Yury G. Udodenko[1,2], Christopher T. Robinson[3], Javzan Choijil[4], Renchinbud Badrakh[4], Jansagsodnom Munkhbat[4], Elena S. Ivanova[2], Victor T. Komov[1,2]

Mercury levels in sediment, fish and macroinvertebrates of the Boroo River, northern Mongolia, under the legacy of gold mining.

[1]Papanin Institute for Biology of Inland Waters, Russian Academy of Sciences, Borok, Nekouz district, Yaroslavl oblast, Russia 152742

[2]Department of human health and biology, Cherepovets State University, Lunacharskogo, 5, Cherepovets, Russia 162600

[3]Department of Aquatic Ecology, Eawag, 8600 Duebendorf, Switzerland and Institute of Integrative Biology, ETH-Zürich, 8092 Zurich, Switzerland

[4]Institute of Geography-Geoecology, Mongolian Academy of Science, Baruun Selbe 15, Ulaanbaatar, Mongolia 210620

Author for correspondence: Yury G. Udodenko; udu@mail.ru, Tel. +7 960 109 37 89

Declarations

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We did not use special application or codes for this study

Abstract

Gold mining is currently one of the main anthropogenic sources of mercury in the environment. In this study, the total mercury content was measured in bottom sediments, benthic macroinvertebrates (mayfly larvae), and fish (Siberian dace) along the Boroo River in northern Mongolia. There was a gold recovery plant in the middle reaches of the river until the mid-twentieth century; an accident there in the 1950s caused a mercury spill. We found an increased content of mercury in measured ecosystem components near the plant compared to upper reaches of the river. The mercury content in sediments varied from trace amounts in the upper Boroo to 2200 ng/g dry weight (dw) in the vicinity of the plant ruins. The mercury content in mayfly larvae ranged from 50 to 2940 ng/g dw and had spatial pattern as sediments, with highest concentrations near the plant. The mercury content in sediments was lower at the mouth of the Boroo River than near the plant, reflecting the lower boundary of the mercury spill. Maximum values of mercury content in fish muscle were found at the river's mouth, and were several times higher than in other rivers of northern Mongolia. Median mercury content in muscles of dace from the lower Boroo in 2016 has doubled since studies in 2010–2012, which may be the result of current mercury releases from gold mining.

Keywords: bottom sediments, mayflies, Siberian dace, artisanal small gold mines

Introduction

Mercury and its compounds are ubiquitous in the environment and toxic for most biota, including humans (Beckers and Rinklebe 2017; Driscoll et al. 2013). Natural processes (volcanic eruptions, weathering) and anthropogenic activity (urban discharges, agricultural materials, mining, and industrial combustion and discharges) can release mercury into the environment (Pacyna et al. 2010). Mercury occurs in nature in several forms, the most toxic of which is MeHg, which can accumulate in living organisms (Broadley et al. 2019; Riva-Murray et al. 2011).

Mercury is easily extracted from mineral deposits, it has been extremely useful in various industrial processes, most notably to concentrate (amalgamate) gold. Until the beginning of the twentieth century, amalgamation was widely used in gold mining around the world. Currently, the method is mainly used in artisanal small-scale gold mines in developing countries. Indeed, small-scale gold mining now makes the largest contribution to anthropogenic mercury emissions (UN Environment 2019). Such mining accounts for about 38% of all anthropogenic mercury emissions, with the bulk occurring in South America, Africa, South-East and East Asia. During gold mining, mercury enters the aquatic environment in the form of metallic mercury and in the form of amalgam with gold (Hg-gold amalgam), which accumulates in bottom sediments (Balzino et al. 2015; Lacerda and Salomons 1998; Moreno-Brush et al. 2020). Here, Hg is stabilized by mineral particles as they are progressively buried and likely only transported downstream during high flow events. In areas where gold was amalgamated previously or is still being mined, the mercury content in bottom sediments is higher compared to areas with less anthropogenic impacts (Kaus et al. 2017; Martinez et al. 2018; Mason et al. 2019).

Under certain conditions, mercury from the bottom sediments can bioaccumulate in trophic webs. The trophic position of organisms in food web structure is often correlated to Hg concentration and can represent the intensity of Hg bioaccumulation in the environment (Scheulhammer et al. 2015). Levels of mercury in fish muscle tissue are also often used to assess the response of an ecosystem to mercury entering waterbodies (Wiener et al. 2002). Fish living in small rivers (Cyprinidae, Percidae, Umbridae) occupy the top positions in the structure of the local food network and are considered a good indicator of mercury bioaccumulation (Riva-Murray et al. 2011).

Mining became one of the most important sectors of the Mongolian economy in the period 1990–2005 (Bulag 2009). A large number of both abandoned and active gold mines are located in the north of the country within the catchment area of the Kharaa River, which belongs to the Selenga River basin – the largest tributary of the lake Baikal (Karthe et al. 2015). Illegal gold mining is also widespread in the region (Grayson et al. 2004; Murray 2003). In the first half of the twentieth century, a gold recovery plant that used mercury operated in the middle reaches of the Boroo River – small tributary of the Kharaa River (Tumenbayar et al. 2000). The plant stopped operating in 1956 as a result of an explosion that spilled several tons of mercury into the river. The area around the former plant is known as Boroo Mercury placer, and mercury is mined illegally there by panning the river sediments and digging makeshift mines. This mercury is taken to nearby villages where it is used to recover gold from artisanal mines. The Boroo River currently discharges waste water from an active gold mine, which contributes to the ongoing flow of toxic substances, including mercury, into the river (Hofmann et al. 2010; Inam et al. 2011).

Previous studies of the Boroo River ecosystem have found high mercury concentrations in fish tissues near an abandoned plant (Kaus et al. 2017) and in the river bottom sediments and soils (Brumbaugh et al. 2013; Coufalík et al. 2012), with levels several times higher than other rivers in northern Mongolia. However, no survey of mercury levels in biotic and abiotic components of ecosystems in along the Boroo River has been conducted. When mercury has been in a river system for a considerable period, a study limited to the area immediately surrounding the initial site of the contamination may not be sufficient to fully understand the footprint of the contaminant throughout the ecosystem (Jackson et al. 2019). Therefore, the aim of the study was to determine the extent of mercury bioaccumulation in the food web along the entire linear gradient of the Boroo River.

