Environmental Science & Technology

Article

Cite This: Environ. Sci. Technol. XXXX, XXX, XXX-XXX

pubs.acs.org/est

Temporal and Longitudinal Mercury Trends in Burbot (*Lota lota*) in the Russian Arctic

Alexander R. Pelletier,*[,†] Leandro Castello,[†] Alexander V. Zhulidov,[‡] Tatiana Yu. Gurtovaya,[‡] Richard D. Robarts,[§] Robert M. Holmes,[∥] Daniel A. Zhulidov,[‡] and Robert G. M. Spencer[⊥]

[†]Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061, United States

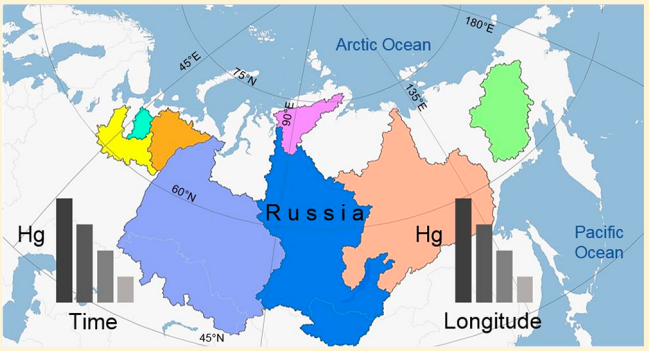
[‡]South Russian Centre for Preparation and Implementation of International Projects, Rostov-on-Don, Russia

[§]World Water and Climate Foundation, Saskatoon, Saskatchewan Canada

[∥]Woods Hole Research Center, Falmouth, Massachusetts 02540, United States

[⊥]Florida State University, Tallahassee, Florida 32306, United States

ABSTRACT: Current understanding of mercury (Hg) dynamics in the Arctic is hampered by a lack of data in the Russian Arctic region, which comprises about half of the entire Arctic watershed. This study quantified temporal and longitudinal trends in total mercury (THg) concentrations in burbot (*Lota lota*) in eight rivers of the Russian Arctic between 1980 and 2001, encompassing an expanse of 118 degrees of longitude. Burbot THg concentrations declined by an average of 2.6% annually across all eight rivers during the study period, decreasing by 39% from 0.171 $\mu g\ g^{-1}$ wet weight (w.w.) in 1980 to 0.104 $\mu g\ g^{-1}$ w.w. in 2001. THg concentrations in burbot also declined by an average of 1.8% per 10° of longitude from west to east across the study area between 1988 and 2001. These results, in combination with those of previous studies, suggest that Hg trends in Arctic freshwater fishes before 2001 were spatially and temporally heterogeneous, as those in the North American Arctic were mostly increasing while those in the Russian Arctic were mostly decreasing. It is suggested that Hg trends in Arctic animals may be influenced by both depositional and postdepositional processes.



INTRODUCTION

Mercury (Hg) contamination is a matter of serious concern in the Arctic.[e.g.,1−4] A meta-analysis of 83 long-term data sets showed that Hg in Arctic animals, predominantly in the toxic form of methylmercury (MeHg), has increased at a median annual rate of 0.6% between 1968 and 2008.[3,4] Increasing rates of Hg poisoning have consequently been documented in animals and humans throughout the Arctic.[5,6]

Rising Hg trends in Arctic animals were first suspected to be due to increasing emissions of gaseous elemental mercury (GEM) to the atmosphere.[1,3] However, Hg concentrations in some European and North American Arctic locations have increased since the 1970s[3,4,7,8] despite stable or even declining GEM concentrations in these locations during the same period.[9,10] This lack of correlation between atmospheric GEM and Hg concentrations has also been observed elsewhere, such as in over 800 lakes in Minnesota, United States, where increasing Hg trends[11] occurred with concurrent declines in GEM emissions.[12] The complex dynamics of Hg concentrations in light of Hg deposition and GEM concentrations in Arctic freshwater ecosystems have more recently led to the notion that depositional Hg processes may be less important controls of Hg concentrations in Arctic animals than postdepositional ecosystem processes such as Hg methylation rates and food web dynamics.[7,8,13−15] In support of this notion, Hg concentrations in Arctic freshwater ecosystems have been found to correlate negatively with temperature,[8] positively with Hg methylation rates by sulfur-reducing bacteria,[16] and nonlinearly with concentrations of dissolved organic carbon (DOC).[e.g.,17]

While the bulk of research on the topic has focused on the mechanistic processes leading to Hg concentrations in Artic animals, relatively little attention has been given to understanding broad spatial and temporal patterns of Hg throughout the entire pan-Arctic region. A lack of data for the Russian Arctic has hampered understanding of Hg dynamics in about half of the pan-Arctic watershed.[18] The prevailing notion that Hg concentrations in the Arctic have been mostly increasing is based on data from Alaska and Canada where most data are readily available. But in Greenland and northern Europe, Hg trends have been stable or even declining.[3,4,14,15,19] A study of

Received: July 31, 2017
Revised: October 3, 2017
Accepted: October 28, 2017
Published: October 30, 2017

DOI: 10.1021/acs.est.7b03929
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

Table 1. Sampling Locations, Effort, And Yields from Eight Russian Arctic Rivers[a]

river	sample latitude (°N)	sample longitude (°E)	watershed area (million km^2)	sampling years*	mean annual sampling yields (min, max)	mean total length (cm)
North Dvina	64.5	40.6	0.29	1980−2001	16.8 (13, 22)	87.6
Mezen	65.6	44.5	0.08	1980−2001	15.4 (9, 20)	87.4
Pechora	67.7	53.1	0.31	1980−2001	14.0 (8, 19)	87.6
Ob'	66.5	66.6	2.95	1988−2001	18.2 (16, 24)	87.1
Yenisey	69.4	86.2	2.56	1988−2001	21.2 (19, 25)	87.4
Pyasina	70.5	89.2	0.18	1988−2001	14.0 (14, 14)	86.4
Lena	70.4	127.2	2.40	1988−2001	24.1 (20, 28)	86.3
Kolyma	68.7	158.7	0.65	1988−2001	12.0 (12, 12)	87.2

[a]Rivers are arranged from west (top) to east (bottom) across the Russian Arctic. *Fish were sampled every year within the specified time period except for 1992, in which no fish were sampled from any of the rivers.

