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Contamination of the Lake Baikal Basin by Organochlorine Pesticides and Polychlorinated Biphenyls: the Gusinoye Lake

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Abstract

In the bottom sediments and surface water of the Gusinoye Lake (the Lake Baikal basin) there were content and profile values for organochlorine pesticides and polychlorinated biphenyls determined. An evaluation was performed concerning the ways of coming thereof in the Gusinoye Lake It has been found that the sources of these ecotoxicants are mainly local.

Key words: organochlorine pesticides, polychlorinated biphenyls, the Lake Baikal basin

INTRODUCTION

Organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) represent especially hazardous ecotoxicants those exhibit a pronounced mutagenic and carcinogenic activity, high bioaccumulation level in organisms, high toxicity and environmental persistence.

Organochlorine pesticides such as dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCCH), due to the pronounced insecticidal properties, were widely used in Russia from 1940 to 1970. They were also actively used as heat sink fluids in electrical equipment, metalworking fluids, as well as in the manufacture of plasticizers, pesticides, paints, lacquers [1].

The Gusinoye Lake is the largest lake after the Lake Baikal in the Western Transbaikalia. The water catchment area of the lake basin is equal to 924 km^2 and the maximum depth thereof amounts to 25 m. The lake is about 25 km long, the maximum width being of 8.5 km. The Bain-Gol River flowing from the southeastern part of the lake flows into the Selenga River, the main tributary of the Lake Baikal [2].

The anthropogenic load on the Gusinoye Lake is significant: the Gusinoozersk Thermal Power Plant (TPP) located on the shore of the lake consumes 90 % of the total water take-off with respect to the surface waters of the Republic of Buryatia. In addition to the TPP heat discharges, into the lake there are discharged purified wastewaters from the Gusinoozersk TPP, as well as waste water from Vodokanal Municipal Enterprise of the Gusinoozersk, from the Gusinoye Lake Ltd. On the shores of the lake there are other sources of man-caused contamination: the Gusinoozersk City with the population of about 25 thousand people, railway station and the Gusinoye Lake Settlement, fuel storage, as well as a currently closed coal mine and an open-cast mine with the piles of rocks.

