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Effects of Polymetallic Ore Occurrences on Mercury Accumulation by Aquatic Biota in River Ecosystems

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Abstract—The study represents the data on metal content in the body of stoneflies *Perla pallida* Guerin-Menville, 1838 (Plecoptera, Perlidae) and in muscles of the fish family Cyprinidae from the small tributaries of the Belaya River (Republic of Adygea, Northwest Caucasus) with ore mineralization of various metals including mercury. It was found that mercury (Hg) concentrations in stoneflies are corresponds to from the water basins without local Hg sources. Mercury content in stoneflies depends on season and is more intensive in early ontogenesis in winter than in the warm season. Mercury concentration in 0.01–0.72 mg/kg dry weight can cause the pathomorphological changes in the structure of organs and decrease the adaptive potential in competitive population of stoneflies, in general. The Hg concentration reached 0.09–0.69 mg/kg in the studied fish species (bleak, gudgeons, and barbels) and was similar to concentrations in stonefly larvae. This can be related with low size-weight parameters of fishes in samples as wells as with similar feeding patterns of hydrobionts.

Keywords: mercury, aquatic biota, river ecosystems, ore occurrences in catchment areas

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INTRODUCTION

The global emission of Hg from natural and anthropogenic sources into the atmosphere, followed by the migration and deposition to the surface of aquatic and terrestrial ecosystems, is still the main reason for the increase in its content and negative impact on biota (Kolka et al., 2011; Eagles-Smith et al., 2016). Various compounds of this metal, including the most toxic and bioavailable methylated mercury, form strong chemical bonds with compounds that make up the tissues of living organisms and accumulate in concentrations hazardous to human health. It has been experimentally revealed that, in vertebrates, high levels of Hg in tissues lead to chromosomal aberrations and blood pathologies and negatively affect the nervous system and embryonic development of the fetus (Topashka-Ancheva et al., 2003; Scheuhammer, 2007; Tavshunsky et al., 2017). In invertebrates, this relates to deformations of vital structures, the deceleration of larval growth, and processes of metamorphosis and regeneration of damaged organs (Medvedev and Komov, 2005; Gremyachikh et al., 2006; Jensen et al., 2007).

Studies of the dependence of the accumulation of Hg by living organisms on the parameters of the environment have been and are still carried out mainly in

aquatic and near-aquatic ecosystems, where the activity of bacterial methylation is high, contributing to the inclusion of mercury in food webs (Gilmour et al., 2013; Tavshunsky et al., 2017). Elevated concentrations of Hg in the muscles of fish from rivers and lakes in Russia were recorded both when metal was originated from industrial wastewater (Leonova et al., 2006) and in the absence of local sources in the catchment area, for example, in acidic (water pH < 5.0) lakes in the northwest of Russia (Haines et al., 1992, 1995; Stepanova and Komov, 1997). The most significant concentrations of Hg in the lithosphere are found the regions of its deposits and ore occurrences, although they contain only 0.02% of all mercury (Saukov, 1975). The bulk of Hg is in the form of compounds in mercury minerals and as isomorphic or mechanical impurities in other minerals and often accompanies metals of the platinum group. Therefore, in the presence of polymetallic and especially mercury ores in the catchment area, the probability of Hg in the trophic webs of aquatic ecosystems increases.

In the foothill regions of the North Caucasus, in the Belaya River basin, several ore fields with isolated points of ore occurrences of tungsten, polymetallic ores, gold ore and placer gold, mercury, and manganese have been identified (Volkodav, 2012). However,

Fig. 1. Locations of sampling stations in the tributaries to the Belaya River (numbering from the river head to mouth): (1) Lipovaya River; (2) Khamyshinka River; (3) Bzykha River; (4) Syuk River; (5) Belaya River, near the settlement of Nikel'; (6) Dakh River, downstream of a bridge; (7) Mishoko River; (8) Maykopskaya River; (9) Shuntuk River; and (10) Kurdzhips River, near the settlement of Krasnyi Most.

studies on the content of mercury in aquatic ecosystems of this region have never been carried out.

The goal of the present paper is to investigate the effect of polymetallic ore outcrops found in the Belaya River catchment area on Hg accumulation in representatives of amphibiotic invertebrates (Plecoptera larvae) and fish.