Hg in relation to longitude found that Hg has been mostly increasing in Alaska and Canada and mostly stable or decreasing in Greenland and northern Europe.[4] This trend was recently supported by newly published data for the Russian Arctic, showing that Hg concentrations in burbot (*Lota lota*) from the Lena and Mezen Rivers declined by 2.3% annually between 1980 and 2001.[20]

Nevertheless, question remains as to whether Hg trends throughout the Russian Arctic region have behaved similarly to the declining Hg trends observed in the Lena and Mezen rivers.[20] Furthermore, the previously reported longitudinal gradient in Hg trends was based mostly on data for the North American Arctic and many of the data sets used in that analysis were either statistically inadequate[21] or lacked statistical significance,[4] putting into question the validity of the longitudinal trend to the Russian Artic. An analysis of new Hg data in burbot throughout the Russian Arctic region will provide insights about the degree to which temporal and longitudinal Hg trends observed elsewhere in the Arctic hold true throughout the entire pan-Arctic region.

This study quantified temporal and longitudinal trends in Hg concentrations in burbot throughout the Russian Arctic region using one of the largest Arctic Hg data sets currently available in terms of the number of specimens and the number of continuous years of data collection.[3,4] Data were collected between 1980 and 2001 from eight rivers that encompass the majority (118 degrees of longitude) of this region. While Castello et al.[20] reported temporal Hg trends in burbot from the Lena and Mezen rivers during the same time period, this study reports cumulative temporal and longitudinal Hg trends in burbot in much of the Russian Arctic, making use of data for six previously unstudied rivers. Together with existing data from the North American and European Arctic regions, our study facilitates discussion of Hg dynamics across the entire pan-Arctic region, as contrasted with current analyses of Arctic Hg dynamics which are limited to the national or hemispheric scale at best. Additionally, our data will allow for broader-scale temporal comparisons between historical Hg dynamics presented here and more recent Hg data to be collected from burbot and other species in the future. Burbot are ideal fish for comparing Hg trends among multiple Arctic locations because they inhabit freshwater ecosystems throughout the pan-Arctic region.[22] They are also long-lived apex predators that occupy small home ranges, so Hg trends in these fish are expected to represent cumulative food-web processes affecting Hg cycling from their respective sampling locations.[20,22,23]

MATERIALS AND METHODS

Study Area. Burbot were sampled near the mouths of the North Dvina, Mezen, Pechora, Ob', Yenisey, Pyasina, Lena, and Kolyma Rivers (from west to east) to fulfill internal directives for heavy metal monitoring from the Russian State Service of Observation and Control of Environmental Pollution, as well as the Federal Russian Service for Hydrometeorology and Environmental Monitoring. These rivers are the eight largest in the Russian Arctic according to watershed area, covering a total drainage area of 9.3 million km^2 (Table 1). Together our sites encompass a watershed area of about half of the total pan-Arctic watershed as defined by Holmes et al.,[18] covering the entire expanse of the Russian Arctic from west to east (Figure 1). All eight rivers flow into the Arctic Ocean.

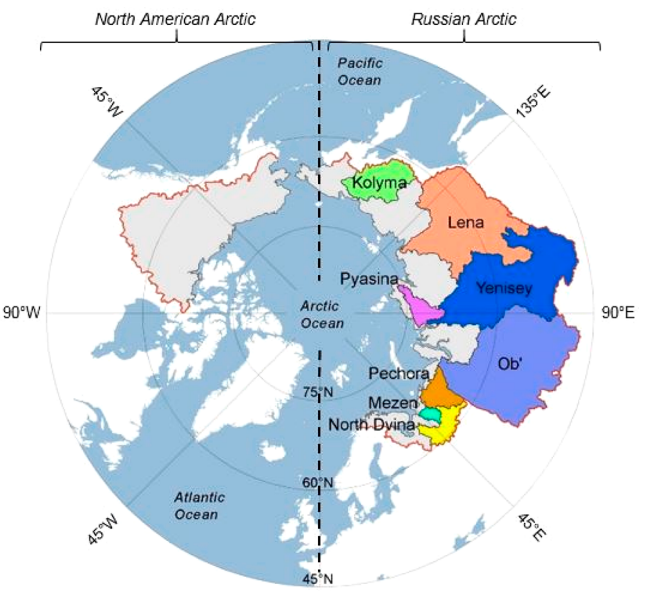


Figure 1. Map of the pan-Arctic region. The geographic north pole is in the center, with the hashed vertical line representing the prime meridian. Colored regions indicate sampled watersheds, all of which lie within the Russian Arctic. The pan-Arctic watershed, as defined by Holmes et al.,[18] is outlined in red.

The vast majority of the watershed area which drains into these eight rivers is mostly removed from direct human influence. However, human populations and industrial activities occurring throughout the study area are more prevalent in the westernmost watersheds (North Dvina, Mezen, and Pechora rivers) as compared with the central (Ob', Pyasina, and Yenisey

DOI: 10.1021/acs.est.7b03929
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

