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Low and Declining Mercury in Arctic Russian Rivers

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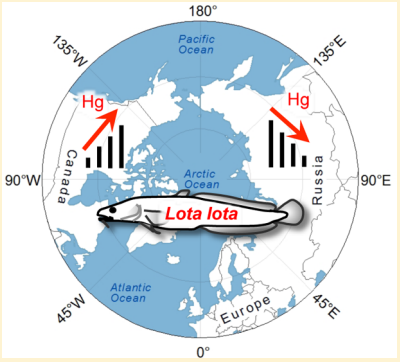
ABSTRACT: Mercury (Hg) dynamics in the Arctic is receiving increasing attention, but further understanding is limited by a lack of studies in Russia, which encompasses the majority of the pan-Arctic watershed. This study reports Hg concentrations and trends in burbot (*Lota lota*) from the Lena and Mezen Rivers in the Russian Arctic, and assesses the extent to which they differ from those found in burbot in arctic rivers elsewhere. Mercury concentrations in burbot in the Lena and Mezen Rivers were found to be generally lower than in 23 other locations, most of which are in the Mackenzie River Basin (Canada). Mercury concentrations in burbot in the Lena and Mezen Rivers also were found to have been declining at an annual rate of 2.3% while they have been increasing in the Mackenzie River Basin at annual rates between 2.2 and 5.1% during roughly the same time period. These contrasting patterns in Hg in burbot across the pan-Arctic may be explained by geographic heterogeneity in controlling processes, including riverine particulate material loads, historically changing atmospheric inputs, postdepositional processes, and climate change impacts.



INTRODUCTION

Mercury (Hg) contamination is a problem of growing interest in the Arctic. Atmospheric Hg entering arctic environments is transformed by microorganisms into methylmercury, which is a powerful toxin that bioaccumulates in biota and people.[1]

The current paradigm in biological Hg dynamics in the Arctic seeks to explain biological Hg levels based on processes related to food web structure and Hg emissions, atmospheric transport, and surface deposition.[1] Previous studies have shown that the origin and quantity of Hg reaching Arctic environments changed in the 1970s following the industrialization of Asia and implementation of Hg-emission regulations. Until the 1970s, atmospheric Hg amounts over the Arctic were on the rise and stemmed mostly from Europe and North America, but after the implementation of emission controls they stabilized or even began to decline and originated mostly from Asia.[1−5] However, such stable or declining atmospheric Hg trends have recently been found to differ from Hg in arctic biota, which have been found to be increasing at an overall annual rate of change of 0.8%.[6−8] This finding has led Macdonald and Loseto[8] to question the validity of the current paradigm by postulating that biological Hg levels in the Arctic are better explained by postatmospheric Hg depositional processes, including methylation and biomagnification, among others. Postatmospheric Hg depositional processes in the Arctic are generally complex and poorly understood, and are increasingly impacted by climate change.[8,9] Climate change induces many physical, biological, and ecological changes that further exacerbate the difficulties of understanding biological Hg dynamics in the Arctic.[10] For example, Hg in burbot (*Lota lota*) in the Canadian Arctic has increased over time, despite stable or declining atmospheric concentrations, because rising temperatures appear to have increased methylation rates.[11]

Despite these advances in knowledge, little consideration has been given to the problems created by the current uneven geographical distribution of research on Hg in the Arctic. Most biological Hg data sets and studies are for Europe and North America (i.e., western Arctic),[6] and there are no published data sets and studies for Russia (i.e., eastern Arctic), which covers the majority of the pan-Arctic watershed (Figure 1). Such disproportionate geographical emphasis limits current capacity to accurately understand biological Hg dynamics in the Arctic.

