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# Mercury in the Barents region – River fluxes, sources, and environmental concentrations $\stackrel{\star}{\succ}$

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# ABSTRACT

Arctic rivers are receiving increased attention for their contributing of mercury (Hg) to the Arctic Ocean. Despite this, the knowledge on both the terrestrial release sources and the levels of Hg in the rivers are limited. Within the Arctic, the Barents region has a high industrial development, including multiple potential Hg release sources. This study presents the first overview of potential Hg release sources on Norwegian and Russian mainland draining to the Barents Sea. Source categories cover mining and metallurgy industry; historical pulp and paper production; municipal and industrial solid waste handling; fossil fuel combustion; and past military activities. Available data on Hg in freshwater bodies near the identified potential release sources are reviewed. Levels of Hg were occasionally exceeding the national pollution control limits, thereby posing concern to the local human population and wildlife. However, the studies were sparse and often unsystematic. Finally, we present new data of Hg measured in five Barents rivers. These data reveal strong seasonality in the Hg levels, with a total annual flux constituting 2% of the panarctic total. With this new insight we aspire to contribute to the international efforts of reducing Hg pollution, such as through the effective implementation of the Minamata Convention. Future studies documenting Hg in exposed Barents freshwater bodies are warranted.

# 1. Introduction

Mercury (Hg) continues to pose a concern for humans and wildlife in the Arctic due to its neurotoxic properties (as reviewed e.g., by Kirk et al., 2012). Decades of long-range Hg transport to the Arctic has resulted in elevated levels in fish, marine mammals (Dietz et al., 2022), and in the local human population (Basu et al., 2022). Despite international efforts to reduce global Hg pollution, such as through the Minamata Convention on Mercury<sup>1</sup> (hereafter the Minamata Convention), a continuing 65 Mg Hg is estimated to be annually deposited onto the Arctic Ocean (Dastoor et al., 2022). Panarctic rivers have received increased attention for their annual contribution of an additional 41 Mg Hg to the Arctic Ocean (Dastoor et al., 2022). Despite this, few measurements of Hg in panarctic rivers exist (Sonke et al., 2018).

Source categories of Hg releases to rivers comprise diffuse natural pools, that are increased by the legacy of long-range transport, as well as point sources from local industrial and urban activities. While the latter is not usually associated with the Arctic, the Barents region stands out with a higher socio-economic development. Richness in natural resources and a milder Arctic climate have since the 1920s laid the foundation for industries and subsequent urbanisation (Lundgren, 2006). Levels of Hg in surface waters in the region are thus also crucial

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<sup>&</sup>lt;sup>1</sup> The Minamata Convention on Mercury is a global treaty to protect human health and the environment from the adverse effects of Hg.

to the health of local human population and wildlife relying on local water sources for drinking, fishing, and recreational activities. Potential major industrial release sources cover mining and metallurgy in addition to historic production of bleached pulp and paper. With Hg being an impurity of most mineral ores, releases can occur at every step of mining and metal processing (AMAP/UN Environment, 2019; Pirrone et al., 2010). Moreover, large quantities of Hg were until the late 1990s used to produce bleached pulp and paper (chlor-alkali produced by the Hg-cell method) (Crook & Mousavi, 2016), and potentially released to the environment e.g. with the inadequately treated waste water. Additional relevant source categories cover municipal and industrial solid waste handling, burning of fossil fuels, and historical military activities (AMAP/UN Environment, 2019; Pirrone et al., 2010).

Hg is typically released to land and water in its inorganic forms. It is not very water soluble but increases its partitioning to the water phase by forming association with particles or dissolved organic matter (DOM) (O'Connor et al., 2019). Hg bound to particles is anticipated to constitute about half of the total panarctic river transport (Dastoor et al., 2022). Hg bound to particles in soils are mobilized and transported to the water by erosion, while Hg in river sediments may be re-suspend to the water phase (Morel et al., 1998). Release of Hg to the river water occurs during hydrological events, with the greatest export of Hg by the panarctic rivers occurring during the high flow spring snow melt season (Dastoor et al., 2022). The water compartment is important for the transformation of inorganic Hg to the neurotoxic and bioavailable methyl-Hg. This occurs in waterlogged microenvironments with slightly anoxic conditions (Paranjape & Hall, 2017). Based on future climate change scenarios, the downscaled predictions for this region are elevated temperatures and intensification of hydrological events. This will lead to both increased riverine transport of Hg and Hg methylation rates (Chételat et al., 2022).