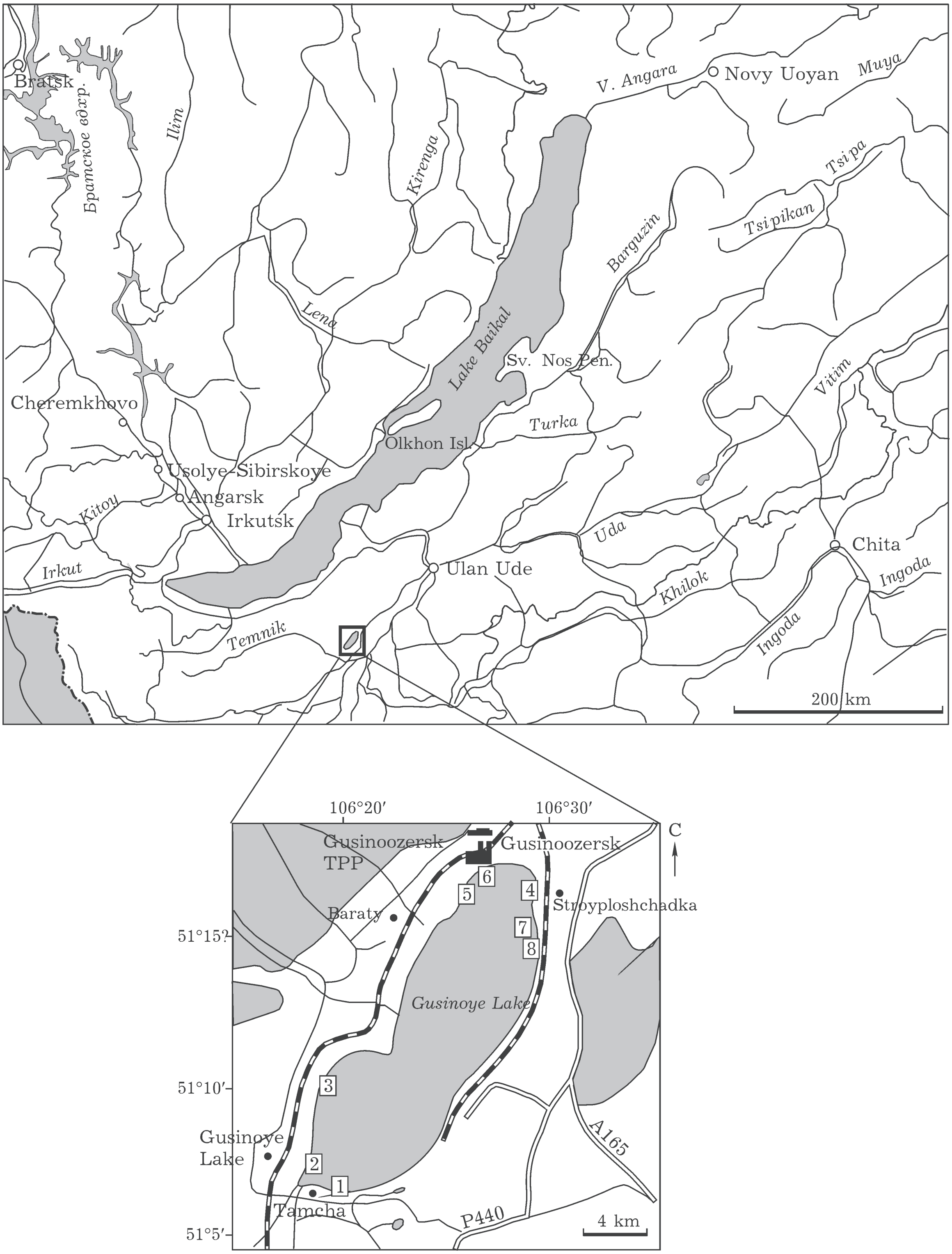

Fig. 1. Schematic map of stations for sampling the experimental material: 1 – Tamcha settlement, 2, 3, 5, 7, 8 – Gusinoye Lake stations, 4 – Stroyploshchadka settlement, 6 – Gusinoozersk TPP.

Earlier, no investigation of the contamination with OCPs and PCBs concerning the Gusinoye Lake and the sources of inflowing them to the basin of the Gusinoye Lake was carried out.

The purpose of this investigation consisted in determining of the present-day levels of OCPs and PCBs in surface water, bottom sediments and the sources of inflowing thereof to the ecosystem of the Gusinoye Lake involved in the environmental buffer zone of the Baikal natural territory.

EXPERIMENTAL

The sampling of surface water and bottom sediments was performed in the autumn of 2009. The map of the location of stations for sampling bottom sediments and surface water is presented in Fig. 1.

The samples of surface water were preserved *via* adding a special purity grade concentrated HCl taking 1 mL of the acid per 1 L of solution. The bottom sediment samples were taken from the surface layer up to 10 cm thick using a Wildlife Supply Co. (USA) bottom sampler. The samples were dried at a room temperature, sieved through a sieve with a mesh size of 0.4 mm.

The preparation of bottom sediment samples (5 g) and of surface water samples (2 L) consisted in the extraction of PCBs and OCPs with methylene chloride and purification of the extracts obtained *via* passing through a column packed with activated alumina. For the analysis of the extracts we used a gas chromatography-mass spectrometry technique (Agilent Technologies 6890N GC) with an AT 597 5N mass selective detector and an AT 7683B autosampler) in the mode of detecting the individual characteristic ions of the compounds under determination. The sample components under analyzing were separated using a HP-5MS quartz capillary column 30 m long with a diameter of 0.25 mm and a coating thickness equal to 0.25 μm. The conditions of gas chromatographic determination were as it follows: detector temperature 280 °C, ion source temperature 230 °C, the quadrupole unit temperature 150 °C, helium being used as the carrier gas. The column oven temperature increased from 50 (holding time 2 min) up to 280 °C (holding time 20 min) at a rate of 10 °C/min. The delay of turning cathode ion source on for missing the solvent peak of was equal to 5 min.