MATERIALS AND METHODS

The Belaya River is the second longest and the most powerful in terms of the discharge volume of the left-bank tributary of the Kuban River, located on the

border of the western and northwestern parts of the Greater Caucasus in the foothill region of the Republic of Adygea. It originates on the slopes of the Fisht-Oshten mountain range and flows into the Krasnodar Reservoir. The river is 277 km in length and the catchment area is 5990 km² (Mel'nikova and Komlev, 2003).

The river basin is a system within which natural, inhabited territories, and agricultural lands interact. The most important components of the natural landscape complexes of the Belaya River basin are small streams which were studied at ten sampling stations (Fig. 1, Table 1).

In the Belaya River basin, the occurrence of ore mineralization of various metals, including Hg, is concentrated. The Khamyshinskoye (Shakhanskoye) mineralization field occupies most of the interfluvium of the Bzykha and Khamyshinka rivers and the left tributaries of the Belaya River (sampling stations 1–3) (Volkodav, 2012). Of the five isolated ore occurrences, the highest Hg content was noted in the Shahanskoye (0.012%–0.2%). Here, in the transverse vertical cracks, in the veins of other minerals, cinnabar is found in the form of inclusions and deposits, which is often accompanied by native mercury. The Hg content in other ore occurrences usually does not exceed hundredths of a percent.

The Belorechenskoye ore field is located in the Syuk River valley (sampling station 4), which flows into the Belaya River (sampling station 5) downstream the settlement of Nikel'. The leading ore-forming minerals of the Belorechenskoye barite ore field are barite, quartz, calcite, and ankerite, while cinnabar is a rare mineral. Mercury mineralization is also con-

Table 1. Hydrographic characteristics of small rivers inflowing the Belaya River

River	Sampling station	River channel length, km	Height above sea level (head/mouth)
Lipovaya	1	8	1340/560
Khamyshinka	2	7	1330/550
Bzykha	3	11	1710/530
Syuk	4	14	990/470
Belaya	5	273	2300/30
Dakh	6	27	—
Mishoko	7	9.4	890/410
Maykopskaya	8	9.5	640/280
Shuntuk	9	12	—/276
Kurdzhips	10	100	2300/224

A dash indicates no data.

tained in the Triassic strata of the Sakhray River area, a tributary of the Dakh River (sampling station 6) (Pekov et al., 2010). The rest of the stations (7–10), located downstream of the Belaya River, do not have manifestations of ore mineralization of metals within their catchment basins.

The studied rivers are typical small watercourses with rocky bottoms. In the bottom invertebrate communities of the Belaya River and its main tributaries, 238 species of invertebrates have been identified, 87.0% of which are insects. They are dominated by Caucasian

littoreophilous representatives of zoobenthos: on biotopes with high water-current velocity, *Epeorus*–*Simuliidae*–*Baetis*–*Blephariceridae*; moderate velocity, *Ecdyonurus*–*Baetis*–*Rhyacophila*–*Hydropsich*–*Perla*–*Gammarus*; and low velocity, *Gammarus*–*Limephilidae* (Shapovalov, 2020).

The materials included 326 stonefly larvae *Perla pallida* Guérin-Meneville, 1838 (Plecoptera, Perlidae), an amphibiotic insect and aquatic organism at the larval stage. In regards to fish, 35 Kuban spirin *Alburnoides kubanicus* Bănărescu, 1964; 29 minnow *Phoxinus* sp.; 9 Caspian gudgeon *Gobio* cf. *holurus* Fowler, 1976; and 3 Kuban barbel *Barbus kubanicus* Berg, 1912. Stonefly larvae were sampled in 2012 and 2013 and fish in 2014 and 2015.