There are at least four reasons to believe that biological Hg dynamics vary across the Arctic. First, historical and geographic changes in atmospheric Hg emissions have produced a spatially and temporally heterogeneous pattern of net Hg deposition in the Arctic.[5,13,14] Second, historical trends in biological Hg have been found to follow a longitudinal gradient, with mostly increasing trends in the western Arctic and mostly declining trends in the eastern Arctic.[6] Third, there is large variation among arctic rivers in water quality, including particulate

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Figure 1. The pan-Arctic watershed (delineated in red), as defined by Holmes et al.,[12] showing the Mezen and Lena Rivers (in yellow). The black dot in the Mezen and Lena watersheds denotes the historical sampling locations for burbot.

organic carbon yields (normalization of the load to the watershed area), which is correlated to riverine Hg export.[15,16] Finally, atmospheric Hg inputs in the eastern Arctic may be influenced by the economic decline of the former Soviet Union, which collapsed in 1991, because, unlike arctic rivers in North America, many arctic rivers in Russia have industrial centers near their headwaters (Figure 1).[17,18]

The objective of this study was to (i) examine Hg concentrations and trends in burbot in two rivers of the Russian arctic, and (ii) assess the extent to which they differ from those in burbot in arctic rivers elsewhere. Burbot were chosen, because they are long-lived piscivores from the top of the food web, so they are expected to integrate many food web processes over relatively long periods of time.[19−21] Burbot are nonmigratory, so the sampled individuals can be assumed to represent conditions of the sampling locations.[22] Burbot are also one of only two freshwater fish species, together with pike (*Esox lucius*), that have a circumpolar distribution, but the only one for which historical Hg data exist in more than one locality in the pan-Arctic.[6] The watershed areas of the two rivers of the Russian arctic analyzed here, the Mezen (0.08×10^6 km^2) and Lena (2.4×10^6 km^2) Rivers, together encompass about 15% of the pan-Arctic watershed.[12] Because the burbot data analyzed here were collected in freshwater environments near the mouths of these two rivers, the data are expected to integrate many of the Hg processes occurring in a significant portion of the pan-Arctic watershed.

MATERIALS AND METHODS

To examine burbot Hg concentrations and trends for the Russian arctic and compare them to those reported elsewhere in the Arctic, three steps were followed. First, burbot Hg concentrations in the Russian arctic were quantified; second, a pan-Arctic data set on burbot Hg concentrations and trends was assembled; and third, burbot Hg concentrations and trends in the Russian arctic were calculated and compared to other data.

Quantifying Hg in Russia. An average of 17 (min = 9, max = 24) burbot were collected annually in the same locations between 1988 and 2001 in the Lena River (70.4° N, 127.2° E) and between 1980 and 2001 in the Mezen River (65.6° N, 44.6° E), totaling 221 samples from the Lena and 357 from the Mezen (Figure 1). The burbot possessed a 1:1 sex ratio, and measured between 80 and 100 cm in total length (mean = 86.3 and 87.4 cm in the Lena and Mezen Rivers, respectively); no change in mean total length occurred during the sampling period. Sampling was done using a longline ice-fishing method called "*nalimnik*" in November−December, which is the peak of burbot fishing season in these rivers. Sampled burbot were stored in hermetically sealed plastic bags, frozen, and transported for laboratory analyses, which were undertaken within two months from sampling.

All laboratory equipment was cleaned prior to analyses using concentrated HNO_3 and 6 M HCl. The chemicals used were of analytical grade or higher. Inorganic mercury (Hg(II)) working standards were prepared daily by dilution of a standard solution 1000 mg L^{-1} Hg (CertiPUR, Merck, Germany) in 2 M HNO_3. Pieces of burbot muscle (without the skin) weighing 20−30 g were extracted from each fish from a location adjacent to the dorsal fin, and were thawed sufficiently so that they could be cut into smaller pieces weighing 0.2−0.3 g. These small muscle samples were homogenized before analysis by digesting the tissue in 5 mL H_2SO_4/HNO_3 (4:1v/v) at 85−90 °C for 2 h, followed by potassium permanganate and then hydrogen peroxide treatment, as in previous studies.[23−26] Total mercury (THg) was measured following the cold vapor atomic absorption spectrometry method[27−29] at the 253.7 nm mercury line. This was accomplished by addition of a reductant, stannous chloride or sulfate.[30] The THg detection limit was 0.005 μg.g^{-1} w.w. of sample. Quality assurance/quality control (QA/QC) procedures included analyses of certified reference materials (CRMs) for THg from the National Research Council of Canada, the National Institute of Standards and Technology, and the International Atomic Energy Agency, Monaco (CRM range: 0.061−4.64 μg.g^{-1}). All CRMs measured values were ±5% of the CRM. These procedures also included analyses of duplicate samples and replicate blanks, which were run after approximately every 20 samples and replicate analysis of the same sample or blank were within ±5%.