For the quantitative determination we used standard reference samples of the mixtures of pesticides and PCBs: (NR No. 8500-6011, GSO 7821–2000), and as a surrogate standard (witness) we used 4,4′-dibromobiphenyl; the accuracy of determination was better than 20 %; the detection limit being of 0.02 ng/g in the case of analyzing 5 g of bottom sediment, and 0.05 ng/L in the case of analyzing 2 L of water, respectively.

In the resulting extracts we determined the following compounds: *p,p*′-DDT and its metabolites: *p,p*′-dichlorodiphenyldichloroethane (*p,p*′-DDD), *p,p*′-dichlorodiphenylethylene (*p,p*′-DDE); HCCH (α-HCCH, β-HCCH, γ-HCCH, δ-HCCH), hexachlorobenzene (HCB), dieldrin, aldrin, endrin, and the sum of PCBs.

RESULTS AND DISCUSSION

Resulting from the investigations, we determined current OCPs and PCBs contamination levels in the Gusinoye Lake ecosystem. Tables 1 and 2 demonstrate data concerning the content of these toxicants in the bottom sediments and surface waters of the Gusinoye Lake.

A comparative analysis was performed with respect to the results obtained and the literature data for other aquatic ecosystems of the Earth (Tables 3, 4).

Hexachlorocyclohexane

Hexachlorocyclohexane is one of common insecticides used in Russia for combating against various noxious insects, including the use thereof as an additive for insecticidal seed protectants in order to protect seedlings against soil pest damage.

The total concentration of HCCH (α-HCCH and γ-HCCH) in bottom sediments (see Table 1) ranges within 0.2–8.9 ng/g of dry solid matter (mean 3.1 ng/g). The values obtained correspond to the level of concentration inherent in slightly contaminated Arctic areas (see Table 3).

TABLE 1

OCPs and PCBs content in the samples of bottom sediments, ng/g

Compounds	Sampling stations							
	A1	A2	A3	A4	A5	A6	A7	A8
α-HCCH	n/d	1.2	n/d	0.5	0.6	0.2	0.1	n/d
β-HCCH	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
γ-HCCH	2.3	5.2	1.5	1.5	8.3	1.2	2.0	0.2
δ-HCCH	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
ΣHCCH	2.3	6.4	1.5	2.0	8.9	1.4	2.1	0.2
p,p′-DDT	0.3	0.8	n/d	n/d	1.3	1.0	0.5	n/d
p,p′-DDE	0.4	0.8	0.2	0.7	2.3	0.5	2.3	0.4
p,p′-DDD	0.2	0.4	n/d	0.8	0.6	0.6	1.5	0.1
ΣDDT	0.9	2.0	0.2	1.5	4.2	2.1	4.3	0.5
HCB	2.5	4.1	3.0	4.0	8.1	3.4	4.5	2.7
Dieldrin	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
Aldrin	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
Endrin	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
ΣOCP	5.8	12.4	4.7	7.4	28.6	8.7	10.9	3.3
ΣPCB	5.2	6.6	2.5	4.7	21.6	17.9	16.6	2.0

Note. n/d – not detected, below the detection limit (0.02 ng/g).

TABLE 2

OCPs and PCBs content in surface water samples, ng/L

Compounds	Sampling stations							
	A1	A2	A3	A4	A5	A6	A7	A8
α-HCCH	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
β-HCCH	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
γ-HCCH	2.0	3.7	0.9	0.6	0.7	1.0	0.6	1.0
δ-HCCH	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
ΣHCCH	2.0	3.7	0.9	0.6	0.7	1.0	0.6	1.0
p,p′-DDT	0.4	1.1	0.3	0.2	0.3	0.4	n/d	0.6
p,p′-DDE	1.1	1.9	0.4	0.3	0.4	0.5	n/d	1.4
p,p′-DDD	0.3	1.4	0.5	0.1	0.4	0.3	n/d	0.7
ΣDDT	1.8	4.4	1.2	0.6	1.1	1.2	n/d	2.7
HCB	1.8	2.3	0.5	0.6	0.5	0.6	0.3	0.6
Dieldrin	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
Aldrin	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
Endrin	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
ΣOCP	5.6	10.4	2.6	1.8	2.3	2.8	0.9	4.3
ΣPCB	14.2	4.7	2.2	4.6	1.1	1.9	1.2	3.6