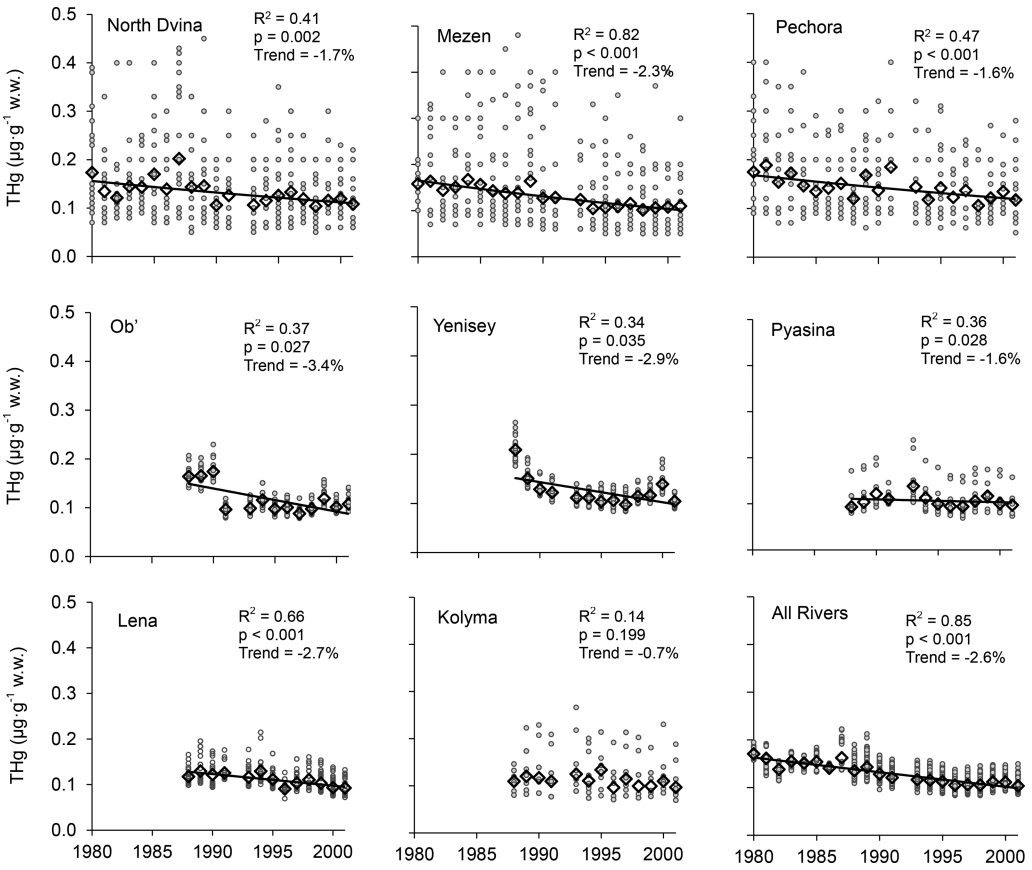


Figure 2. Temporal THg trends in burbot from eight Russian Arctic rivers, arranged from west to east (top to bottom, then left to right). Gray dots represent observed data, black diamonds represent geometric mean THg concentrations, and lines represent log−linear fit models. Trends represent mean annual percent changes in THg concentrations as a function of sampling year. Trends from the Mezen River are from Castello et al.[20].

rivers) and easternmost watersheds of the Russian Arctic (Lena and Kolyma rivers). Despite this, the central and eastern Russian watersheds contain several hydroelectric power plants (HPPs) which the western Russian watersheds lack. These include the Novosibirsk HPP on the Ob' River, the Krasnoyarsk and Sayano-Shushenskaya HPPs on the Yenisey River (the latter of which is the largest power plant in Russia), the Vilyuy HPP on the Vilyuy River (a tributary of the Lena River), and the Kolyma HPP on the Kolyma River. Dams and HPPs have been shown to increase Hg concentrations for many years after construction in fishes including burbot from northern latitudes.[24,25]

Sampling. A total of 2135 burbot were collected from the eight rivers between 1980 and 2001. A total mean of 46 burbot (15 per river, min = 10, max = 20) were collected annually from the Northern Dvina, Mezen, and Pechora Rivers between 1980 and 1987. A total mean of 136 burbot (17 per river, min = 8, max = 28) were collected annually from all eight rivers between 1988 and 2001. No data were collected in 1992 (Table 1). The majority of burbot were collected using a traditional longline ice fishing technique called *nalimnik* and all samples were collected between October and January of each year, which coincides with the peak of the burbot fishing season in the region. Sampled fish had a 1:1 sex ratio and measured between 80 and 100 cm total length (TL). Mean TL of sampled fish did not vary between rivers or between years during this study ($p > 0.05$). Dorsal muscle samples between 20 and 30 g each were removed from each fish from an area adjacent to the dorsal fin and skinned to isolate the muscle tissue for analysis. Tissue samples were frozen in hermetically sealed plastic bags and transported to the laboratory for processing. All chemical analyses were conducted within two months of arrival at the laboratory in order to avoid archiving samples.

Processing. Laboratory processing of Hg data followed identical gprocedures as those described in a previous study[20] and did not change throughout the duration of this study. Prior to analyses, all laboratory equipment was cleaned with concentrated HNO_3 and 6 M HCl. Working standards for inorganic mercury (Hg(II)) were prepared daily by diluting a standard solution of 1000 mg L^{-1} Hg (CertiPUR, Merck, Germany) in 2 M HNO_3; all chemicals used were of analytical grade or higher. The samples of burbot muscle without skin were thawed so the pieces could be cut into smaller sections weighing 0.2−0.3 g. These smaller muscle samples were homogenized prior to analysis in 5 mL H_2SO_4/HNO_3 (4:1 v/v) at 85−90 °C for 2 h, followed by potassium permanganate and then hydrogen peroxide treatment, as in previous studies.[13,26−28] Measurements of total mercury (THg) were conducted under wet weight (w.w.) conditions via the cold vapor atomic absorption spectrometry method[29−31] at the 253.7 nm Hg line. For all samples, this was accomplished by addition of a reductant, either stannous chloride or sulfate.[32] Although MeHg would have preferably been analyzed in the lab, it is expected that 95% or more of THg in these tissue samples is present in the MeHg form.[33] Quality assurance/quality control (QA/QC) procedures included analysis of a suite of certified reference materials (CRMs) for THg from the National Research Council of Canada, the National Institute of

DOI: 10.1021/acs.est.7b03929
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

Standards and Technology, and the International Atomic Energy Agency, Monaco (CRM range: 0.061−4.64 $\mu g \cdot g^{-1}$ w.w.). All CRMs measured values were ±7% of the CRM. QA/QC 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%. The THg detection limit of the outlined method is 0.005 $\mu g \cdot g^{-1}$ w.w. of sample.