Materials and Methods

Study area

The Boroo River is a left tributary in the middle of the Kharaa River (Hofmann et al. 2010). This is a typical small river with an average depth of 1–1. 5 m and a width of 2-5 m. The river is located 110–150 km northwest of Ulaanbaatar, latitude 48°40–50' N, and flows to the north at longitude 106°10' E (Fig 1.). The strongly continental climate of the study region is characterized by very low temperatures in winter (-40 °C) and high temperatures in summer (+40 °C) (Karthe et al. 2015). Average annual precipitation is 260–350 mm, more than 70% of which falls in the summer (from June to August). The river flows through wide (2–5 km) intermountain valleys used for tillage farming and grazing. A floodplain with oxbow lakes and channels is formed in the middle course and in the estuarine zone. The soil cover of the valley is represented by kastanozems, and in floodplains by fluvisols.

According to the Institute of Geography and Geoecology of the Mongolian Academy of Sciences, the chemical composition of the water of the river belongs to bicarbonate-calcium. The water hardness on the Boroo River was 3.50-4.00. The mineralogical composition of bottom sediments in the riverbed does not differ along the river. The results of the chemical analysis of the water are presented in the Supplementary materials.

Sample selection and preparation

In August 2016, samples of bottom sediments, macroinvertebrates (*Batis,* Ephemeroptera) and dace (*Leuciscus baicalensis*) muscle tissue were taken along a linear gradient of the Boroo River. Sampling occurred at sites with different potential anthropogenic impacts: 1) sites 10-13 km upstream from the plant's ruins, with mercury content presumably affected only by inflow from the surrounding farm fields (S1–S3); 2) sites midstream, near the plant's ruins and in the mercury spill zone after the explosion (S4-S5); 3) sites downstream from the ruins of the gold recovery plant (S6-S7); 4) sites near modern gold mine and in the river-mouth area ca 30–40 km from S1 (S8–S10); and 5) two sites on the Kharaa River for comparison ca. 20 km upstream and 2 km downstream of the confluence with the Boroo river (S11–S12).

The surface layer (0–5 cm) of bottom sediments of all 12 sites was sampled using a grab sampler. Three subsamples were collected at each site, mixed, and placed in pre-washed, with 10% nitric acid, plastic bags. During sampling, the outermost layer of the sediments was discarded to avoid possible contamination from the sampler. Conventional techniques recommend freezing sediment and soil samples as quickly as possible after sampling, and freeze-drying before analysis. This is done in order to avoid the decomposition of MeHg under the influence of bacteria, and the loss of gas mercury at high concentrations in the environment (Kodamatani et al. 2017; Reis et al. 2015). Since the objectives of this study were not to determine the content of MeHg in bottom sediments, and in the water environment where the sediments are located, gaseous mercury is present in trace amounts (Kaus et al. 2017) and thus drying samples in air is acceptable. The possible losses of mercury during such drying from the activity of microorganisms are 10% and are within the permissible error of the method used to determine the mercury content (Hojdová et al. 2015). Further, this method of sample preparation has been used in a number of studies, which allows us to compare the results of this study with previously published data (Bolaños-Álvarez et al. 2016; Dauvalter and Kashulin 2018; Heaven 2000; Kot et al. 2010; Odumo et al. 2014; Rimondi et al. 2019; Zhang et al. 2018). Considering this, samples were immediately air-dried in the field, ground in a mortar, sieved through a 2-mm sieve, and packed in bags for transport to the laboratory for analysis.

Mayfly larvae were collected at 7 sites from different parts of the river by kick sampling (250-μm mesh net, 5-minute collection time). Mayflies were chosen because they comprise up to 45-50% of the diet of dace in northern Mongolia (Chandra et al. 2005). Macroinvertebrate samples were divided into 2 parts. One part was fixed with ethanol and taken to the laboratory for enumeration, and a second part, used for mercury analysis, was washed with DI water and air dried in in the field.

Dace (*Leuciscus baicalensis*), a common fish in the river, was collected from each site (7–21 fish per site). *L. baicalensis* is a small-bodied cyprinid that consumes periphyton, benthic invertebrates and terrestrial insects (Chandra et al. 2005). Fish were caught with gill nets and fishing rods at all sites except upstream of the Boroo River (S1) where the river is very shallow. Each fish was weighed and its length measured (total length). A sample of muscle tissue weighing about 5 g without skin and bones was taken from the dorsal side above the lateral line. Fresh tissue samples could not be frozen in the field, so the samples were immediately air dried within 48 hours in a field before being transported. Previous surveys showed that replacing freezing and subsequent lyophilization of samples of different fish tissues with drying at relatively low temperatures (50 °C) does not distort analytical results and nor change the THg content in samples (Schmidt et al. 2013). A similar method of preparing fish muscle tissue samples has been used in other studies (Mason et al. 2019; Panichev and Panicheva 2015). A total of 155 samples of dace muscle tissue were collected from 11 sites.

Analysis

The pH was determined in-situ for sediment using a glass electrode pH meter. The amount of organic matter in bottom sediments was estimated using the standard loss on ignition method (Heiri et al. 2001). Subsamples (2 g) of sediments were combusted at 600 °C in an oven for 6 h and reweighed.

The mercury concentration of each sample (bottom sediments, mayflies, fish muscle) was determined by pyrolysis using direct thermal decomposition atomic absorption spectrometry c (RA-915, Lumex, Russia) using a pyrolytic attachment PYRO. This method of analysis does not require additional chemical preparation of samples. Replicated samples (n =3) of bottom sediments and muscles of dace were analyzed from each site. Because mayfly larvae have a small mass, 2–5 combined samples (each comprising several individuals) was analyzed for each site. The mercury content in all samples is given for dry weight (dw). To compare the obtained concentrations of mercury in the muscles of dace with those studies where the mercury content is expressed as wet weight (ww), we converted from wet weight to dry weight. Assuming 80% moisture in muscle samples, conversion to dry weight can be estimated as ([dw] = 5[ww]) (Campbell et al. 2008). Before analysis, the instrument was calibrated using a liquid calibration solution GSO (Russia). Quality control was assessed by analyzing international certified standards and DORM-2 for the analysis of muscle tissue of fish and mayfly larvae, and MESS-3 for the analysis of bottom sediments. Total mercury concentration measured for DORM-2 was 4.54 ± 0.05 mkg/g dry weight (dw) (certified value 4.64 ± 0.26 mkg/g dw), and for MESS-3, 0.095±0.08 mkg/g dw (certified value 0.091 ± 0.09 mkg/g dw). Quality control was every 10 samples.