Assembling a Pan-Arctic Data Set. Five studies that report burbot muscle Hg data for the pan-Arctic were selected and organized in two groups, depending on the degree of data comparability. The most important studies for this analysis comprise Evans et al.,[19] Carrie et al.,[11] and Rigét et al.[6] The data considered here from these three studies are based on samples of pieces of dorsal muscle (without the skin) from burbot of known total lengths collected during the winter (November−January). The samples were analyzed for THg using the cold vapor atomic absorption spectrometry method, as described above, at the laboratories of the Freshwater Institute of the Department of Fisheries and Oceans, in Winnipeg, Manitoba, Canada. Detection limits were at 0.005 μg.g^{-1} w.w., and replicates, blanks, and CRMs were used. Carrie et al.[11] state that duplicate samples and CRMs were run every eighth sample and that precision was better than 7% for each CRM. Evans et al.[19] state that QA/QC procedures included analysis of known analytical standards and CRMs, and that sample results were within accepted ranges, which is here interpreted to mean to meet the analytical rigor of Carrie et al.[11] and of the present Russian data. The data in Rigét et al.[6] considered here are from Carrie et al.[11] and Evans et al.[19]

The other group of studies comprises Amundsen et al.,[23] which measured burbot Hg in the Pasvik River on the

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Norwegian−Russian border, and Lockhart et al.,[30] which summarized burbot Hg data for many locations north of the Arctic circle across Canada. The data in Amundsen et al.[23] and Lockhart et al.[30] are based on the laboratory methods outlined for the first group, so they appear to be comparable. However, because Lockhart et al.[30] does not provide methodological details for all data sources, and Amundsen et al.[23] sampled burbot in the summer, full comparability of the data they report cannot be ensured here. Lockhart et al.'s[30] data for multiple years for some locations were pooled here per location.

Data Analyses. To estimate burbot Hg concentrations in the Russian arctic in a fashion comparable to those reported elsewhere in the Arctic, the data on burbot Hg concentrations in the Lena River were adjusted by the length of the burbot studied for Hg elsewhere in the Arctic. Regression analyses of total length and (natural log) Hg concentrations showed a positive relation for the Lena River ($R^2 = 0.36$, $p < 0.01$) but no relation for the Mezen River ($R^2 = 0.0$, $p = 0.86$). Although a relation between fish length and Hg concentrations was expected,[26] Carrie et al.[11] also did not observe it in a sample of 385 burbot. Burbot Hg data for the Lena and Mezen Rivers were compared to Lockhart et al.'s[30] length data at 60.4 cm, Carrie et al.'s[11] at 69.3 cm, and Evans et al.'s[19] at 63.4 cm. Because most of the selected studies did not report dispersion measures for the mean, the present study assessed whether the reported means were within the 95% confidence interval of the respective means for burbot Hg in the Lena and Mezen Rivers.

To investigate historical trends in burbot Hg in the Russian arctic, the PIA software[31] was applied using the statistical approach developed by Bignert et al.[32] and the specifications used in the Hg assessment done under the auspices of the Arctic Monitoring and Assessment Programme.[1,6] This allowed comparison of the results reported here for the Lena and Mezen Rivers with those found by Rigét et al.[6] for the Mackenzie River near Fort Good Hope and West Basin (1985−2008 and 1996−2007, respectively), both in the Mackenzie River Basin, Canada. PIA calculated: (a) the test for linear trends using ordinary (least-squares) log−linear regressions based on annual geometric means of Hg concentrations; and (b) the "adequacy" of the data sets to reveal long-term trends in Hg concentrations. Such adequacy was calculated by dividing the number of monitoring years available by the number of years of sampling required to detect a change in Hg concentration with a significance level of $p < 0.05$ and 80% statistical power.[32] According to Bignert et al.,[32] an adequacy index of ≥1 enables reliable trend analyses.