Statistical Analyses. THg concentration data were analyzed using the Plot and Image Analysis software.[34] To investigate temporal trends in THg concentrations in burbot, annual geometric means of THg concentrations were fitted with log−linear regression models with year as a predictor of THg concentration (Figure 2). A total of nine models were developed: one for each river and one as the geometric mean of all eight rivers together. Although no interannual variation in mean fish weight or length was detected over time in any of the rivers ($p > 0.05$), intra-annual variation in fish weight was high, as burbot measuring 100 cm TL weighed an average of 40% more than burbot measuring 80 cm TL (mean total weight = 7.0 and 5.0 kg, respectively). Therefore, total fish weight was added as a covariate in all models in which THg concentrations correlated positively with fish weight, including the Kolyma, Lena, Ob', Pyasina, and Yenisey Rivers, as well as the cumulative model ($p < 0.05$).

To investigate longitudinal trends in THg concentrations, the same log−linear regression model described above with total fish weight as a covariate was fit with the exception that longitude was used as a predictor of THg concentration (Figure 3). Only THg data from 1988 to 2001 were included in the longitudinal model because all eight rivers were sampled during this period. The reliability of resulting conclusions about temporal and longitudinal THg trends was assessed by calculating the adequacy of the data, which is defined as the number of sampling years divided by the number of years required to detect an annual 5% change in contaminant concentration at $\alpha = 0.05$ and statistical power = 80%.[21] Adequacy values greater than or equal to 1.0 indicate reliable trends.[21]

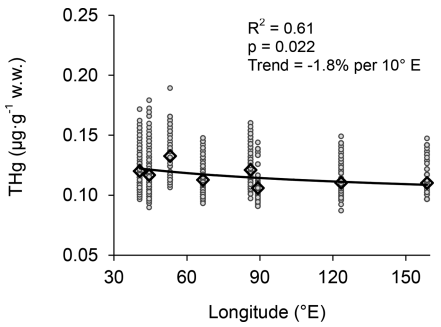


Figure 3. Longitudinal THg trends across Russian Arctic rivers. Gray dots represent observed data, black diamonds represent geometric mean THg concentrations, and line represents log−linear fit model.

RESULTS AND DISCUSSION

Temporal Trends. Concentrations of THg in Russian Arctic burbot during the sampling period declined in seven of the eight rivers by between 1.6 and 3.4% annually (Table 2, Figure 2). THg levels in the Kolyma River also appeared to decline throughout the study period, although trends were not significant ($p = 0.199$). Unlike many Hg time series data sets evaluated in the Arctic,[3,4] all temporal trends analyzed here are robust as adequacy values were greater than 1.0 (Table 2).

These temporal trends are consistent with those of Castello et al.[20] and indicate that THg trends throughout most of the Russian Arctic have declined between 1988 and 2001. These decreasing THg trends coincide with a large-scale decline in both the number and activity of industrial operations (namely machinery plants, oil and gas enterprises, mining and processing of metals and gems, and paper and pulp mills) which surrounded the collapse of the former Soviet Union (USSR) in 1991. For instance, Russian economic output declined by 6.3% annually from 1989 to 1998, amounting to a cumulative decline of about 44% during this time.[35] This decreasing industrial activity may have reduced both atmospheric GEM emissions and release of Hg-containing effluent into nearby rivers,[9,36] thereby decreasing burbot exposure to Hg over time during the study period. Reductions in Hg-containing effluent from an industrial plant in Ontario, Canada had comparable effects on Hg levels in four fish species in the English-Wabigoon river system.[37] At that site, Hg levels declined steadily in all four fish species throughout the 15 year period after the effluent deposits were ceased.[37] The collapse of the former USSR may also help explain the sudden and more drastic decrease in THg concentrations in burbot around 1991 in the Ob' River Basin (Figure 2).

Water temperatures may have also influenced THg trends in Russian Arctic burbot presented here. Increasing water temperatures have been suggested to increase Hg methylation rates by sulfur-reducing bacteria, which increases the amount of bioavailable Hg in the ecosystem.[16] Additionally, rising water temperatures lead to increased food consumption rates in burbot,[38,39] which would be predicted to increase THg exposure in these fish. However, THg trends in the Russian Arctic have decreased from 1980 to 2001 despite concurrent increases in water temperatures in at least the Mezen, Pechora, and Lena river basins during this time[40,41] (water temperatures for the remaining five rivers during this time period remain unavailable). One study from the Mackenzie River Basin in Canada reported a similar inverse relationship between water temperature and Hg trends in both burbot and lake trout (*Salvelinus namaycush*) from the early 1990s to 2012.[8] It is proposed here, as has been suggested previously,[20] that biodilution may have influenced temporal THg trends in Russian Arctic burbot. For example, increasing stream temperatures in parts of the Russian Arctic could increase growth rates of phytoplankton in these rivers, in turn decreasing the concentration of Hg per algal cell.[42] Reductions in Hg concentrations with increased growth rates have also been observed in periphyton[43] and zooplankton.[44] This reduction in Hg at the bottom of the food web would decrease dietary Hg exposure to low-level predators which consume these plankton, and to tertiary predators such as burbot higher up the food web. Additionally, increasing stream temperatures may have caused burbot to grow at faster rates, as has been documented in other coldwater fishes[45] Increased growth rates resulting from warmer water temperatures could have allowed these burbot to recruit to the 80 cm TL threshold (the minimum length at which fish were kept for Hg analyses in this study) at progressively younger ages over the course of the sampling period, decreasing their exposure to THg and thereby reducing their THg concentrations.[20] However, further mechanistic studies are required to clarify the effects of increasing water

DOI: 10.1021/acs.est.7b03929
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