The larvae of stoneflies are widespread in all biotopes of the Belaya River and its tributaries at height of no more than 200 m above sea level. They dwell in cracks on the underside of stones (avoiding a very fast current) and under large stones, where the layer of loose pebbles lies. Large species of fam. Perlidae have a long period of development in a watercourse (up to 3 years) and an active lifestyle. Stoneflies, like the larvae of other insects, may consume detritus of various origins, mycoflora, unicellular and filamentous algae, tissues of macrophytes, and mosses, as well as all kinds of invertebrates (Monakov, 2003). Representatives of carnivorous stoneflies species, which include *Perla pallida*, switch to animal food as they grow, feeding primarily on dipteran larvae, whose share of the total number of food components is 68.6–74.3%, and then on larger organisms: caddisflies larvae and mayflies (Shapovalov, 2020). The feeding spectra of stonefly larvae may expand as they grow and develop. Differences in the composition of food are most often determined by the composition of the population of the watercourse in a particular period.

The fish sampled in the tributaries of the Belaya River are rheophylic cyprinids, whose diet includes insect larvae to varying degrees (Troitskii and Tsunikova., 1988; *Atlas* ..., 2003).

The body length and weight of the sampled animals were measured prior to the processing of samples with further drying at 36°C (whole insects were used; in fish, a fragment of dorsal skeletal muscles was dissected). The mercury content was determined in two

or three replicates by the atomic absorption method on a RA-915+ mercury analyzer with a PIRO (Lumex) attachment without preliminary sample preparation. The accuracy of analytical measurement methods was controlled using DORM-2 and DOLM-2 certified biological material (Institute of Environmental Chemistry, Ottawa, Canada).

Data on Hg concentrations are presented as mean values and their errors ($x \pm mx$) with min–max. Since the distribution of the data differed from the normal (Shapiro–Wilk test), the nonparametric Spearman test was used to identify correlations between the studied indicators, and the Kruskal–Wallis median was used to assess the significance of differences between the samples (the differences are significant at $p < 0.05$) (Sokal and Rohlf, 1995).

RESULTS

The body length and weight of stonefly larvae varied widely: length 5.0–32.0 mm and weight 7.0–455.0 mg. The minimal and maximal concentrations of Hg in the body of insects reached 0.01 and 0.72, respectively; on average, over watercourses, it was 0.06–0.38 mg Hg/kg dry weight (Table 2). The metal content in animals sampled the closest to the source and located in the zone of ore occurrences at sampling station 1 was statistically significantly higher (Fig. 2). The accumulation of Hg by stoneflies at sampling station 2 exceeded that at sampling stations 3–5, 7, and 10; at sampling station 6, it exceeded that at sampling stations 4 and 5. For samples from sampling stations 1, 2, 4, 6, and 10, a significant negative relationship between Hg concentrations in larvae and their body weight was noted: $r_s = (-0.56) - (-0.78)$ at $p < 0.05$; for samples from sampling stations 2, 4, 6, and 10, it was also with body length, $r_s = (-0.55) - (-0.72)$ at $p < 0.05$.

The entire group sample was divided into two subgroups: the first included animals from watercourses located in areas of ore occurrence (sampling stations 1–6); the second, from watercourses without ore occurrence in the catchment (sampling stations 7–10; at station 9, only fish). The metal concentrations in stoneflies from the first group were statistically significantly higher (Kruskal–Wallis test, $H = 21.6$, $p < 0.000$) than from the second: 0.15 ± 0.01 and 0.07 ± 0.004 mg/kg dry weight, respectively. The body length and body weight of the larvae of the first group were significantly less than those of the second group: 17.5 ± 0.3 mm and 19.1 ± 0.6 mm ($H = 5.9$, $p < 0.02$) and 125.3 ± 6.0 mg and 168.0 ± 13.2 mg ($H = 13.9$, $p < 0.002$). With an increase in body length (L), they less intensively gained body weight (W): in the larvae of the first group, $W = -158.56 + 16.18L$ and, in the second, $W = -228.52 + 20.75L$.