Note. n/d – not detected, below the limit of detection (0.005 ng/L).

The total HCCH concentration in the surface water ranges within 1.1–5.1 ng/L (mean 2.2 ng/L). In the samples of bottom sediments and surface water, there were no isomers such as β-HCCH and γ-HCCH detected (see Tables 1, 2).

As it can be seen from Table 4 the concentration of ΣHCCH in surface water exceeds the analogous data for the Arctic regions, being comparable the data for the Bosumtwi Lake, Ghana (0.8–1.6 ng/L). The values obtained do not exceed the MPC level acceptable for water reservoirs for drinking and community water use (20 μg/L) and for the reservoirs of fishery significance (0.01 μg/L) [10, 11].

The presence of HCCH in the samples of bottom sediments and surface water is caused, in our opinion, by an intensive use of HCCH in agriculture in the basin of the Lake Baikal in the past.

Dichlorodiphenyltrichloroethane

Dichlorodiphenyltrichloroethane in Russia was begun to produce and use in 1946, it was used to combat against various insects, the carriers of infectious diseases. In 1970, DDT was excluded from the list of pesticides allowed for using in Russia. Until the end of the 1980s, it was still used in many areas of Russia to prevent spreading the malaria and encephalitis.

The composition of the technical DDT preparation includes *p,p′*-DDT (65–80 %), *o,p′*-DDT (15–21 %), *p,p′*-DDD (4.0 %), 1-(*p*-chlorophenyl)-2,2,2-trihloroethanol (1.5 %) [17].

The total concentration of DDT compounds in bottom sediments of the Gusinoye Lake is insignificant ranging from 0.2 to 4.2 ng/g (see Table 1) (the mean value being of 2.2 ng/g). These results are comparable with the earlier data for the bottom sediments of the Lake Baikal (0.01–2.7 ng/g) [18] and the coast of Norway (0.2–5.4 ng/g), but being much lower than the maximum values established for the Saronic Gulf, Greece (9100–75 600 ng/g) (see Table 3).

The total concentration of DDT in the surface water is equal to 0.6–4.4 ng/L (mean 1.6 ng/L) being comparable with the concentrations obtained for lakes in Vietnam (0.2–15.3 ng/L) (see Table 4). The values obtained do not exceed the MPC level acceptable for water reservoirs for drinking and community water use (100 μg/L) and water reservoirs of fishery significance

TABLE 3

Comparison of OCPs and PCBs content in the bottom sediments of the Gusinoye Lake contaminated bottom sediments of other aquatic ecosystems