Table 2. Annual Trends in THg Concentrations in Burbot in Eight Rivers of the Russian Arctic[a]

river	geometric mean [THg] ($\mu g \cdot g^{-1}$ w.w.) at start of sampling period	geometric mean [THg] ($\mu g \cdot g^{-1}$ w.w.) in 2001	cumulative change in mean [THg]	annual trend in mean [THg] (95% confidence interval)[c]	adequacy[d]
North Dvina[b]	0.173	0.107	−38%	**-1.7%** (−2.6, −0.7)	3.0
Mezen[b]	0.158	0.110	−30%	**-2.3%** (−2.8, −1.8)	3.5
Pechora[b]	0.175	0.117	−33%	**-1.6%** (−2.5, −0.8)	3.0
Ob'	0.164	0.107	−34%	**-3.4%** (−6.3, −0.4)	1.4
Yenisey	0.209	0.104	−50%	**-2.9%** (−5.5, −0.2)	1.6
Pyasina	0.093	0.097	4%	**-1.6%** (−3.0, −0.2)	2.2
Lena	0.117	0.093	−21%	**-2.7%** (−4.0, −1.4)	2.2
Kolyma	0.110	0.097	−12%	−0.7% (−1.9, 0.4)	2.2
All Rivers	0.171	0.104	−39%	**-2.6%** (−3.1, −2.1)	3.5

[a]Rivers are arranged from west (top) to east (bottom) across the Russian Arctic. Mean THg concentrations control for the effect of fish weight as a covariate in the Ob', Yenisey, Pyasina, Lena, and Kolyma Rivers, as fish weight correlated positively with THg concentration. [b]Rivers which were sampled between 1980 and 2001. All other rivers were sampled between 1988 and 2001. [c]Bold trends indicate those which are statistically significant at $\alpha = 0.05$. [d]Adequacy is defined as the number of sampling years divided by the number of years required to detect an annual 5% change in contaminant concentration.[21] Adequacy values greater than or equal to 1.0 indicate reliable trends.

temperatures on THg accumulation in burbot throughout the pan-Arctic region.

Longitudinal Trends. Concentrations of THg in all eight Russian rivers decreased by an average of 1.8% per 10° longitude from west to east across the study area ($R^2 = 0.61$, $p = 0.022$, Figure 3). This trend is robust as the adequacy of the trend is greater than 1.0. Because THg concentrations were higher in western than in eastern Russia (Figure 3), and because the only three rivers sampled from 1980 to 1987 were also the three westernmost rivers, longitude could have acted as a confounding variable in the cumulative temporal model that included THg data from all eight rivers. To remove the effect of longitude on THg concentrations across the entire Russian Arctic in this cumulative temporal model, data from 1980 to 1987 were removed from this model, leaving only years in which all rivers were sampled. No change in the original cumulative model was observed when these early sampling years were excluded (annual trend = −2.5%, $R^2 = 0.82$, $p < 0.001$), providing support to the declining longitudinal trend from west to east across the Russian Arctic (Figure 3).

A longitudinal gradient in North American and European Arctic THg concentrations similar to the one observed in this study was described in 2011 by Rigét et al. for all Arctic animals for which data exist, including burbot and other freshwater fishes.[4] This longitudinal gradient showed that the quantity and magnitude of increasing THg trends in these Arctic animals decreases moving from west to east across the Arctic, beginning in Alaska and western Canada where Hg trends are increasing at the highest rates, and ending in Scandinavia where Hg trends are mostly stable or even declining.[4] When this longitudinal trend is forecast even further eastward into the Russian Arctic, Rigét et al.'s regression model predicts mostly decreasing Hg trends there. The longitudinal and temporal THg trends presented in this study therefore agree with the longitudinal trend first reported by Rigét et al.,[4] showing that this spatial gradient holds true across the entire pan-Arctic region for which data exist.[4,14,15,19,20]

The longitudinal gradient in THg trends observed here may be influenced in part by a spatial gradient in industrial activities which occurred throughout the Russian Arctic. In general, more industrial activities and higher human population densities occurred in the western Russian Arctic, whereas fewer industrial activities and lower human population densities occurred in the central and eastern Russian Arctic between 1980 and 2001, as is still true today. As previous studies have reported decreasing THg concentrations in fish with increasing distance from point sources of Hg contamination,[37,46] it could be that THg concentrations in burbot declined longitudinally in response to decreasing proximity to industrial activities moving west to east across the Russian Arctic. This hypothesis is also supported by sediment cores collected across the Russian Arctic showing that Hg concentrations in surface sediments decrease longitudinally from west to east[47] in a strikingly similar fashion to THg trends observed here in burbot. Because surface Hg levels are predominantly attributed to deposition of anthropogenic Hg emissions,[47] it is suggested here that the longitudinal trend in THg observed in Russian Arctic burbot may at least in part be influenced by spatial variation in exposure to anthropogenic Hg emissions and effluent across the region. Conversely, the presence of dams and HPPs in the Russian Arctic do not appear to have influenced longitudinal THg trends in burbot between 1988 and 2001. As earlier studies observed increasing Hg concentrations with the presence of dams and HPPs,[24,25] we observed lower THg concentrations in regions with dams and HPPs (the central and eastern Russian Arctic) than in regions without these infrastructures (the western Russian Arctic).