Both countries bordering the Barents Sea, Norway and Russia, have signed the Minamata Convention, but to date only Norway has ratified the treaty (Minamata Convention on Mercury, 2022b). The Minamata Convention works to reduce global Hg pollution, focusing on e.g., supply sources and trade, industrial processes using Hg, as well as proper treatment of Hg-containing waste and contaminated sites (Lennett & Gutierrez, 2018). More focus has been devoted to reducing sources of emission to air compared to sources of release to land and water. For example, while guidance documents on emission to air, i.e., Best Available Technique (BAT)/Best Environmental Practice (BEP), have been available since before the convention entered into force in 2013, corresponding guidelines are still not available for release sources (UNEP and Minamata Convention, 2021).

Focusing on the Barents region, we present a scoping literature review combining qualitative information on Hg release sources with quantitative data on Hg in lakes and rivers. The pollution status of the region is evaluated with respect to the identified sites of potential Hg release. Strengthening the data foundation, we present new seasonal Hg data from five Barents rivers along with estimated annual Hg fluxes. With this study, we aspire to contribute towards the development of BATs/BETs for Hg release sources under the Minamata Convention, and to highlight the role of the Barents region in the assessment of riverine transport of Hg to the Arctic Ocean.



**Fig. 1.** Map illustrating the study region shaded with different colours for the Norwegian county Troms and Finnmark and the Russian Murmansk Oblast, Republic of Karelia, and Arkhangelsk Oblast. Locations of the Identified potential point sources of Hg are indicated (fossil fuel combustion, past military activities, mining and metallurgical industry, historic pulp and paper production, and waste handling sites) along with lakes (light blue droplet) and rivers (dark blue droplet) with available Hg measurements. Monitored rivers are identified by yellow stars. Note that River Niva is not indicated due to the few samples collected. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

#### 2. Materials and methods

## 2.1. Study region

The study region covers mainland of Norway and Russia draining to the Barents Sea. In Norway this is comprised by Troms and Finnmark county, and in Russia by Murmansk Oblast, the Republic of Karelia, and Arkhangelsk Oblast, as outlined in Fig. 1 (excluding Nenetsia). The region, comprised by the arctic, subarctic, and boreal zones, is known for its harsh climate conditions. The natural environment in these areas is diverse, with flat plains and tundra, dense taiga forests, towering mountain ridges, and limited agriculture in the southern regions. However, these regions are abundant in lakes and bogs, which contribute to their overall ecological richness. The region is relatively sparsely populated with towns/cities often located near some historical or ongoing industrial activity. For more details on the region see Table S1.

# 2.2. Study rivers

Six rivers were selected for seasonal monitoring of Hg based on the following criteria: *i*) drainage to the Barents- or the White Sea; *ii*) larger size representative to the region; *iii*) vicinity to potential Hg release sources or representative for background levels; and *iv*) accessibility for sampling. The water quality in the four large rivers, Målselv, Alta, Tana, and Pasvik, are measured monthly by the Norwegian monitoring programme (Braaten et al., 2020; kaste et al., 2021). River Pasvik has been documented to be influenced by nearby metallurgical complexes in Nikel. The two Russian Rivers, Tuloma and Northern Dvina, are large and in proximity to potential point pollution sources (Fig. 1). The pollution status of the Northern Dvina River has been documented, but little data exists for the Tuloma River. Note that River Niva in Murmansk Oblast was originally included, but continued sampling was not possible to due to the Covid-19 pandemic. For details of the rivers see Table 2.