Data analysis

The level of anthropogenic impact on the mercury content in bottom sediments was estimated using the geoaccumulation index (Igeo) of Forstner et al. (1990):

$$Igeo = \log_2(C_{Hg} / 1.5 \cdot B_{Hg})$$

where C_{Hg} is the mercury concentration measured in the sediment, B_{Hg} the background concentration in the area, and 1.5 a correction factor for lithogenic effects. Due to the lack of data on the background content of mercury in the ores of Mongolia, concentrations for quaternary rocks of Transbaikal region were taken as background concentrations for this research (18 ng/g) (Ivanov and Kashin 2010). Transbaikal region is the territory of Russia adjacent to Mongolia, which has a landscape that is homogeneous with the territory of northern Mongolia. Therefore, it is quite acceptable to use background mercury values for Transbaikalia in this study. The following classification is given for the Igeo index: <0 no anthropogenic impact; 0–1 weak anthropogenic impact; 1–2 and 2–3 moderate to strong impact; 3–4 strong impact; 4–5 strong to very strong impact, and >5 very strong impact.

The level of mercury accumulation in mayfly larvae was estimated using the Biota Sediment Accumulation Factor (BSAF) (Gobas and Morrison 2000):

$$BSAF = C_O/C_S;$$

where, C_O is the mercury concentration in the organism and C_S is the mercury concentration of sediment. A BSAF >1 indicates the bioaccumulation potential of mercury in relation to sediment mercury levels.

Since there were only 2 different trophic levels of the Boroo River trophic web (mayflies larvae and dace) evaluated, the intensity of mercury accumulation in the trophic network was estimated using a simplified biomagnification coefficient. (Mackay and Fraser 2000):

$$BMF = C_B/C_A,$$

where, Hg concentration in the organism C_B (in this study, dace) to Hg concentration in the organisms diet C_A (in this study, mayfly larvae).

The Shapiro–Wilk test was used to determine normality. Since the mercury content in all samples was not normally distributed, the Kruskal-Wallis H-test was used to compare mercury concentrations among the different sites. To determine the dependence of the mercury content in sediments from amount of organic matter and pH, and the mercury content in fish muscles on the length of the fish, the Spearman rank correlation coefficient was used. Simple linear regressions between log10-transformed mean Hg concentrations in mayflies and fish muscles, BMF and log10-transformed Hg concentration in sediments were run to evaluate the potential for Hg accumulation through the food web. A p-value <0.05 was considered significant in all statistical tests.

Results

The organic matter content in bottom sediments of the Boroo and Kharaa Rivers ranged from 3.5 to 13.5% (Table 1). The sediments were neutral to slightly alkaline. The total mercury content in sediments of the Boroo River varied widely (Table 1). The minimum mercury content was observed in the upper reaches of the river (5–10 ng/g dw). Downstream, the mercury content in sediments increased and reached a maximum 2–6 km below the ruins of gold recovery plant (1697–2230 ng/g dw). In sediments of the Boroo River's mouth, the concentration of mercury in sediments decreased to 103±8 ng/g dw. At sites of the Kharaa River above and below the mouth of the Boroo River, the mercury content in the sediment was 10±8 and 31±21 ng/g dw, respectively. There are no significant correlations between the properties of bottom sediments (organic matter content, pH) and the content of total mercury.

The geoaccumulation index (Igeo) for Boroo River sediments varied from -2.4 to 6.4, and a different degree of anthropogenic influence on the mercury content was evident in different parts of the river (Fig. 2A). No mercury pollution was observed in the upper reaches of the Boroo River and in the Kharaa River above the confluence of the Boroo River. The strongest anthropogenic impact was found at sites near the plant's ruins. At sites directly adjacent to the plant, the anthropogenic impact was moderate.

The most abundant genus of mayfly was *Baetis*, making up 92–100% of the total number of mayflies at all sites along the Boroo River. Along with chironomid larvae, they were the dominant macroinvertebrate in the river. Their abundance at different stations varied between 35–1900 specimens/per sample (Table 1). The presence of *Baetis* also was recorded near plant ruins and at the river's mouth. There was no significant correlation between the abundance of mayflies, the content of mercury and organic matter in sediments ($p>0.05$). The minimum mercury concentrations in *Baetis* were found at the upper Boroo sites and Kharaa Rivers sites (50±10 and 130±20 mkg/g dw, respectively), while the maximum mercury values were at sites downstream from the plant's ruins and at the Boroo River mouth (2940±420 and 2360±130 mkg/g dw, respectively). The highest BASF values were observed at sites near of the explosion area and in the estuarine area of the river (S4, S10) (Fig. 3). The BASF was 0.9-1.3 at sites of the river with the greatest anthropogenic impact on the mercury content in bottom sediments.

There were no significant differences in weight and length between dace at different sites in the Boroo River and Kharaa River. The mercury content in the muscles of the dace in the Boroo River ranged from 450 to 15,300 ng/g. The spatial distribution of mercury in dace was heterogeneous and concentration levels can be divided into 4 groups: 1) upper reaches of the river (S2) – average mercury content = 810±350 ng/g; 2) sites upstream and downstream of the explosion area and the plant's ruins (S3–S7) – average mercury content = 3140±1250 – 4770±1810 ng/g; 3) sites 10 km below the plant ruins and to the mouth of the river (S8–S10) – average mercury content = 8040±2870 – 9600±2670 ng/g; and 4) sites on the Kharaa River (S11–S12) – average mercury content = 880±390 – 1390±990 ng/g. Differences in mercury concentrations among groups were statistically significant (Kruscal–Wallis test: $H(2; 155) = 10.6$; $p<0.001$), with the exception of the upper reaches of the Boroo River and Kharaa River (Fig. 2B). No correlation was found between the mercury content in fish muscle tissue and the length or weight of fish. The BMF values along the river varied from 1.2 to 16.2 (Fig. 3). Minimum values were observed at sites with strong anthropogenic impact on the mercury content in sediments, and maximum values were observed in the upper reaches of the river and the Kharaa River. There were no significant correlations between BMF and fish length.