RESULTS AND DISCUSSION

Pan-Arctic Concentrations. Total Hg concentrations in burbot in the Lena and Mezen Rivers were found to be generally lower than those reported for burbot elsewhere in the Arctic. Mean length-adjusted Hg in the Lena River (0.05 $\mu g.g^{-1}$ w.w. at 63.4 cm and 0.06 $\mu g.g^{-1}$ w.w. at 69.3 cm) were lower than those in Fort Good Hope (0.33 $\mu g.g^{-1}$ w.w. at 69.3 cm) and West Basin (0.13 $\mu g.g^{-1}$ w.w. at 63.4 cm; Figure 2). Those in the Mezen River (0.15 $\mu g.g^{-1}$ w.w. at 69.3 and 63.4 cm) were lower than in Fort Good Hope but slightly higher than in West Basin (Figure 2). Considering data from Lockhart et al.[30] for 20 locations across Canada and Amundsen et al.'s[23] data for the Norway-Russian border, mean length-adjusted burbot Hg concentrations in the Lena River (0.05 $\mu g.g^{-1}$ w.w. at 60.4 cm) were among the lowest. Those in the Mezen River (0.15 $\mu g.g^{-1}$ w.w. at 60.4 cm) were intermediate, being lower than 10 other locations but higher than eight other locations (Figure 2).

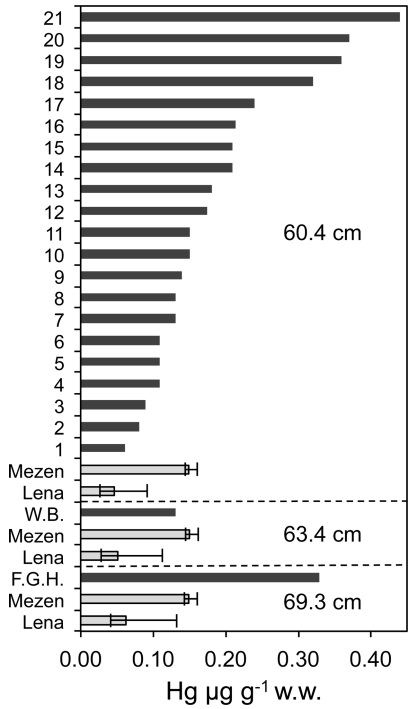

Figure 2. Mean burbot Hg concentrations in the Lena and Mezen Rivers in Russia are compared to available data across the pan-Arctic at three length-adjusted measures. The estimated mean and associated 95% confidence limits are given for the data from the Lena and Mezen Rivers in Russia. Only the means are given for the data from Fort Good Hope (F.G.H.)[11] near the Mackenzie River, West Basin (W.B.)[19] in Great Slave Lake, and 22 locations across Canada and Europe,[23,30] as dispersion measures were not available. The 21 locations are (1) Trout, (2) Great Slave, Resolution Bay, (3) Marsh, (4) Yukon River (Takhini), (5) Colville, (6) Aubry, (7) McEwan, (8) Great Slave, Fort Resolution, (9) Great Slave, Lutsel K'e, (10) Slave River, (11) Great Slave AR2, (12) Great Slave All, (13) Belot, (14) Willow, (15) Mackenzie Delta, (16) Manuel, (17) Fox, (18) Laberge, (19) Kelly, (20) Pasvik River (Norway-Russia border), and (21) Mackenzie River at Ramparts.

Burbot Hg concentrations in the Russian arctic were below the safe levels issued by Health Canada and the U.S. Environmental Protection Agency for commercial sale of fish (0.5 $\mu g\ g^{-1}$ w.w.) and also for people who consume large amounts of fish (0.2 $\mu g\ g^{-1}$ w.w.). Thus, unlike other freshwater fishes in the Arctic,[33] burbot in the Lena and Mezen Rivers appear to be safe for consumption with respect to Hg.