In stoneflies from the first group, the same statistically significant negative relationship between the

Table 2. Content of mercury in stoneflies larvae from the tributaries to the Belaya River

Number of sampling station	Data	<i>n</i>	Body length, mm	Body weight, mg	Hg, mg/kg dry weight
1	April, 2012	25	$\frac{15.9 \pm 0.9}{9.0-27.5}$	$\frac{96.7 \pm 16.3}{16.0-325.5}$	$\frac{0.38 \pm 0.03}{0.17-0.72}$
	May 4, 2013	20	$\frac{19.4 \pm 1.4}{10.0-29.8}$	$\frac{166.7 \pm 28.3}{19.0-403.0}$	$\frac{0.24 \pm 0.02}{0.14-0.42}$
	June 19, 2013	19	$\frac{13.5 \pm 1.5}{7.8-31.2}$	$\frac{84.2 \pm 28.2}{15.0-455.0}$	$\frac{0.26 \pm 0.02}{0.12-0.38}$
	Mean				
Total for station 1	2012–2013	64	$\frac{16.3 \pm 0.8}{7.8-31.2}$	$\frac{114.9 \pm 14.2}{15.0-455.0}$	$\frac{0.31 \pm 0.02}{0.12-0.72}$
2	May 4, 2013	43	$\frac{21.5 \pm 0.6}{15.0-32.0}$	$\frac{185.2 \pm 14.0}{69.0-450.0}$	$\frac{0.11 \pm 0.02}{0.05-0.55}$
	June 19, 2013	28	$\frac{18.3 \pm 1.0}{10.8-29.0}$	$\frac{136.1 \pm 19.7}{26.0-396.0}$	$\frac{0.15 \pm 0.01}{0.07-0.22}$
	Mean				
	2013	71	$\frac{20.3 \pm 0.6}{10.8-32.0}$	$\frac{165.8 \pm 11.8}{26.0-450.0}$	$\frac{0.13 \pm 0.01}{0.05-0.55}$
3	May 4, 2013	11	$\frac{23.1 \pm 1.0}{18.0-28.6}$	$\frac{192.4 \pm 21.9}{97.0-314.0}$	$\frac{0.06 \pm 0.006}{0.03-0.11}$
	June 19, 2013	39	$\frac{18.2 \pm 0.9}{8.0-28.1}$	$\frac{139.5 \pm 16.6}{17.0-376.0}$	$\frac{0.06 \pm 0.003}{0.03-0.11}$
	October 2012	33	$\frac{12.1 \pm 0.5}{5.0-22.5}$	$\frac{54.5 \pm 6.3}{7.0-212.0}$	$\frac{0.07 \pm 0.006}{0.02-0.17}$
	October 2012	32	$\frac{15.7 \pm 0.5}{10.0-21.0}$	$\frac{77.2 \pm 6.1}{19.0-163.0}$	$\frac{0.14 \pm 0.005}{0.08-0.19}$
6	May 4, 2013	20	$\frac{19.5 \pm 1.0}{13.0-28.2}$	$\frac{144.0 \pm 21.2}{42.0-363.0}$	$\frac{0.06 \pm 0.006}{0.03-0.10}$
	Mean				
	2012–2013	52	$\frac{17.2 \pm 0.5}{10.6-28.2}$	$\frac{102.9 \pm 9.9}{19.0-363.0}$	$\frac{0.11 \pm 0.01}{0.03-0.19}$
	Total for station 6				
7	May 2013	15	$\frac{16.5 \pm 1.2}{8.0-24.0}$	$\frac{136.0 \pm 25.5}{18.0-368.0}$	$\frac{0.06 \pm 0.01}{0.01-0.09}$
8	May 18, 2013	26	$\frac{19.9 \pm 0.7}{13.0-30.5}$	$\frac{179.7 \pm 19.6}{39.0-451.0}$	$\frac{0.09 \pm 0.01}{0.02-0.16}$
10	July 3, 2013	15	$\frac{20.3 \pm 1.2}{12.2-28.0}$	$\frac{179.8 \pm 24.8}{52.0-330.0}$	$\frac{0.07 \pm 0.006}{0.04-0.13}$
Total for all stations	Mean				
	2012–2013	326	$\frac{17.8 \pm 0.3}{5.0-32.0}$	$\frac{132.6 \pm 5.5}{7.0-455.0}$	$\frac{0.13 \pm 0.01}{0.01-0.72}$

Here and in Table 3: top is the mean and error of mean; bottom is the min–max, and *n* is the number of analyzed stonefly larvae; numbers of sampling stations are the same as in Fig. 1.