Heterogeneity in THg Trends. These results provide strong support to previous indications that trends in THg concentrations varied throughout the pan-Arctic region preceding and immediately following the turn of the twenty-first century, having mostly increased in the North American Arctic[3,4] and mostly decreased in the Russian Arctic.[20] Although there are no additional environmental data (e.g., aqueous Hg concentrations, water temperatures) to jointly analyze the Hg data reported here, it appears that this spatial and temporal heterogeneity in burbot THg trends may have also been driven in part by varying rates of GEM emissions from sub-Arctic source regions. Available evidence suggests that atmospheric GEM is transported to the North American Arctic predominantly from Hg emissions in sub-Arctic Asia,[48] which increased steadily from 1990 to 2005.[3] Conversely, GEM is transported to the Russian Arctic predominantly from Hg emissions in sub-Arctic North America and Europe,[3,48] which decreased steadily from 1990 to 2005[3] in response to the implementation of environmental regulations.[9,36] Thus, the increasing Hg trends in North American Arctic animals and the decreasing Hg trends in Russian Arctic animals both follow similar patterns as Hg emissions trends from their respective

DOI: 10.1021/acs.est.7b03929
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

source regions. Because of this, and because about 70% of Hg present in interior Arctic tundra ecosystems is derived from the deposition of anthropogenic Hg emissions,[49] it appears likely that deposition of GEM transported from sub-Arctic regions could influence THg trends in animals across the entire pan-Arctic region, as has been observed in other regions of the world.[50,51] However, further study of long-range GEM transport and associated rates of deposition to the Arctic, perhaps utilizing stable isotopic tracers, is necessary to create a more thorough understanding of the relationships between sub-Arctic GEM emissions and Hg trends in animals throughout the pan-Arctic region.

Interpretation of temporal and longitudinal trends in THg in Russian Arctic burbot suggest that both depositional and postdepositional processes may explain the observed spatial and temporal heterogeneity in Hg trends in Arctic animals. In order to further understand Hg dynamics throughout the entire pan-Arctic region, integrative analyses must be conducted based on spatially complete data sets of the emission, transport, and deposition of GEM to the Arctic, as well as on biotic and abiotic environmental controls of Hg bioavailability and bioaccumulation.

AUTHOR INFORMATION

Corresponding Author

*E-mail: palex93@vt.edu.

ORCID

Alexander R. Pelletier: 0000-0001-5032-6912

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

Data collection for this project was funded by internal directives of the Russian State Service of Observation and Control of Environmental Pollution, the Federal Russian Service for Hydrometeorology and Environmental Monitoring, and the South Russian Centre for Preparation and Implementation of International Projects Ltd. (Rostov-on-Don, Russia). Additional funds were provided by the Kajima Foundation (Japan), the Finnish Environment Institute, and the Hydrochemical Institute (Rostov-on-Don, Russia). A.P. acknowledges funding from an internal directive within the Fralin Life Science Institute at Virginia Tech (Blacksburg, VA). R.S. and M.H. acknowledge funding from the U.S. National Science Foundation (NSF-1107774). We thank Drs. Simon Wilson and Anders Bignert for their helpful instruction in using the PIA software, as well as the many people on the Kolyma, Lena, Mezen, North Dvina, Ob', Pechora, Pyasina, and Yenisey Rivers for assisting us in the field and making this work possible. Lastly, we thank three anonymous reviewers for their insightful comments which greatly improved our work.

REFERENCES

(1) AMAP. *AMAP Assessment 2002: Heavy metals in the Arctic*; Arctic Monitoring and Assessment Programme (AMAP): Oslo, Norway, 2005.

(2) Evans, M. S.; Muir, D.; Lockhart, W. L.; Stern, G.; Ryan, M.; Roach, P. Persistent organic pollutants and metals in the freshwater biota of the Canadian Subarctic and Arctic: An overview. *Sci. Total Environ.* **2005**, *351*−*352*, 94−147.

(3) AMAP. *AMAP Assessment 2011: Mercury in the Arctic*; Arctic Monitoring and Assessment Programme (AMAP): Oslo, Norway, 2011.

(4) Rigét, F.; Birgit, B.; Bignert, A.; Wilson, S.; Aars, J.; Born, E.; Dam, M.; Dietz, R.; Evans, M.; Gamberg, M.; Gantner, N.; Green, N.; Gunnlaugsdóttir, H.; Kannan, K.; Letcher, R.; Muir, D.; Roach, P.; Sonne, C.; Stern, G.; Wiig, Ø. Temporal trends of Hg in Arctic biota, an update. *Sci. Total Environ.* **2011**, *409*, 3520−3526.

(5) Committee on the Toxicological Effects of Methylmercury, Board on Environmental Studies and Toxicology, Commission on Life Sciences, Division on Earth and Life Studies, National Research Council. *Toxicological Effects of Methylmercury*; National Academy Press: Washington, D.C., 2000.

(6) Dietz, R.; Sonne, C.; Basu, N.; Braune, B.; O'Hara, T.; Letcher, R. J.; Scheuhammer, T.; Andersen, M.; Andreasen, C.; Andriashek, D.; Asmund, G.; Aubail, A.; Baagøe, H.; Born, E. W.; Chan, H. M.; Derocher, A. E.; Grandjean, P.; Knott, K.; Kirkegaard, M.; Krey, A.; Lunn, N.; Messier, F.; Obbard, M.; Olsen, M. T.; Ostertag, S.; Peacock, E.; Renzoni, A.; Rigét, F. F.; Skaare, J. U.; Stern, G.; Stirling, I.; Taylor, M.; Wiig, Ø.; Wilson, S.; Aars, J. What are the toxicological effects of mercury in Arctic biota? *Sci. Total Environ.* **2013**, *443*, 775−790.

(7) Carrie, J.; Wang, F.; Sanei, H.; Macdonald, R. W.; Outridge, P. M.; Stern, G. A. Increasing contaminant burdens in an Arctic fish, Burbot (*Lota lota*), in a warming climate. *Environ. Sci. Technol.* **2010**, *44*, 316−322.

(8) Evans, M.; Muir, D.; Brua, R. B.; Keating, J.; Wang, X. Mercury trends in predatory fish in Great Slave Lake: The influence of temperature and other climate drivers. *Environ. Sci. Technol.* **2013**, *47* (22), 12793−12801.

(9) Faïn, X.; Ferrari, C. P.; Dommergue, A.; Albert, M. R.; Battle, M.; Severinghaus, J.; Arnaud, L.; Barnola, J.; Cairns, W.; Barbante, C. Polar firn air reveals large-scale impact of anthropogenic mercury emissions during the 1970s. *Proc. Natl. Acad. Sci. U. S. A.* **2009**, *106*, 16114−16119.

(10) Li, C. S.; Cornett, J.; Willie, S.; Lam, J. Mercury in Arctic air: The long-term trend. *Sci. Total Environ.* **2009**, *407*, 2756−2759.