Sampling place	Year	Content (mean value), ng/g of dry solid matter				Ref.
		ΣHCCH	ΣDDT	HCB	ΣPCB	
Gusinoye Lake	2009	0.2–8.9 (3.1)	0.2–4.2 (2.2)	0.2–8.1 (4.0)	2.0–21.6 (9.6)	
Ny-Elesund Lake, Norway	2005	0.8–6.5 (3.6)	0.2–5.4 (2.2)	n. d.	0.2–13.0 (3.1)	[3]
Novaya Zemlya, the Sinyeye Lake, Lebedinoye Lake	2006	n/f	0.1–0.1	n. d.	2.1–3.4	[4]
Kola Peninsula lakes	2006	n/f	0.5–27.7	0.4–0.9	11.2–37.7	[4]
Greenland Sea, Gren-fiord Bay	2006	0.3–0.4	0.6–6.1	0.3–0.4	5.1–5.6	[4]
White Sea	2006	0.02–0.9	0.1–5.2 (1.8)	n. d.	2.3–7.2 (4.1)	[5]
Baltic Sea, Sweden	2007	0.1–6.4 (0.8)	0.04–52.4 (3.8)	n. d.	0.01–15.4 (1.5)	[5]
Taihu Lake, East China	2000	n. d.	n. d.	0.06–9.69	n. d.	[6]
Saronic Gulf, Greece	1998	100–300 (200)	9100–75 600 (47 090)	300–2800 (1810)	47.8–351.8 (168.7)	[7]
Barents Sea	1997	0.1–0.7 (0.3)	0.3–36.7 (10.5)	0.3–1.8 (1.1)	1.1–37.9 (12.8)	[8]
Salton Sea Lake, California	2000–2001	0.13–8.3 (2.3)	6.8–40.2 (22.4)	0.02–1.8 (0.5)	116–304 (217.2)	[9]

Note. n/f – not found; n. d. – no data.

(0.01 μg/L) [10, 11]. The total concentration of compounds belonging to the DDT group in the surface water does not exceed the total concentration of DDT isomers and metabolites in the surface waters of the majority of background areas in Russia (≤30 ng/L) (see Table 4).

The half-life of DDT in the environment is equal to 18–20 years. Nowadays, the most part of the DDT amount should convert into metabolites (DDD and DDE), mainly into the major metabolite DDE. To assess the character of DDT degradation and the time of the DDT entering to the Gusinoye Lake we used ratio values such as DDD/DDE and (DDE + DDD)/ΣDDT [19, 20]. The ratio value (DDD + DDE)/ΣDDT > 0.5 indicates that the DDT entering

TABLE 4

Comparison of OCPs and PCBs content in the surface water the Gusinoye Lake with the background contamination of surface water in other aquatic ecosystems

Sampling place	Year	Content (mean value), ng/L				Ref.
		ΣHCCH	ΣDDT	HCB	ΣPCB	
Gusinoye Lake	2009	1.1–5.1 (2.2)	0.6–4.4 (1.6)	0.3–2.3 (0.9)	1.1–14.2 (4.2)	
Background territories in Russia	2006	n.d.	30	n.d.	n. d.	[4]
Baltic Sea, Gulf of Finland	2006	n/f–0.4 (0.3)	n/f–0.7 (0.3)	n.d.	n/f–0.7 (0.3)	[4]
Selenga River	2007	1.0–27.7 (9.1)	3.3–163.3 (44.6)	n.d.	n. d.	[5]
Bosumtwi Lake, Ghana	2004–2006	0.8–1.6	0.2–1.8	n.d.	n. d.	[12]
Lakes of Yunnan, South China	2003	240–21 950 (7390)	90–1860 (270)	10–720 (110)	n. d.	[13]
Lakes of Vietnam	1998–1999	0.3–122.	0.2–15.3	n.d.	n. d.	[14]
Gaobeidian Lake, North China	2005	13.2–26.7	0.2–14.4	1.7–4.5	4.8–43.8	[15]
Mediterranean Sea, France	2001–2002	n.d.	n.d.	n.d.	0.5–11.3	[16]

Note. n/f – not found; n. d. – no data.

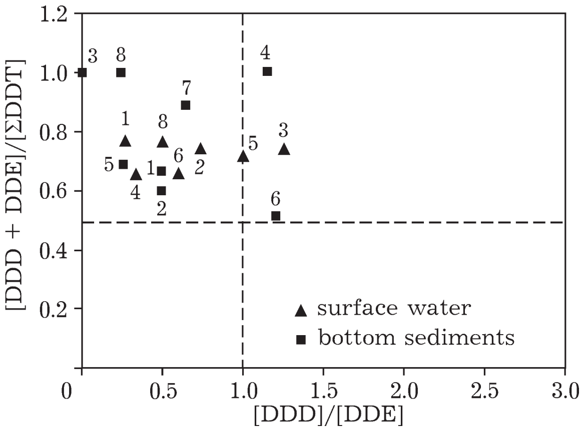

Fig. 2. Correlation between the concentration ratio values (DDD + DDE)/ΣDDT and DDD/DDE in the bottom sediments and surface water of the Gusinoye Lake. 1–8 – sampling stations.