Regression models showed significant positive relationships between log10-mercury content in sediments, mayfly larvae, and fish muscles (Fig. 4). The strongest relationship was found between the log10-mercury content in mayfly larvae and fish muscles (slope = 0.45; R^2-adjuted = 0.74; p = 0.008). A weak negative relationship of BMF on the Log10-content of mercury in bottom sediments was found (slope = - 4.47; R^2-adjuted = 0.54; p = 0.037).

Discussion

The mercury content in the bottom sediments and biota in the Boroo River increased from minimum values in the upper to middle part of the river to maximum values below the ruins of the gold processing plant, and then to low values again in the Kharaa River above and below the confluence with the Boroo.

River sediments in the upper Boroo and Kharaa are mercury-depleted relative to the upper continental crust; average content in the crust is 70 ng/g (Kabata-Pendias 2011). The concentration of mercury in sediments can be considered natural, since they are comparable to the background values for the Transbaikal region (Ivanov and Kashin 2010). In contrast, sediments in central part of the river are considerably enriched in mercury (15–30 times). According to the Igeo index, the increased mercury content in bottom sediments is due to anthropogenic impacts. Further, it should be noted that the mercury content in the bottom sediments of rivers with large metallurgical and chemical industries located on their banks can significantly exceed those in the mercury spill zone on the Boroo River. For instance, the sediment mercury content was 105,000–147,000 ng/g in the Nura River (Central Kazakhstan), and 1 million ng/g in the Idria River (Slovenia) (Gosar et al. 1997; Ullrich et al. 2007). The most likely source of mercury in the Boroo River ecosystem is gold mining in the past and present, as the river is remote from industrial areas of

Mongolia. Apparently, the mercury spill still affects the mercury concentration in sediments, since the spatial distribution of Hg in sediments coincides with the selected boundaries of the spill spread along the river valley (Tumenbayar et al. 2000). The earlier elevated mercury concentrations found in soils and tailings near the plant ruins were 5-15 times higher (Coufalík et al. 2012), thus this study confirms that sediments of the middle Boroo River represent a mercury ‘hotspot’ in the Kharaa River basin.

Studies in other countries show that the mercury content in river bottom sediments in areas of modern gold mining is generally higher than local background concentrations, and in some areas the excess can be tens of thousands of times background levels (Table 2). On rivers with intensive gold mining, the mercury content of bottom sediments in the vicinity of mines can reach values exceeding the maximum concentrations in Boroo by 100-150 times. Therefore, the current levels of mercury are mainly related to its intake in the past due to gold mining and the explosion. This assumption is supported by the fact that in other rivers of the Selenga River basin, where gold is currently mined, mercury in the sediments is represented in trace amounts (Orkhon, Tuul), (Brumbaugh et al. 2013). However, the use of mercury on the Boroo River by the local population when searching for gold in tailing dumps cannot be completely excluded.

Typically, at locations non-impacted by local sources of Hg contamination, sedimentary Hg correlates with the organic content (Gao et al. 2016; Ma et al. 2020; Mendes et al. 2016; Zhang et al., 2018). While there were no significant correlations with organic matter when sediments were heavily contaminated with mercury (Cukrov et al. 2020; Mason et al. 2019; Vöröš et al. 2018), the absence of correlations in the case of the Boroo River is due to the fact that there is more mercury in bottom sediments than can be retained by the amount of organic matter.

The spatial distribution of mercury in mayfly larvae and dace along the river length differed from the pattern in bottom sediments. The mercury content in the larvae of mayflies and invertebrates of other taxonomic groups living in rivers and reservoirs in Mongolia has not been previously studied, so direct regional comparison is not possible. High concentrations of mercury in mayflies from the Boroo River at stations in the vicinity of gold recovery plant are due to the increased mercury content in the river's sediments. It should be assumed that the mercury content of mayfly larvae is significantly lower in other rivers of northern Mongolia, and comparable to the mercury content in *Baetis* from the upper Boroo and Kharaa Rrivers.

Comparison of the mercury content in mayflies in other rivers of the world shows that the mercury content in mayflies of the same genus ranges widely. For example, the high mercury concentration of *Baetis* larvae in the upper reaches of the river El Harrach in Algeria (median: 900 ng/g dw, maximum: 4100 ng/g dw) (Bouchelouche and Arab 2020) was higher than that of *Baetis* larvae from the upper Boroo and. Kharaa Rivers, but lower than that at the mouth of the Boroo River. The mercury content of mayflies from Lake Sunapee (New Hampshire, USA) was also comparable to mayflies from the upper Boroo River (Broadley et al. 2019). In contrast, the mercury content of *Baetis* living in the Rybinsk Reservoir (the center of the European part of Russia) was 10 times lower than that of mayflies from the upper reaches of the Boroo Rriver (Komov et al. 2017). The reason for such differences can be both the different content of mercury in the bottom sediments and its different bioavailability.

At all sites, the mercury content in the dace muscles was higher than the mercury content in the bottom sediments and mayfly larvae. The levels of mercury in dace from the upper Boroo River and Kharaa River are comparable to dace from other rivers of the Selenga basin, so they can be considered natural (Kaus et al. 2017; Komov et al. 2014). Similarly, only these sites had mercury levels below the US EPA recommended levels (460 ng/g ww = 2300 ng dw). For the most part of Boroo, the mercury content in fish is higher than recommended values and regular consumption of fish is not encouraged.