Temporal Trends. The regression analyses indicated that Hg concentrations in burbot in the Lena and Mezen Rivers declined at an annual rate of 2.3% in both rivers ($r^2 > 0.6$, $p < 0.001$, Figure 3). The adequacy and power indexes calculated for these two time-series data sets, which were equal to or greater than 1.6 and 2.7%, respectively, indicated these trends are robust (Table 1). Geometric mean Hg concentrations in burbot declined by 38% in the Mezen River from 0.165 $\mu g\ g^{-1}$ w.w. in 1980 to 0.101 $\mu g.g^{-1}$ w.w. in 2001, and by 26% in the Lena River from 0.129 $\mu g\ g^{-1}$ w.w. in 1988 to 0.095 $\mu g\ g^{-1}$ w.w. in 2001.

These results support the longitudinal gradient established by Rigét et al.[6] in which biological Hg trends are mostly increasing in North America and Greenland and mostly declining in the

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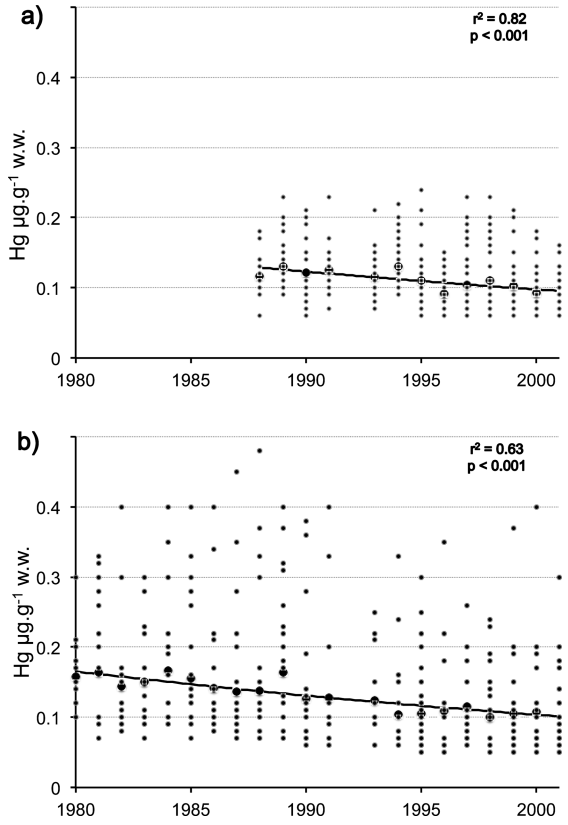

Figure 3. Historical decline of Hg in burbot in (a) the Lena River and (b) the Mezen River. Gray dots represent observed data, black dots are geometric means, and the lines are log−linear regressions.

Table 1. Trend Statistics of Historical Hg Concentrations in the Lena and Mezen Rivers[a]

statistics	Mezen	Lena
adequacy	2.6	1.6
power	2.8%	2.7%
slope	−2.3% (−3.5, −1.1)	−2.3% (−2.9, −1.8)

[a]"Adequacy" reports an index the capacity of the data set to reveal long-term trends. "Power" reports the lowest detectable trend change in the current time series with a significance level of $p < 0.05$ and 80% statistical power. "r^2" reports the coefficient of determination of the log-linear regression with a p-value for a two-sided test. "Slope" reports the slope of the significant log−linear regression together with 95% confidence intervals.

European arctic. Such a gradient is maintained for burbot, all freshwater fishes, and across all species for which data are available.[6] Among the four burbot muscle Hg time-series data sets, the two in Russia showed declining trends (Figure 3), whereas the two in Canada showed increasing trends at mean annual rates ranging from 2.2 to 5.1%.[6] Considering the Russian burbot Hg data presented here together with the data for freshwater fishes analyzed by Rigét et al.,[6] most data sets in the western Arctic (9 out of 14) showed increasing trends and most data sets in the eastern Arctic (3 out of 4) showed declining trends. Similarly, if the Russian burbot Hg data are considered together with the data analyzed by Rigét et al.[6] for all animal species, biological Hg concentrations are seen to mostly decline in the eastern Arctic and mostly increase in the western Arctic.

