				
n = 60	max.	–	159.2	140	49	56	0.260
	min.	–	48	37	15	5	0.024
	av.	–	92.7	96	33	22	0.011
			Middle and lower Ob (midstream verticals), 1999				
n = 12	max.	95	89.2	71.1	12.1	6.8	0.48
	min.	12.5	11.2	5.6	4	0.03	0.12
	av.	35.4	28.4	24.8	5.9	1.3	0.24
ε		±16.1	±12.6	±11.4	± 1.6	±1.26	± 0.16
			Middle and lower Ob (still-water places), 1999				
n = 27	max.	282.04	87.5	281	28.5	7.2	0.58
	min.	71.31	11.7	29.5	5.4	0.66	0.11
	av.	164	46.6	79.8	14.6	2.62	0.37
ε		±25.4	±7.05	± 32	± 2	± 0.74	± 0.4
			Middle and lower Ob (still-water places), 2001				
n = 31	max.	155	37	24	25	3.7	0.38
	min.	8.4	8	1.7	2	0.2	0.4
	av.	66.5	16.7	9.6	14	1.2	0.10
ε		±11.9	±2.4	±1.6	±2.4	± 0.27	±0.23
			Basin of the Irtysh, 2000 [22]				
n = 284	max.	5870	1039	12690	4061	4138.5	178.9
	min.	190	1.0	21.2	11.6	10.8	0.2
	av.	1040	91.0	1223	230	251.7	15.3
ε		±140	±14.5	±217	±29.2	±47.9	±2.3
Background [10]		5–100	10–50	5–50	15–60	15–50	0.1–10

Note. n is the number of samples, and ε is the confidence interval.

8800 for Fe, 400–3800 for Mn, 600–8000 for Cd, 2–22 for Pb, 186–7500 for Cu, and 35–150 for Zn.

The calculated coefficients may be arranged as follows: Fe ≈ Cd ≈ Cu > Mn >> Zn > Pb. Zinc and lead are the end members of the series; for these metals, water-soluble forms make a considerable contribution to the total concentration of the metal in water. As shown by the data of yearlong observations, no significant excess has been found over the abundance ratios of these metals in the BS of the middle and lower Ob.

Comparing the results presented in Table 1, one can see that the increased amounts of suspended Pb forms on section line *1* (see Fig. 4, *c*) is due to the increased content of small SM fractions in the tail water of the reservoir, determining the distribution of these forms in the BS of the lower part of the latter. Increased concentrations of all HM in bottom sediments in the delta of the Irtysh and below its junction with the Ob were attributed [22] to the far greater anthropogenic load (according to HM) on the basin of the Irtysh than on the middle Ob. Thus small fractions of SM of the Irtysh river are an additional source of HM in the lower Ob. At the junctions with tributaries and in regions of large oil producing centers the bottom sediments contain more HM than do sediments (on the average) all over the Ob below the reservoir; the corresponding figures can serve as indicators of the anthropogenic pollution of the ecosystem of the river on these segments.

CONCLUSIONS

Analysis of changes in the concentrations of pollutants coming to the Novosibirsk reservoir with the upper Ob flow permits one to speak about the positive role of the latter in self-purification of the river waters. It revealed that heavy metals mostly travel with the small fractions of suspended substances and settle in the lower lake-like part of the reservoir.

The hydrochemical runoff of the middle and lower Ob is most seriously affected by the large tributaries: Tom', Chulym, Tym, Vakh, and especially Irtysh, as well as by oil and gas producing centres: Nizhnevartovsk, Surgut, and Khanty-Mansiysk.

Water pollution of the Ob below its junctions with tributaries and near industrial centers increases dramatically, leading to considerable changes in the species diversity of hydrobionts and to accumulation of chemical elements including heavy metals (iron, manganese, zinc, copper, lead, and cadmium) in water and bottom sediments.

Currently, oil pollution of rivers is of special concern especially because the areal of pollution has been expanding steadily, covering not only the middle and lower Ob, but also many segments of the Irtysh and its tributaries. Under conditions of moderate temperatures of air and water in northwestern Siberia, decreasing self-purification, this is the most serious (leading to adverse consequences) man-caused factor for river ecosystems.

Further hydrological and hydrochemical monitoring should include work on current greatest challenges associated with finding segments with increased ecological load on the Ob and its tributaries. This will make it possible to take appropriate decisions and to develop preventive measures to avoid crises in the ecological state of water.

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