While along most of the length of the Boroo River, the mercury content in dace was significantly high. The lack of significant differences between S3 and S7 suggests that the dace migrate along the river. The high biomagnification factor at S3 and S4 and the uniformity of mercury content in the muscles of the dace and in the sediments, compared with the other sites, confirm the assumption that the fish come from areas located downstream. There are practically no studies on the range of fish movement in small rivers. Earlier it was shown that the dace are able to migrate a distance of 400–600 m in a month (Clough and Beaumont 1998). In the case of a short frost-free period in Mongolia, this distance can be 1.5–2.5 km per season. Further, conditions of the Boroo River, such as its small size, it is possible to expect long distance migration, which is confirmed by the nature of the distribution of mercury in fish compared to precipitation. Studies on rivers in southern regions of the European part of Russia have shown that the fish, in 1-2 months, can move distances of up to 160 km both upstream and downstream from the tagging site (Fedorov at al. 1965). It is possible that in streams with a limited food resource, for example, the Boroo, migration opportunities of fish are greater and are limited exclusively by the length of the river sections free of barriers. The typical mercury content at site S2 in comparison with the downstream sites and among other Mongolian rivers is probably due to the presence of natural barriers on the river (rapids or shoals) that restrict the migration of fish, since there are no hydraulic structures between the sites. At the same time, there are no dace at site S1 because of the dam located in below the site. The increase in the concentration of mercury in fish muscles at stations S8–S10 compared to the upper sites may be caused by the presence of an unidentified mercury source. This assumption is supported by the fact that there is a modern mine nearby, so there is a certain probability that the local population still uses mercury to extract gold from tailing dumps and mercury enters the river leading to an increase in the mercury content in fish.

Comparison of the results of this study with previous studies on the Boroo River (Komov et al. 2015) showed that mercury concentrations in fish muscle at part of the river between S8-S9 have increased twofold from 2011–2016 (Fig. 5). Such increases can occur when there is a permanent source of mercury to the river. Since the river is regulated and fish from upstream sections of the river are unlikely to pass through the dam, it is obvious that fish from the lower section of the river bioaccumulate mercury near the catch site. By length, the studied population of dace from the Boroo River is dominated by fish aged 4–5 years, the same age that dominated in previous studies. Thus, the new generation of dace accumulated more mercury than the previous one. This result can be explained by the fact that the local people use mercury in illegal gold mining.

Length is known to be a strong predictor of fish Hg concentration, as it can represent an individual's exposure time and often increased Hg content of prey items (Wiener et al. 2002). In contrast to dace from the Boroo River, significant correlations between the length and mercury content in the muscles of fish with a homogeneous diet (planktivores, piscivores, detritivores) were observed earlier under anthropogenic pollution (Bastos et al. 2015; Mills et al. 2019; Scerbo et al. 2005). This happens when mercury enters the body evenly throughout life. If the accumulation is more intense in non-predatory fish species in the early stages of life, and the diet changes significantly and the intake of mercury decreases, then the effect of biological dilution is observed, and with an increase in the length of the fish, the concentration of mercury in organs and tissues decreases. The large range between the maximum concentrations of mercury in dace of the Boroo Rriver in the same size category, and consequently, approximately the same age, can be explained by the fact that the intensity of mercury accumulation by fish is determined both by the concentration of mercury in food and by individual physiological and biochemical processes in the body.

The uneven intake of mercury into the food web along the river length is due to the different proportion of bioavailable mercury compounds at different sites. It is known that with an increase in the content of gross mercury under the influence of ASGM in bottom sediments, the content of MeHg also significantly increases (Marrugo-Negrete et al. 2015; Pinedo-Hernández et al. 2015). This explains the higher concentrations of mercury in mayflies at Boroo

River sites with a high mercury content in bottom sediments compared to sites with moderate or weak anthropogenic impact. However, in addition to MeHg, the content of forms of mercury inaccessible to biota also increases, which probably leads to a decrease in BASF values at stations in the middle reaches of the river. Earlier, it was shown in amphipods that the feeding rate of invertebrates decreases with increased concentrations of mercury in the sediment (Bundschuh et al. 2011), which may also be the reason for the decrease in BASF. In addition, mercury can enter the river ecosystem and become part of the food web with solid particles removed from the soil surface after heavy rains or snowmelt (Chalov et al. 2016). It is possible that this migration of mercury from adjacent landscapes is associated with increased BASF values at S3 and S10.

Despite the regression model on the dependence of the Hg content in the muscles of dace on the Hg content in bottom sediments being statistically significant ($p=0.001$), the low value of R^2-adjusted (0.49) requires caution in interpretation. Perhaps, this may be both the migration of fish along the river, and the different proportion of bioavailable mercury in different parts of the river. It is also impossible to adopt a similar model for mayfly larvae. The results of the regression analysis are consistent with the assumption of a different proportion of bioavailable mercury compounds in the bottom sediments at different sites. At the same time, the average mercury concentrations in dace for each station are significantly correlated with the mercury concentrations in mayflies, and the value of R^2-adjusted (0.74) allows us to adopt the model. Thus, mayfly larvae are one of the main sources of mercury for dace in the Boroo River. Earlier studies carried out on rivers of Mongolia showed that the basis of the diet of dace consists of benthic invertebrates, algae and terrestrial insects that have fallen into the water (mainly grasshoppers) and does not differ in individuals of different age (Chandra et al. 2005). Due to the small size of the Boroo river and, consequently, the limited choice of food types, benthic invertebrates may be the most likely source of mercury for dace, which is confirmed by regression analysis. The regression model also showed a decrease in BMF with an increase in mercury concentrations in sediments, but R^2-adjusted does not allow full interpretation. However, the general direction of the regression line is consistent with the assumption of a different proportion of bioavailable mercury compounds in areas with little or no anthropogenic influence on the concentration of mercury in sediments. In order to build a reliable model, it is necessary to increase the number of points in the future, including other rivers flowing in areas of intensive gold mining in Mongolia.

Conclusion

This is the first study of the distribution of mercury in ecosystem components along the linear gradient of the Boroo River under a legacy of gold mining and mercury spill in the middle reaches. The results of the study show that despite the fact that the mercury spill occurred more than 60 years ago, the Boroo River still has high levels of mercury compared to other rivers in Mongolia. The spatial distribution of mercury in the bottom sediments along the profile of the river better reflects the boundaries of mercury distribution than the larvae of mayflies and dace. Fish move along the river for long distances and apparently do not always accumulate mercury in the place of capture. Therefore, the concentration of mercury in the organs and tissues of fish in small rivers may not correctly indicate the location of potential sources of mercury.

The study showed that by 2016, the mercury content in the muscles of the Yelets from the areas below the ruins of the plant increased by 2 times compared to 2010-2012. The reason for this increase may be the supply of mercury as a result of illegal gold mining. Studies should be conducted in Boroo in the future to further assess trends in the mercury content of fish muscles.