mainly due to leaching and weathering the earlier entered DDT from the agricultural soils. The ratio value DDD/DDE < 1.0 is typical for the biological degradation of DDT mainly under aerobic conditions [19, 20]. The values of DDD/DDE and (DDD + DDE)/ΣDDT ratios exhibit varying between 0.3–1.2 and 0.6–0.9, respectively (Fig. 2).

The results obtained indicate that the biological degradation of DDT under aerobic conditions led to the DDT conversion mostly into the metabolites thereof such as DDD and DDE [19]. Indirectly, this also indicates the fact that there is a relatively high temperature of the lake water used for cooling the aggregates of the Gusinoozersk TPP, which temperature promotes an intense process of DDT degradation.

Hexachlorobenzene

Hexachlorobenzene was used in Russia until 1991 as a fungicide agent for the seeds of cereal crops in the mixtures with other fungicides to protect the cereals against the leaf smut. For the agricultural purposes, in Russia annually there were produces 120–150 t of hexachlorobenzene, which substance was a part of such preparations as gammahexane (30 % HCB), hexathiuram (30 % HCB), mercurbenzene, fagus, and so on. At the present time, it is used in the defense industry for the production of fireworks, in ferrous and nonferrous metallurgy, in pulp and paper industry, and as an intermediate in the preparation of chemicals. Hexachlorobenzene is thermally and chemically stable, so it can be remain intact for a long time in the soil to enter into the food chains of animals and humans and even to be present in the milk of lactating women [1]. The worldwide consumption of this substance is estimated to be equal to 1–2 million t [21].

The concentrations of HCB in the bottom sediments of the Gusinoye Lake (see Table 1) are quite high being comparable with the data for the Taihu Lake, East China (0.1–9.7 ng/g) (see Table 3), whereas the bottom sediments of the Lake Baikal exhibit the HCB content to be insignificant ranging within 0.01–0.2 ng/g [18]. It should be noted that the data obtained are to a considerable extent lower than the level of HCB in bottom sediments of the Saronic Gulf, Greece (300–2800 ng/g) (see Table 3).

The HCB content in the surface water ranged from 0.3 to 2.3 ng/L (see Table 2). The average concentration of HCB is equal to 0.9 ng/L. The values obtained are comparable with the data for the Gaobeidian Lake in North China (1.7–4.5 ng/L) (see Table 4), and do not exceed the MPC level acceptable for water reservoirs for drinking and community water use (0.001 mg/L) [10].

The presence of HCB in the samples of bottom sediments and surface water could be caused, in our opinion, by the HCB entering due to an explosion emergency at military ammunition depot near the railway station The Gusinoye Lake in July 2001, which led to a severe contamination of the lake ecosystem.

Dieldrin, aldrin, endrin

In the surface water and bottom sediments of the Gusinoye Lake, there were no these pesticides detected. The dieldrin was used in very small amounts (it was actively used in 1966–1967), whereas endrin was not used in Russia. Aldrin is banned for using in the Russian Federation since 1972. The dieldrin and endrin were not used in the Baikal region. For comparison, the dieldrin and endrin content in South China in the bottom sediments of the bays is high ranging within 0.1–214.7 and 0.1–39.5 ng/g, respectively [23].

Polychlorinated biphenyls

Polychlorinated biphenyls (PCBs) represent heavy high-boiling oily liquids with high dielectric properties. The PCB compounds are extremely inert being poorly soluble in water. The solubility level of PCBs in water decreases with increasing the number of chlorine atoms in the molecule [22].