Fig. 2. Mercury content in stonefly larvae at sampling stations located in the ore occurrence zone (sampling stations 1–6) and outside it (sampling stations 7, 8, and 10).

mercury content and length and weight was revealed: $r_s = -0.24$, $p < 0.05$. In stoneflies from the second group, the decrease in metal concentration with the growth of animals is also significant, but less pronounced: with length $r_s = -0.17$, $p < 0.05$, weight $r_s = -0.11$, $p < 0.05$. A seasonal (from spring to autumn) decrease in the average length and weight of stonefly larvae (in both cases, $r_s = -0.38$, $p < 0.05$) was noted at a relatively stable level. accumulation of Hg. In two groups of different sizes (in the first, the length of stoneflies is ≤ 15 mm and, in the second, > 15 mm) sampled in the tributaries of the mineralization zone, spring individuals were significantly larger than summer–autumn ones. In the first group (smaller individuals), the metal content decreased from spring to autumn and, in the second, it did not change significantly (Fig. 4).

The minimal and maximal concentrations of Hg in the muscles of the studied fish species varied within 0.09–0.69 mg/kg dry weight (Table 3, Fig. 5). The metal content in the muscles of the minnow is statistically significantly higher than that of the Kuban spirlin. The accumulation of mercury by minnows at sampling stations 1 and 2 did not differ; in the spirlin at sampling station 9, it was higher than that of fish at sampling stations 2 and 10. The same concerns the fish

body weight. A significant positive relationship between Hg concentrations in muscles and fish body weight was noted for the spirlin: $r_s = 0.53$ at $p < 0.05$. The content of metal in the muscles of the spirlin from the stations located in the ore occurrence zones and outside them did not differ statistically significantly.

Fig. 3. Correlation between body length and weight at sampling stations located in the ore occurrence zone (1) and outside it (2).

Fig. 4. Seasonal changes in Hg content in stonefly larvae ≤ 15 mm in length (a) and > 15 mm (b) sampled in tributaries in the ore occurrence zone.

DISCUSSION

The input of Hg from catchment areas with outcrops of polymetallic ore occurrences into waterbodies and streams is both of a natural and anthropogenic nature. In the first case, this is due to natural processes and, in the second, with the more rapid erosion of mineralization zones as a result of human economic activities for the extraction, preliminary processing of minerals, and storage and disposal of waste dumps at their enrichment (Kocman et al., 2011; Hsu-Kim et al., 2018). According to recent estimates, artisanal and small-scale industrial gold mining may be significant sources of Hg for freshwater ecosystems (880 t/year), both industrial production and wastewater emissions (220 t/year); the mobilization of land resources; and changes in land use, forestry, and frequency and intensity of wildfires (170–300 t/year) (Obrist et al., 2018).

The Belaya River basin in the upstream reaches is located in the zone of gold-bearing, copper and lead–zinc mineralization interspersed with mercury-bearing minerals (in particular, cinnabar and native Hg) (Volkodav, 2012). The development of the Belaya River gold placers began in 1932 and continued until 1941, although prospecting works in 1934 and 1935 did not reveal new placers on the river or industrial mineralization (Volkodav, 2012).

The small valleys of the Belaya River Basin were developed by prospectors at the beginning of the last century and continue to interest local metal miners to

this day. Any economic activity in the catchment of rivers (mining of polymetallic ores, deforestation, and

regulation of watercourses) may lead to an increase in the removal of finely dispersed suspension of mercury mineral cinnabar and the accumulation of mercury in bottom sediments within slowly flowing river sections (Hsu-Kim et al., 2018).

The metal content in the larvae of stoneflies and muscles of the studied fish species from the Belaya River and its tributaries in the zone of ore fields and outside it is comparable to that revealed in larvae of amphibiontic insects and fish from waterbodies and watercourses of various regions lacking local sources of pollution in the catchment area and predominantly prone to atmospheric pollution. Thus, the concentration of total Hg in chironomid larvae from lakes in the Canadian Arctic varies within 0.09–0.50 mg/kg dry weight (Ganter et al., 2010). In larvae of dragonflies from subtropical freshwater Lake Caddo (Texas and Louisiana, United States), it is 0.17–0.31 (Chumchal et al., 2011); in Losha River (Vologda oblast), Savala River (Voronezh oblast), and an artificially dug channel opening in Rybinsk Reservoir and Sunozhka River (Yaroslavl oblast), it is 0.02–0.07, 0.04–0.15, and 0.01–0.38 mg/kg dry weight, respectively (Gremyachikh et al., 2013).