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Table 1. The pH, organic matter content and mercury concentrations in bottom sediments at sampling sites

Site	Location	Sediments				Mayflies		
		n	pH	OM %	Hg±SD mg/kg dw	n	Hg±SD mg/kg dw	Abudance specimens/sample
S1	Upstream Boroo riverbed	3	7.70	7.0	0.010 ± 0.001		-	-
S2		3	7.57	7.4	0.005 ± 0.004	3	0.05 ± 0.01	280
S3		3	7.68	9.7	0.042 ± 0.001	3	0.27 ± 0.01	220
S4	Boroo riverbed near gold recovery plant	3	7.68	13.5	0.051 ± 0.001	5	1.53 ± 0.26	1400
S5		3	7.55	11.7	0.557 ± 0.059		-	-
S6		3	7.84	4.9	2.230 ± 0.044	2	2.94 ± 0.42	35
S7	Boroo riverbed downstream from ruins of gold recovery plant	3	7.76	6.6	1.103 ± 0.315		-	-
S8		3	7.81	8.3	1.310 ± 0.026		-	-
S9		3	7.38	12.6	1.697 ± 0.061	5	1.56 ± 0.27	1900
S10	Boroo river mouth	3	7.70	3.5	0.103 ± 0.007	3	2.36 ± 0.13	230
S11	Kharaa river bed	3	7.50	3.7	0.011 ± 0.008		-	-
S12		3	7.50	7.7	0.032 ± 0.021	2	0.13 ± 0.02	80

n – numbers of observations, OM – organic matter content, SD – standard deviation,

Table 2. The values of THg in Boroo river valley and surface sediments in rivers under gold mining impact reported by other researchers.

Location	THg concentration (ng/g)	Reference
	Mongolia	
Upper reaches and river's mouth of Boroo River	5-103	This study
Middle reaches of Boroo River, Mercury spill area	557–2230	This study
Middle reaches of the Boroo River	780	Kaus et al., 2017
	Indonesia	
Buru island, Wamsait River	548–9280	Male et al., 2013
	Kenya	
Rivers and streams Migori–Transmara gold mining complex	30–2380	Odumo et al., 2013
	Cote d'Ivoire	
Different river s under gold mining impact	2.4–147	Mason et al., 2019
	Colombia	
San Martin de Loba	100–232800	Olivero-Verbel et al., 2014
The Mojana region	198–1187	Pinedo-Hernández et al., 2015
	Peru	
Madre de Dios	13900-367200	Martinez et al., 2018
	Ecuador	
Nangaritza River's sediments high-exposure by gold mining activity	930–18600	(González-Merizalde et al., 2016)
Local background concentrations	20–30	
	Suriname	
Gold Mining area	1300–220	(Ouboter et al., 2012)

Figure captions

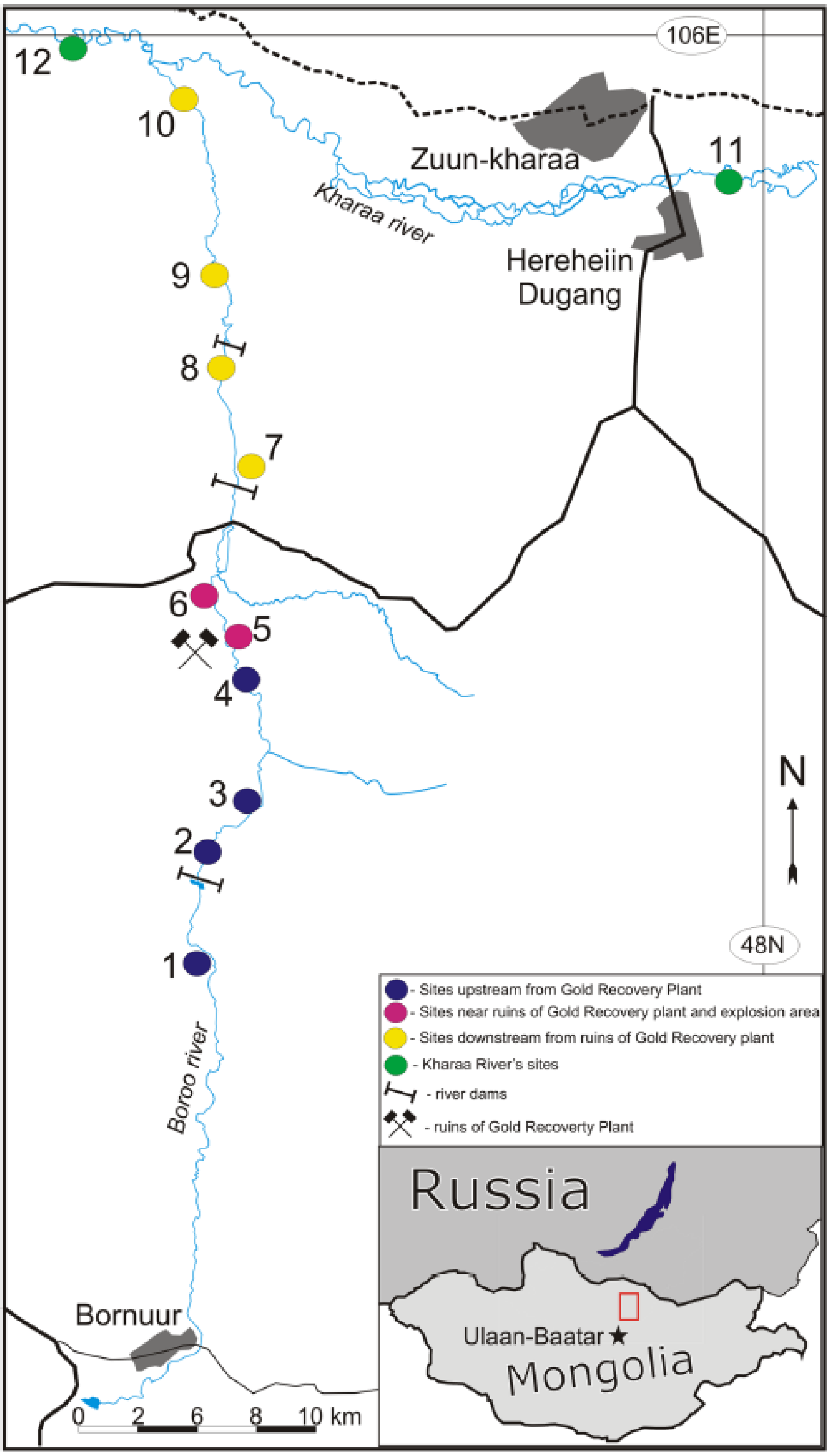

Figure 1. Locations of sampling sites along the Boroo river and the Kharaa river. Sites were selected to represent the full longitudinal profile of the Boroo river. Sites on the Kharaa were used as comparison with the Boroo river sites and to see whether mercury has moved out of the Boroo river. The Kharaa is a tributary of the Selenga river that eventually flows into Lake Baikal in Russia.