Pan-Arctic Heterogeneity. Why are Hg concentrations in burbot in the Russian arctic rivers generally lower than in burbot elsewhere in the Arctic? And why have biological Hg concentrations mostly declined in the eastern Arctic and mostly increased in the western Arctic? Many factors may heterogeneously influence burbot Hg dynamics across the pan-Arctic watershed. However, there are no ancillary environmental data from the time period of this study (e.g., relevant water chemistry, Hg concentrations in water or sediment) with which to jointly analyze the burbot Hg data here reported, making the following explanations tentative.

Pan-Arctic differences in burbot Hg may be explained by differences in water quality, geological bedrock formations, and proximity to polluting sources, among other factors (e.g., food webs). Many water quality parameters, including pH and dissolved and particulate organic carbon, which are known to vary across the Arctic,[15,16] strongly control Hg availability to methylation, and hence can be expected to explain pan-Arctic differences in burbot Hg, in part at least. The presence of metallurgic industries in the Murmansk region of Russia and smelter companies in the Pasvik watershed can be assumed to explain the high burbot Hg concentrations observed in the Pasvik River on the Norway-Russian border (no. 21 in Figure 2). High levels of heavy metal contamination have been recorded in the sediment and water of this watershed.[34,35] Finally, the type of geological bedrock formations is expected to explain some of the observed variation in burbot Hg (Figure 2). Metamorphic, intrusive, and volcanic bedrocks have been shown to lead to higher fish Hg concentrations than sedimentary bedrocks.[30]

The declining Hg trends observed in burbot in the Lena and Mezen Rivers are here suggested to be primarily explained by a combination of historical reductions of atmospheric inputs and increased water temperatures. In line with the prevailing paradigm,[1] it is expected that the declining Hg concentrations in burbot in the Lena and Mezen Rivers are due to declining atmospheric Hg inputs stemming from the implementation of Hg emission controls and the economic decline of the former Soviet Union. Atmospheric Hg inputs in the Arctic are thought to have stabilized and even started to decline since the mid 1970s[4,5] and the economic decline and subsequent collapse of the former Soviet Union lowered industrial polluting activity near the headwaters of the Lena and Mezen Rivers. Our previous studies implicated the economic collapse of the Soviet Union in dramatic historical declines in organochlorine pesticides (e.g., DDT) in water and burbot in eastern arctic rivers, including the Lena and Mezen Rivers, between 1988 and 1996.[36,37] At the same time, it is expected that the observed declining Hg trends in burbot in the Lena and Mezen Rivers may be due to increased river water temperatures caused by climate change and construction of dams, which have been recorded in the Lena River.[38] Increased temperatures increase fish body growth rates in a manner similar to that documented for pike across the globe,[39] thereby decreasing fish Hg concentrations through reduced exposure to Hg.[10,40] Although increased water temperatures are expected to have influenced burbot Hg dynamics in the Lena and Mezen Rivers, it is unknown why they contrast with climate change effects observed in the Mackenzie River, where increased methylation rates have been suggested to have led to increases in Hg concentrations in burbot.[11]

In summary, this study suggests that there is a significant amount of heterogeneity in biological Hg dynamics across

rivers of the pan-Arctic. Hg concentrations in burbot are influenced by many controlling processes, including historically changing atmospheric inputs, postdepositional processes, and global climate change effects. Geographic heterogeneity in such controlling processes have the potential to explain why burbot Hg concentrations vary among rivers and are generally lower in the Russian Arctic, and why Hg concentrations in burbot and other biota have been declining in the eastern Arctic and increasing in the western Arctic. Understanding how biological Hg dynamics varies throughout the pan-Arctic thus appears to be essential to develop a paradigm that adequately explains arctic biological Hg concentrations.

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Notes

The authors declare no competing financial interest.

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