In the USSR, the PCBs and preparations containing them were produced in large amounts 1934–1995. They were used mainly for the production of dielectric fluids sold under the trademarks of Sovol and Sovtol, as well as plasticizers in the manufacture of lacquers and polymeric materials, lubricants and fungicides for wood protection. The mixture of sovol and 1,2,4-trichlorobenzene entitled Sovol was made in different variants: Sovtol-1 (containing 75 % of sovol and 25 % of 1,2,4-trichlorobenzene); Sovtol-2 (64 % of sovol and 36 % of 1,2,4-trichlorobenzene); Sovtol-10 (90 % of sovol and 10 % of 1,2,4-trichlorobenzene). Furthermore, there were preparations released such as Trichlorobiphenyl (TCB) (85 % of sovol), Hexol (25 % of sovol and 75 % of hexachlorobutadiene) [1].

The total concentration of PCBs in the bottom sediments of the Gusinoye Lake ranges from 2.0 to 21.6 ng/g (see Table 1) (average value 9.6 ng/g). The values obtained are comparable with the data for the bottom sediments of the Barents Sea (1.1–37.9 ng/g), but they are much lower than the maximum concentrations of PCBs in the bottom sediments of the Saronic Gulf, Greece (47.8–351.8 ng/g) (see Table 3).

The total concentration of PCB in the surface water varies from 1.1 to 14.2 ng/L (mean value 4.2 ng/L) (see Table 2) being comparable with the values obtained for the northwestern Mediterranean Sea, France (0.5–11.3 ng/L) (see Table 4).

For all the samples, the concentrations of PCBs did not exceed the MPC level acceptable for water reservoirs with drinking and community water (0.001 mg/L) [10]. However, in the sample A1, taken near the Tamcha settlement, exhibits a maximum concentration of PCBs that exceeds the MPC level for water reservoirs having fishery significance (0.01 μg/L) [11].

Figure 3 demonstrates the content of di-, tri-, tetra-, hexa-, heptachlorobiphenyls composing the УPCB in the bottom sediments of

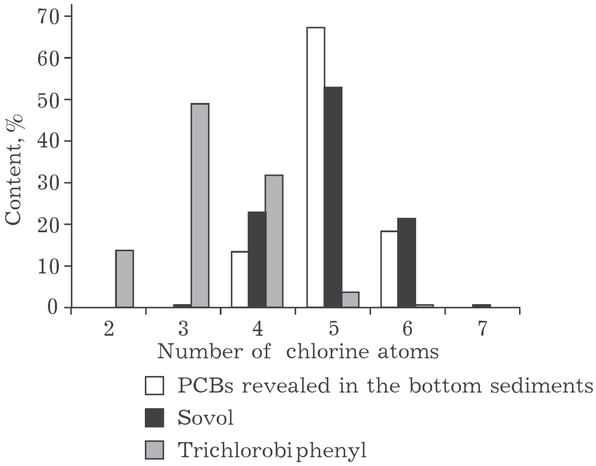

Fig. 3. Content of compounds with different numbers of chlorine atoms among the PCBs revealed in the bottom sediments of the Gusinoye Lake as well as the content of those in Sovol and Trichlorobiphenyl technical products.

the Gusinoye Lake as well as the content of technical products Sovol and TCD. A comparative consideration of the set of compounds with different number of chlorine atoms demonstrated that among the compounds found in the bottom sediments of the Gusinoye Lake, there prevail highly chlorinated PCBs such as tetra-, penta- and hexachlorobiphenyls, whose content is to a greater extent corresponding to a technical product Sovol rather than to a technical product TCD (mostly presented by di-, tri- and tetrachlorinated PCBs) [24]. Such a ratio between polychlorinated biphenyls was found earlier in the bottom sediments of the southern part of the Lake Baikal [18].

Thus, the PCBs detected in the bottom sediments of the Gusinoye Lake are of local origin, since the set of the compounds exhibit the PCB with a great number of chlorine atoms, whereas the global transfer is characterized by much more volatile PCBs, with a small number of chlorine atoms [25]. A proposed source of PCBs in the ecosystem of the Gusinoye Lake could be caused by a widespread use of electrical equipment containing PCBs (the Gusinoozersk power plant, boilers, and electricity supply networks).