The seasonal decrease in the body length and weight of larvae of *Perla pallida* Guerin-Meneville, 1838 is due to the fact that in the spring samples a sig-

nificant part of the animals is represented by large overwintered specimens (the development cycle in stoneflies from Adygean watercourses is about 3 years). Later samples contain individuals hatched in spring (much less often in summer and autumn) at early stages of development (Cherchesova and Zhiltsova, 2013). Low concentrations of Hg in summer and autumn larvae in the small-sized group are associated with seasonal changes in the composition of food: at a specific time period, animals choose the most numerous and available food items (Monakov, 2003). In winter and early spring, it is mainly detritus and bacteria that have developed in and on it; from late spring to autumn, it is algae and invertebrates. According to de Wit et al. (2012), the consumption with food of the bacteria transforming inorganic forms of mercury into organometallic ones more efficiently than algae explains the increased metal content in aquatic organisms in the winter–spring period. In addition, overwintering larvae do not molt, and each molt during the growing season is accompanied by rapid growth and, accordingly, dilution of the accumulated Hg (Ross et al., 1985). In large larvae of stoneflies, the metal content does not change significantly with the change of seasons due to obligate zoophagy: detritus and algae enter the stomachs of predators from the intestines of eaten prey (Monakov, 2003).

Fish with both low and increased concentrations of metal in muscles are found in the rivers and lakes of nature reserves in European Russia remote from sources of Hg emissions, which is determined by the biogeochemical (ecological) characteristics of the waterbodies and their drainage basins. In perch from the Usman River (the Voronezhsky Nature Reserve), a content of 0.35–1.30 mg/kg dry weight was recorded; in the lakes of the Rdeisky Nature Reserve, 0.20–12.00 (Komov et al., 2009); in Darvinsky Reserve, 0.10–4.95 (Stepanova and Komov, 1997); and in Oksky Reserve, 0.10–2.50 mg/kg dry weight (Gremyachikh et al., 2012).

Unlike stoneflies, the levels of metal in the muscles of the Kuban spirlin from the Belaya River and its tributaries located closer to the river head in the zone of ore fields (sampling station 2) and outside it (sampling stations 7, 8, and 10) are low, but upon approaching the mouth of the river, these values, on the contrary, increase. However, this is determined by the small size of fish caught in the upper reaches: for many species, a correlation was noted between the size and content of mercury in the internal organs and muscle tissue (Komov et al., 2009; Gremyachikh et al., 2012). The metal content in the muscles of predatory fish from the Tapajos River (Brazil), one of the main tributaries of the Amazon, from the mid-1980s, where traditional gold mining takes place, reaches 20 mg/kg dry matter (Nevado et al., 2010). With an increase in the distance from the mining sites downstream of the river, it decreases, as is observed in the Katun and Selenga rivers with numerous mineralization zones in the catch-

Table 3. Content of mercury in the muscle tissues of fish the Belaya River tributaries

Number of sampling station	<i>n</i>	Fish body weight, g	Hg, mg/kg dry weight
Kuban spirlin			
2	6	0.11 ± 0.02 0.06–0.17	0.15 ± 0.005 0.15–0.18
8	7	3.1 ± 0.7 1.6–6.6	0.26 ± 0.03 0.13–0.34
9	6	5.1 ± 1.1 2.9–8.9	0.51 ± 0.07 0.26–0.69
10	16	3.2 ± 0.7 0.8–13.4	0.16 ± 0.01 0.09–0.30
Mean		3.0 ± 0.5 0.06–13.4	0.24 ± 0.03 0.09–0.69
Minnow			
1	14	0.03 ± 0.007 0.02–0.15	0.27 ± 0.02 0.16–0.40
2	15	0.11 ± 0.009 0.07–0.18	0.25 ± 0.01 0.17–0.40
Mean		0.07 ± 0.01 0.02–0.18	0.26 ± 0.01 0.16–0.40
Gudgeon			
9	5	1.6 ± 0.7 0.4–4.2	0.20 ± 0.02 0.12–0.24
Kuban barbel			
9	3	2.0 ± 0.8 0.8–3.6	0.19 ± 0.02 0.15–0.23

ment area (Vasil'ev and Sukhenko, 1993; Komov et al., 2014), in which the concentrations of Hg in fish vary in the range of 0.25–0.55 and 0.24–2.33 mg/kg dry weight, respectively.