(11) Monson, B. A. Trend reversal of mercury concentrations in piscivorous fish from Minnesota lakes − 1982−2006. *Environ. Sci. Technol.* **2009**, *43* (6), 1750−1755.

(12) Schmeltz, D.; Evers, D. C.; Driscoll, C. T.; Artz, R.; Cohen, M.; Gay, D.; Haeuber, R.; Krabbenhoft, D. P.; Mason, R.; Morris, K.; Wiener, J. G. MercNet - A national monitoring network to assess responses to changing mercury emissions in the United States. *Ecotoxicology* **2011**, *20* (7), 1713−1725.

(13) Evans, M. S.; Lockhart, W. L.; Doetzel, L.; Low, G.; Muir, D.; Kidd, K.; Stephens, G.; Delaronde, J. Elevated mercury concentrations in fish in lakes in the Mackenzie River Basin: The role of physical, chemical, and biological factors. *Sci. Total Environ.* **2005**, *351*, 479−500.

(14) Åkerblom, S.; Bignert, A.; Meili, M.; Sonesten, L.; Sundbom, M. Half a century of changing mercury levels in Swedish freshwater fish. *Ambio* **2014**, *43*, 91−103.

(15) Braune, B. M.; Gaston, A. J.; Mallory, M. L. Temporal trends of mercury in eggs of five sympatrically breeding seabird species in the Canadian Arctic. *Environ. Pollut.* **2016**, *214*, 124−131.

(16) Pućko, M.; Burt, A.; Walkusz, W.; Wang, F.; Macdonald, R. W.; Rysgaard, S.; Barber, D. G.; Tremblay, J. É.; Stern, G. A. Transformation of mercury at the bottom of the Arctic food web: An overlooked puzzle in the mercury exposure narrative. *Environ. Sci. Technol.* **2014**, *48* (13), 7280−7288.

(17) French, T. D.; Houben, A. J.; Desforges, J. P. W.; Kimpe, L. E.; Kokelj, S. V.; Poulain, A. J.; Smol, J. P.; Wang, X.; Blais, J. M. Dissolved organic carbon thresholds affect mercury bioaccumulation in Arctic lakes. *Environ. Sci. Technol.* **2014**, *48*, 3162−3168.

(18) Holmes, R. M.; Coe, M. T.; Fiske, G. J.; Gurtovaya, T.; McClelland, J. W.; Shiklomanov, A. I.; Spencer, R. G. M..; Tank, S. E.; Zhulidov, A. V. Climate change impacts on the hydrology and biogeochemistry of Arctic rivers. In *Climate Change and Global Warming of Inland Waters: Impacts and Mitigation for Ecosystems and Societies*; Goldman, C. R., Kumagai, M., Robarts, R. D., Eds.; Wiley, 2013.