For the bottom sediments of the Gusinoye Lake, the following content OCPs and PCBs has been revealed (%): ΣPCB 50.9, ΣHCCH 16.4, HCB 21.9, ΣDDT 11.5.

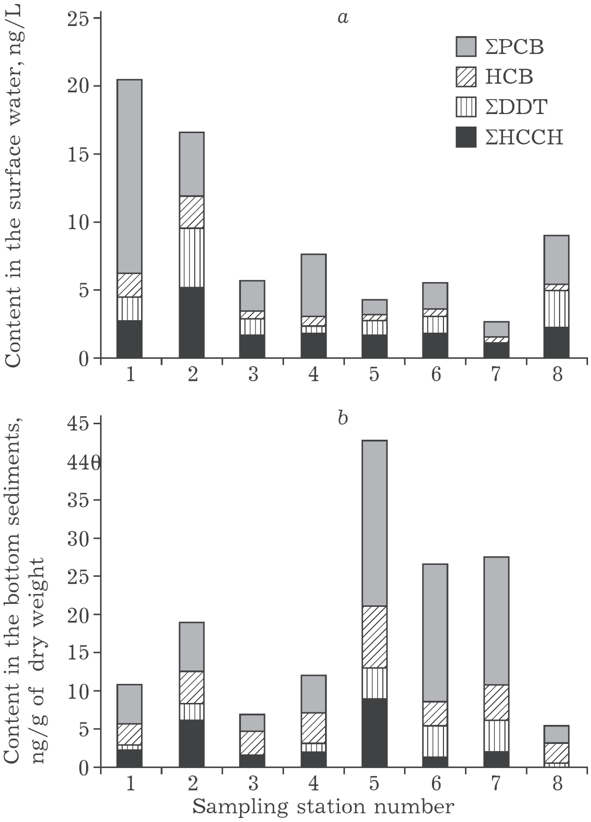

Fig. 4. Content of OCPs and PCBs in the surface water (*a*) and in the bottom sediments (*b*) of the Gusinoye Lake.

For the surface water, the mentioned values are as it follows (%): ΣPCB 46.9, ΣHCCH 24.9, ΣDDT 18.2, HCB 10.1.

Data concerning the total concentration of OCPs and PCBs in the bottom sediments and surface water of the Gusinoye Lake are demonstrated in Fig. 4.

CONCLUSION

Resulting from the studies concerning the contamination of the Gusinoye Lake with OCPs and PCBs, the following conclusions were drawn.

1. The contamination of bottom sediments and surface water of the Gusinoye Lake by compounds DDT and HCB can be described to be a relatively weak one, being comparable with the contamination of the Arctic areas; whereas the contamination HCCH by for the surface water exceeds that inherent in the Arctic areas.

2. The content of DDT, HCCH, HCB and PCB in the surface water of the Gusinoye Lake does not exceed the MPC level acceptable for water reservoirs for drinking and community use. The average concentrations of OCPs and PCBs do not exceed the MPC level acceptable for water reservoirs having fishery significance.

3. It has been found that the total concentration of the compounds of DDT group in the surface water of the Gusinoye Lake is not higher than that for the surface waters inherent in the background territories of Russia.

4. The ratio values between the concentrations such as DDD/DDE and (DDE + DDD)/ΣDDT allow one to evaluate the nature of degradation and the time of DDT entering the ecosystem of the Gusinoye Lake. It has been found that DDT was mostly transformed into the metabolites such as DDD and DDE.

5. The contamination of bottom sediments and surface water by PCB compounds exceeds the PCB contamination of the Arctic areas.

6. It has been found that the content of tetra-, penta- and hexachlorobiphenyls in the samples of bottom sediments is almost corresponding to a technical product Sovol.

7. It has been found that supplying the OCPs and PCBs to the ecosystem of the Gusinoye Lake is mainly caused by local anthropogenic sources.

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