In water, Hg is present in small amounts in a mixture of various forms: mainly suspended and colloidal particles and, to a lesser extent, a true solution (Han and Gill, 2005; Hsu-Kim and Sedlak, 2005; Balogh et al., 2008; Schuster et al., 2008; Dittman et al., 2010). In a study conducted on the Idrijetsa River (Slovenia), draining the territory of the former mercury mine, it was found that Hg is in the form of a finely suspended material, including colloids, and mercury is transported downstream during short but extreme hydrometeorological events when the metal is remobilized from the bottom sediments of the river (Kocman et al., 2011). During such events, the concentrations of Hg in water and bottom sediments rapidly decrease and, at the same time, bacterial methylation of the metal occurs, which increases the propor-

Fig. 5. Mercury content in the muscles of fish sampled in Belaya River watercourses.

tion of methylmercury (MeHg) in the total Hg content and, consequently, the risk of accumulation in biota (Ullrich et al., 2007; Kocman et al., 2011; Jackson et al., 2019).

The bioavailability of Hg for biota depends in a complex nonlinear manner on a variety of biological and ecological factors that affect the patterns and pathways of metal migration along food webs (Lavoie et al., 2013; Karimi et al., 2016; Polito et al., 2016; Eagles-Smith et al., 2018; Hsu-Kim et al., 2018). This is why bottom sediments of waterbodies and soils in the catchment areas containing relatively high Hg concentrations with low bioavailability of metal may be sources of its accumulation in aquatic organisms only to low levels. For instance, in the muscles of bream from Lake Serebry located in the immediate vicinity of the Karabash copper-smelting plant in Chelyabinsk oblast, Hg concentrations are several times lower than in the muscles of bream from Lake Seliger in Tver oblast, in the vicinity of which there is practically no industry (Tatsii et al., 2017).

Since the second half of the last century, there has been no active industrial and artisanal mining of gold or polymetallic ores in the Belaya River basin. This is why the input of Hg occurs mainly with atmospheric precipitation and runoff from the river catchment area as a result of the erosion (leaching) of rocks, as well as of mountain, floodplain-meadow, and forest soils (Volkodav, 2012). The specificity of the river hydrological regime (closer to the source, glacial, snow and storm water supply the sections closer to river headwaters; there is a high water-current velocity) does not create favorable conditions for the bacterial methylation of mercury compounds increasing its bioavailability in either the water column or in bottom sediments. The accumulation of Hg by animals depends on many environmental factors: the water pH level, the area of wetlands in the drainage basin, and the concentration of dissolved organic matter (Harding

et al., 2006). However, the main mechanism for an increase in the concentration of metal in each subsequent link is its trophic transport along the food web (Eagles-Smith et al., 2018).

The predatory larvae of stoneflies participate in the transfer of Hg along the food web of aquatic ecosystems as secondary and tertiary consumers, which are at the same trophic level with some fish and at the same time are a food object for omnivorous and predatory fish species. In the bottom fauna of mountain streams, amphibiontic insects are one of the most abundant and widespread groups of invertebrates. Potential food items for predatory stonefly larvae are >40% of the species composition of benthic invertebrates in the studied rivers (Shapovalov, 2011, 2020). By consuming large insect larvae (in a specific case, dragonflies), fish in the watercourse limit the Hg flux into the terrestrial ecosystem, but do not affect the mercury flux passing through the small larvae of chironomids and caddis flies (Tweedy et al., 2013).