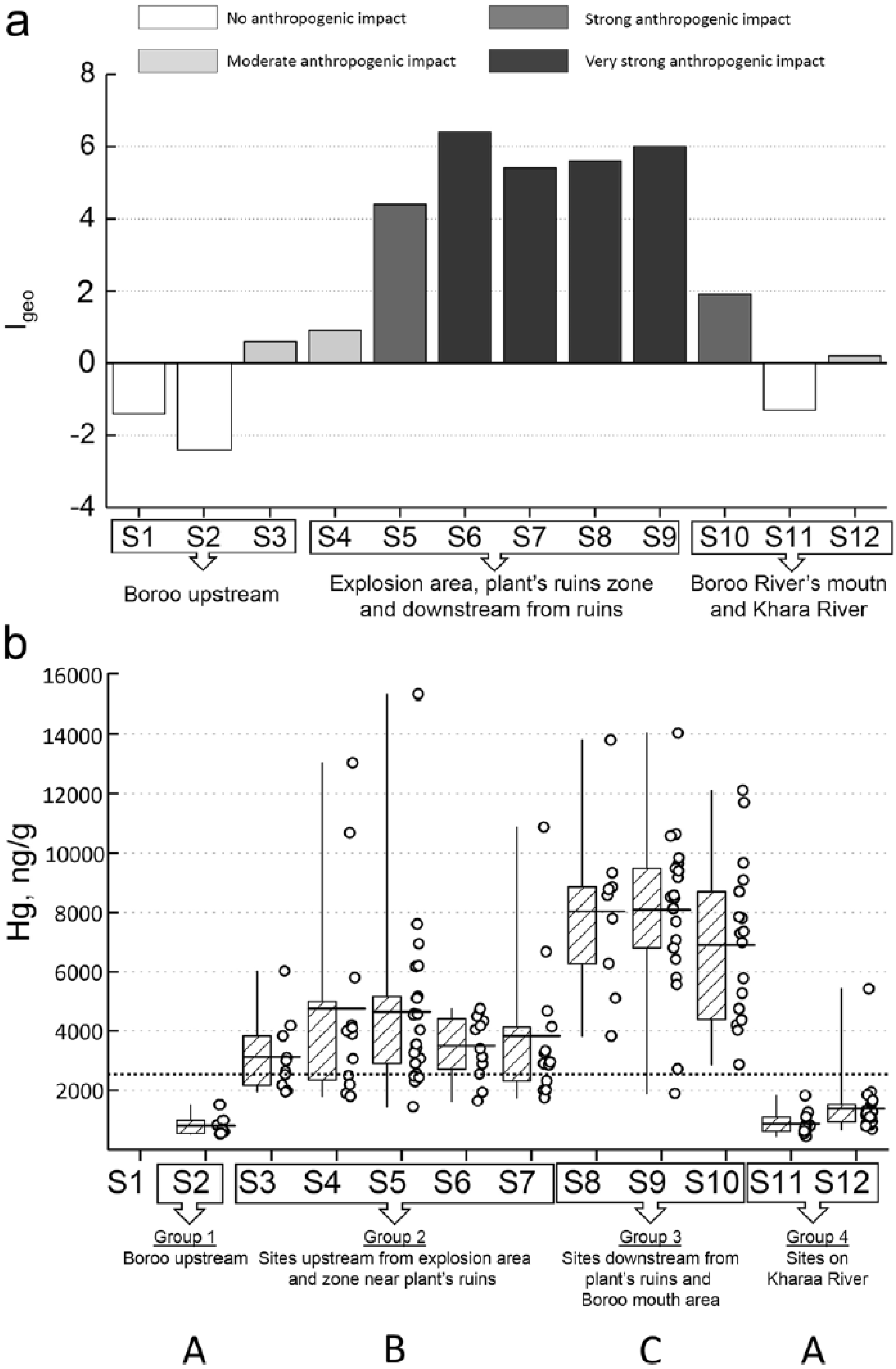

Figure 2. Plots of the Igeo Index (A) grouped anthropogenic impact and mercury content (ng/g dry weight) in dace muscle (B) collected from the different sites grouped by longitudinal location on the river. Box plots in B show median, quartiles, range and data points. Letters represent significance at $p < 0.05$. The dotted line indicates the permissible by US EPA level of mercury in fish suitable for food (460 ng/g ww = 2300 ng/g dw).