DOI: 10.1021/acs.est.7b03929
Environ. Sci. Technol. XXXX, XXX, XXX−XXX

(19) Åkerblom, S.; Nilsson, M.; Yu, J.; Ranneby, B.; Johansson, K. Temporal change estimation of mercury concentrations in northern pike (*Esox lucius* L.) in Swedish lakes. *Chemosphere* **2012**, *86* (5), 439−445.
(20) Castello, L.; Zhulidov, A. V.; Gurtovaya, T. Yu.; Robarts, R. D.; Holmes, R. M.; Zhulidov, D. A.; Lysenko, V. S.; Spencer, R. G. M. Low and declining mercury in Arctic Russian rivers. *Environ. Sci. Technol.* **2013**, *48* (1), 747−752.
(21) Bignert, A.; Rigét, F.; Braune, B.; Outridge, P.; Wilson, S. Recent temporal trend monitoring of mercury in Arctic biota - how powerful are the existing data sets? *J. Environ. Monit.* **2004**, *6*, 351−355.
(22) McPhail, J. D.; Paragamian, V. L. *Burbot biology and life history. In BURBOT: Biology, Ecology, and Management*; Paragamian, V. L., Willis, D. W., Eds.; Fisheries Management Section of the American Fisheries Society, *128* (9), 2008.
(23) Paragamian, V. L.; Wakkinen, V. D. Seasonal movement of burbot in relation to temperature and discharge in the Kootenai River, Idaho, USA and British Columbia. *Canada. Am. Fish. Soc. Symp.* **2008**, *59*, 55−77.
(24) Verdon, R.; Brouard, D.; Demers, C.; Lalumiere, R.; Laperle, M.; Schetagne, R. Mercury Evolution (1978−1988) in fishes of the La Grande Hydroelectric Complex, Quebec, Canada. *Water, Air, Soil Pollut.* **1991**, *56*, 405−417.
(25) Porvari, P. Development of fish mercury concentrations in Finnish reservoirs from 1979 to 1994. *Sci. Total Environ.* **1998**, *213*, 279−290.
(26) Amundsen, P.; Staldvik, F. J.; Lukin, A. A.; Kashulin, N. A.; Popova, O. A.; Reshetnikov, Y. S. Heavy metal contamination in freshwater fish from the border region between Norway and Russia. *Sci. Total Environ.* **1997**, *201*, 211−224.
(27) Nikanorov, A. M.; Zhulidov, A. V.; Pokarzhevskij, A. D. *Biomonitoring of Heavy Metals in Freshwater Ecosystems*; Hydrometeoizdat: Leningrad, 1985; (Published in Russian).
(28) Nikanorov, A. M.; Zhulidov, A. V. *Biomonitoring of Metals in Freshwater Ecosystems*; Hydrometeoizdat: Leningrad, 1991; (Published in Russian). ISBN 5−286−00314−1.
(29) Hendzel, M. R.; Jamieson, D. M. Determination of mercury in fish. *Anal. Chem.* **1976**, *48*, 926−928.
(30) Trebacz, E.; Boila, G.; Wagemann, R. *Analytical Methods Manual: Metals in Tissues; Department of Fisheries and Oceans*; Central and Arctic, Freshwater Institute: Winnipeg, Manitoba, Canada, 1998; Vol. 7.
(31) NLET: *National Laboratory for Environmental Testing. Schedule of Services*; National Water Research Institute, Environment Canada: Ontario, Canada, 2003.
(32) Lockhart, W. L.; Stern, G. A.; Low, G.; Hendzel, M.; Boila, G.; Roach, P.; Evans, M. S.; Billeck, B. N.; DeLaronde, J.; Friesen, S.; Kidd, K.; Atkins, S.; Muir, D. C. G.; Stoddart, M.; Stephens, G.; Stephenson, S.; Harbicht, S.; Snowshoe, N.; Grey, B.; Thompson, S.; DeGraff, N. A history of total mercury in edible muscle of fish from lakes in northern Canada. *Sci. Total Environ.* **2005**, *351*, 427−463.
(33) Bloom, N. S. On the chemical form of mercury in edible fish and marine invertebrate tissue. *Can. J. Fish. Aquat. Sci.* **1992**, *49*, 1010−1017.
(34) Bignert, A. PIA statistical application developed for use by the Arctic Monitoring and Assessment Programme (available from www.amap.no). 2007.
(35) Kvintradze, E. *Russia's Output Collapse and Recovery: Evidence from the Post-Soviet Transition*; Working Paper 10/89 of the International Monetary Fund. 2010.
(36) Zhulidov, A. V.; Emetz, V. M. *Biogeochemical regionalization of the USSRManuscript*; Hydrochemical Institute, Rostov-on-Don, Russian Federation, 1991, 625 p.
(37) Kinghorn, A.; Solomon, P.; Chan, H. M. Temporal and spatial trends of mercury in fish collected in the English-Wabigoon river system in Ontario, Canada. *Sci. Total Environ.* **2007**, *372*, 615−623.
(38) Pääkkönen, J. P. J.; Marjomäki, T. J. Feeding of Burbot, *Lota lota*, at different temperatures. *Environ. Biol. Fishes* **2000**, *58* (1), 109−112.
(39) Hofmann, N.; Fischer, P. Impact of temperature on food intake and growth in juvenile burbot. *J. Fish Biol.* **2003**, *63* (5), 1295−1305.
(40) Yang, D.; Liu, B.; Ye, B. Stream temperature changes over Lena river basin in Siberia. *Geophys. Res. Lett.* **2005**, *32* (5), 1−5.
(41) Lammers, R. B.; Pundsack, J. W.; Shiklomanov, A. I. Variability in river temperature, discharge, and energy flux from the Russian pan-Arctic landmass. *J. Geophys. Res.* **2007**, *112* (G4), 15.
(42) Pickhardt, P. C.; Folt, C. L.; Chen, C. Y.; Klaue, B.; Blum, J. D. Algal blooms reduce the uptake of toxic methylmercury in freshwater food webs. *Proc. Natl. Acad. Sci. U. S. A.* **2002**, *99*, 4419−4423.
(43) Hill, W. R.; Larsen, I. L. Growth dilution of metals in microalgal biofilms. *Environ. Sci. Technol.* **2005**, *39*, 1513−1518.
(44) Karimi, R.; Chen, C. Y.; Pickhardt, P. C.; Fisher, N. S.; Folt, C. L. Stoichiometric controls of mercury dilution by growth. *Proc. Natl. Acad. Sci. U. S. A.* **2007**, *104*, 7477−7482.
(45) Rypel, A. L. Meta-analysis of growth rates for a circumpolar fish, the Northern Pike (*Esox lucius*), with emphasis on effects of continent, climate, and latitude. *Ecol. Freshwater Fish* **2012**, *21*, 521−532.
(46) Fimreite, N.; Reynolds, M. R. Mercury contamination of fish in northwestern Ontario. *J. Wildl. Manage.* **1973**, *37*, 62−68.
(47) Rognerud, S.; Skotvold, T.; Fjeld, E.; Norton, S. A.; Hobaek, A. Concentrations of trace elements in recent and preindustrial sediments from Norwegian and Russian Arctic lakes. *Can. J. Fish. Aquat. Sci.* **1998**, *55* (6), 1512−1523.
(48) Durnford, D.; Dastoor, A.; Figueras-Nieto, D.; Ryjkov, A. Long range transport of mercury to the Arctic and across Canada. *Atmos. Chem. Phys.* **2010**, *10*, 6063−6086.
(49) Obrist, D.; Agnan, Y.; Jiskra, M.; Olson, C. L.; Colegrove, D. P.; Hueber, J.; Moore, C. W.; Sonke, J. E.; Helmig, D. Tundra uptake of atmospheric elemental mercury drives Arctic mercury pollution. *Nature* **2017**, *547* (7662), 201−204.
(50) Harris, R. C.; Rudd, J. W. M.; Amyot, M.; Babiarz, C. L.; Beaty, K. G.; Blanchfield, P. J.; Bodaly, R. A.; Branfireun, B. A.; Gilmour, C. C.; Graydon, J. A.; Heyes, A.; Hintelmann, H.; Hurley, J. P.; Kelly, C. A.; Krabbenhoft, D. P.; Lindberg, L. E.; Mason, R. P.; Paterson, M. J.; Podemski, C. L.; Robinson, A.; Sandilands, K. A.; Southworth, G. R.; St.Louis, V. L.; Tate, M. T. Whole-ecosystem study shows rapid fish-mercury response to changes in mercury deposition. *Proc. Natl. Acad. Sci. U. S. A.* **2007**, *104* (42), 16586−16591.
(51) Hutcheson, M. S.; Smith, C. M.; Wallace, G. T.; Rose, J.; Eddy, B.; Sullivan, J.; Pancorbo, O.; West, C. R. Freshwater fish mercury concentrations in a regionally high mercury deposition area. *Water, Air, Soil Pollut.* **2008**, *191* (1−4), 15−31.

DOI: 10.1021/acs.est.7b03929
Environ. Sci. Technol. XXXX, XXX, XXX−XXX