The present study revealed that concentrations of Hg in larvae of stoneflies and muscles of fish from the small streams of Adygea are low and comparable to each other: 0.01–0.72 and 0.09–0.69 mg/kg dry weight, respectively, which may be associated with the low size and mass parameters of fish in the samples and, as a result, the similarity of the food spectra of the aquatic organisms under study. As food items, the larvae of stoneflies (and also adults, even taking into account the possible increase in metal concentration as a result of metamorphosis), accumulating Hg within the established values, cannot significantly harm the predatory representatives of the aquatic, near-water, and terrestrial ecosystems in the Belaya River basin.

As was shown on the chironomids from Arctic lakes, during metamorphosis, the concentration of methylated forms of mercury increases two- to three-fold (Chetelat et al., 2008). Insect imagoes emerging from waterbodies contribute to the removal of metal water and its transfer into terrestrial ecosystems (Speir et al., 2014; Chumchal and Drenner, 2015; Williams et al., 2017) and, at the same time, pose a threat to representatives of the arachno- and avifauna, both near-water and those unconnected to aquatic ecosystems (Cristol et al., 2008).

Can Hg at the recorded concentrations be harmful for the species under study and representatives of near-water biota? The few experimental and field studies evaluating the potential toxicity of Hg have been done on a dose–response basis. The concentrations of metal in the medium (water, bottom sediments, and substrate) was revealed to cause lethal (mortality, % for a certain number of hours), sublethal (slowing growth, motor and respiratory activity, metamorphosis, reproduction, and having a teratogenic effect), and an absence of visible effects of the reaction of living organisms. Thus, the exposure of *Chironomus riparius*

Meigen, 1804 larvae to a solution of Hg chloride (1.58 mg/L) resulted in the immobilization of 50% of the individuals after 48 h (Rodrigues et al., 2013); keeping these larvae on bottom sediments with total Hg concentrations of 0.93, 2.42, and 3.84 mg/kg dry weight leads to a decrease in the survival rate of chironomids (88, 80, and 26%, respectively) and a suppression of growth and development rate compared to control (the percentage of individuals hatched from eggs is 94, 74, and 8%, respectively) (Chibunda, 2009).

However, it is preferable to judge the danger of Hg for biota (a set of negative deviations in the development and functioning of biological systems of all levels) by the concentrations of its inorganic and organic forms not in abiotic components of the environment, but in living beings of different levels of the food web, which reflect the features of biogeochemical transformations of metal in specific ecosystem conditions (Jain et al., 2007). In *C. riparius* larvae reared on mercury-containing food or bottom sediments, the accumulation of 0.6–0.8 mg Hg/kg dry weight slows down the metamorphosis, and the proportion of individuals with deformations of the mouth structures of the head capsule increases to 19–20% (Gremyachikh et al., 2006; Tomilina and Grebenyuk, 2019). The concentrations of Hg (0.01–0.72 mg/kg dry weight) revealed in the larvae of stoneflies, like in chironomids belonging to the group of amphibiotic insects, may lead to pathomorphological deviations in the formation of vital structures and, accordingly, to a decrease in the competitiveness of animals for food resource, increasing their availability to predators.

CONCLUSIONS

Mercury concentrations in stoneflies from small tributaries of the Belaya River are comparable to those in amphibiotic insects from waterbodies and streams lacking local sources of Hg in the catchment area and predominantly subject to atmospheric pollution. Higher levels of metal accumulation were found in the larvae of stoneflies from watercourses in the Khamyshinsky field of mineralization (sampling stations 1 and 2), the runoff from which represents an additional, albeit insignificant, source of metal input into the ecosystems of watercourses. The four fish species studied (small specimens predominated in the samples) accumulated Hg in the same concentrations as the stoneflies. The predatory and large larvae of the stoneflies are not so much food items for small fish as their food competitors. In stoneflies accumulating mercury at concentrations of 0.01–0.72 mg/kg dry weight, an increase in the proportion of individuals with pathomorphological deviations in the structure of the oral apparatus is possible, leading to a decrease in the viability of the population and, accordingly, the provision of food for animals of higher trophic levels.

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COMPLIANCE WITH ETHICAL STANDARDS

Conflict of interests. The authors declare that they have no conflicts of interest.

Statement on the welfare of humans or animals. All applicable international, national, and/or institutional guidelines for the care and use of animals were followed.

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