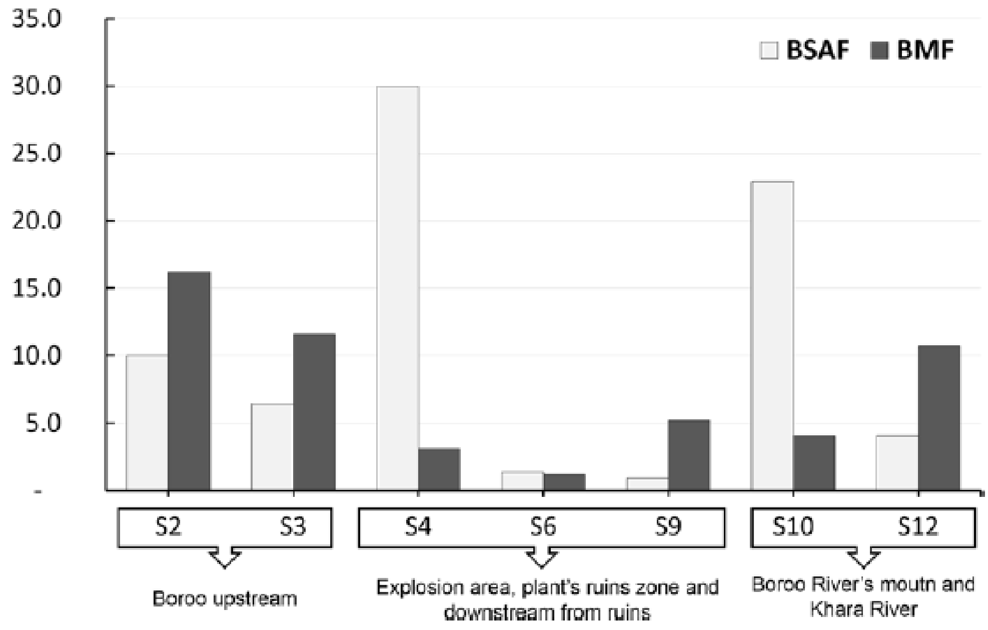

Figure 3. Histogram of BSAF and BMF scores based on the 7 sites along the river at which macroinvertebrates were collected and analyzed. Sites grouped as in figure 2.

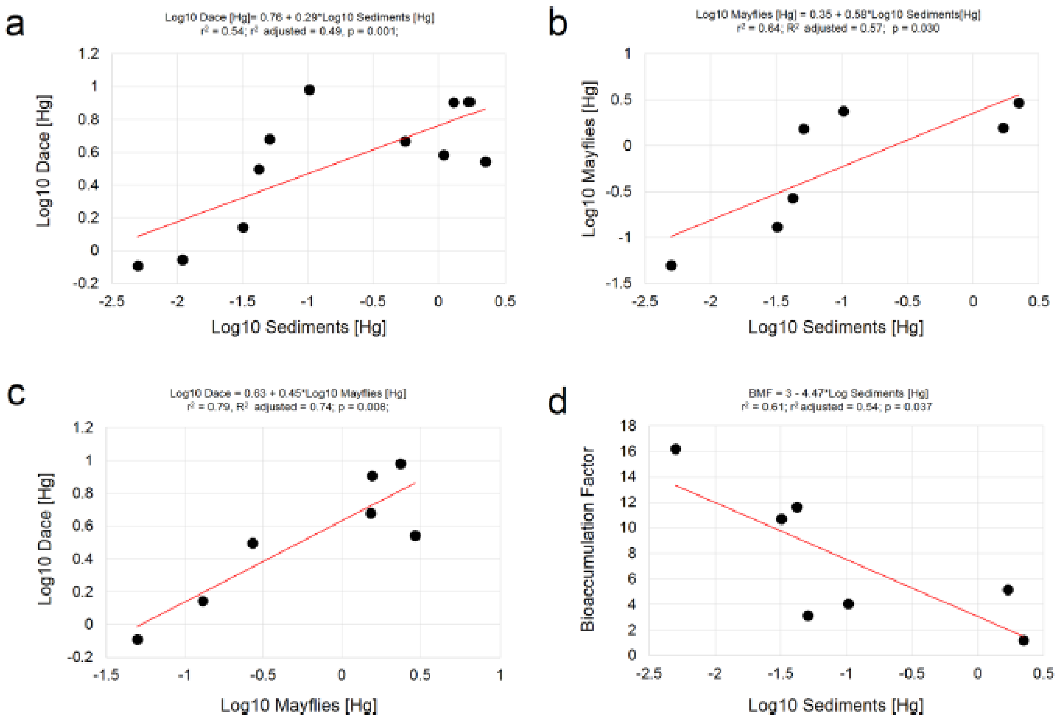

Figure 4. Correlations between log10-transformed mercury content in sediments and that in dace (A), sediments and mayflies (B), mayflies and dace (C), and sediments and bioaccumulation factor (D). Plots B-D based on the 7 sites where mayflies were collected.

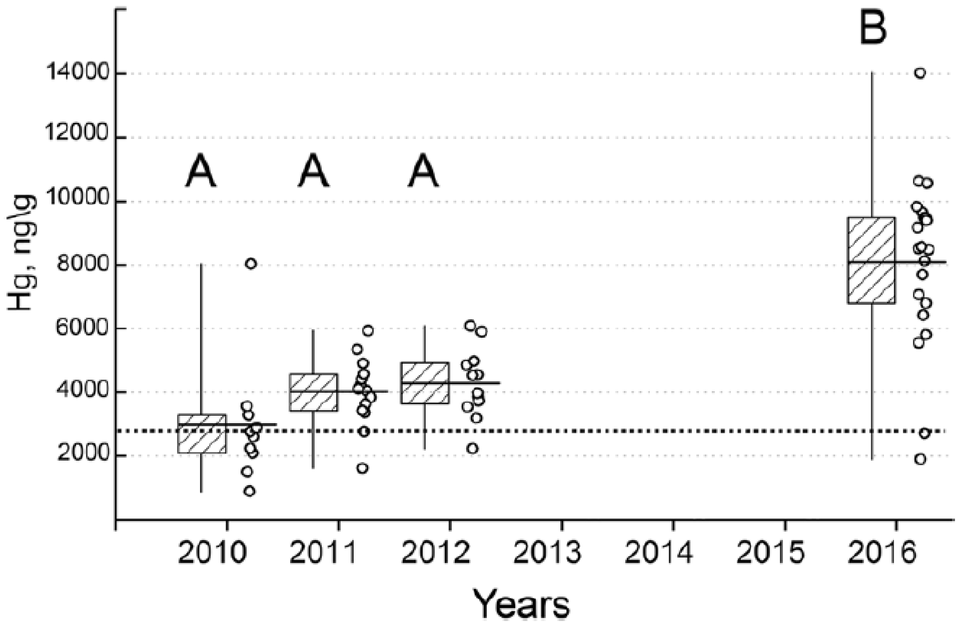

Figure 5. Box plots of mercury content (ng/g dry wt) in dace between 2010 and 2016. No studies were conducted in 2013-2015. Box plots represent median, quartiles, range and data points. Letters represent significance at $p < 0.05$. The dotted line indicates the permissible by US EPA level of mercury in fish suitable for food (460 ng/g ww = 2300 ng/g dw).