#### 2.3. Compilation of potential Hg pollution sources and measured Hg data

Information on potential Hg pollution sources was collated from numerous sources, including peer-reviewed literature, reports, existing databases, e.g., the Barents Euro-Arctic Cooperation (BEAC), Working Group on Environment (WGE), Subgroup on Hot Spots Exclusion (SHE), ongoing monitoring programmes, and expert opinions from researchers, governmental employees, and local stakeholders in Norway and Russia. The compilation was an iterative process. First, categories of activities with potential Hg release relevant to the region were identified. Subsequently, the region was carefully scanned to identify the sites. Google maps in the satellite mode was used to identify water bodies located nearby the sites. Data of Hg concentrations was confined to studies of lakes and rivers, and to the environmental matrices of water, sediment, and fish muscle. Data sources comprised peer-reviewed scientific articles, publicly available reports, and new data generated by this study. The research publication database of Google Scholar was used with the keyword mercury in combination with local names of regions/cities/ water bodies and/or categories of pollution sources. The data were compiled and presented by averages and ranges (where available). Data of Hg concentration in fish provided as dry weight (d.w.) was converted to wet weight (w.w.) by dividing by a factor of 4.5. In fish muscle, data from the same species and site was averaged, weighted by the number of observations. In a few studies where the number of observations was not available it was set to one for the calculations. In sediments, only data from surface layers were included which was most often defined by 0-5 cm depth. To link the identified potential Hg release sites to the lakes and rivers, a radius of 40 km was used, following the findings by Moiseenko et al. (2018).

Finally, 106 publications were included in this review, of which 32 publications provided data on Hg in water/sediment/fish muscle from

32 different sites.

#### 2.4. Sampling of river water

Norwegian rivers (Målselv, Alta, Tana, and Pasvik) were sampled monthly during 2019 and 2020. For details see Braaten et al. (2020). Monthly samples were in 2020 also collected from the Russian river Northern Dvina. Samples from the Tuloma river were only acquired for four of the originally planned 12 months of 2019 due to Covid-19 travel restrictions. For the same reasons, the Russian River Niva (UTM 36/WGS 84, 474974, 7446178) was only sampled once in 2019 and is thus excluded from most of the following discussions. Only the Northern Dvina was sampled in December due to challenging winter conditions at the other sites. For details on months of sampling see Table S3.

The water samples were collected in new 250 mL fluoropolymer bottles, following ultra-clean sampling procedures to avoid contamination (USEPA, 1996). Sample bottles were kept in double plastic bags and as soon as possible shipped to the NIVA laboratory in Oslo, Norway.

# 2.5. Determination of total Hg

Determination of total Hg in river water followed the USEPA Method 1631 by oxidation, purge and trap, and cold vapor atomic fluorescence spectrometry (CVAFS) (USEPA, 2002) using an automated Brooks Rand Inc. system. The method limit of quantification (LOQ) was  $0.0003 \,\mu g \, L^{-1}$  (10 x standard deviations of method blanks). For details, see Braaten et al. (2014). For every batch of Hg analysis (n = 10–50 individual samples), quality assurance and quality control (QA/QC) measures included method blanks (n = 3), blank spikes (n = 3), drift control, sample duplicates, and matrix spikes. The relative difference of sample duplicates was <15% and recovery of drift control, blank spikes and matrix spikes was 80–120%. If QA/QC measures were not met, samples were re-analysed.

#### 2.6. River water flow data

For the Norwegian rivers (Målselv, Alta, Tana, and Pasvik), data on water flow was retrieved through the Norwegian River Monitoring Programme (Braaten et al., 2020; Kaste et al., 2021). Data were summarised to monthly values. For Northern Dvina River in Russia (Usy-Pinega mouth station) 2020 data was acquired from the Automated Information System of State Monitoring of Water Bodies (AIS GMVO, 2022) and from the Arctic great rivers observatory (Shiklomanov et al., 2021). For River Tuloma only historic data was available, from which monthly data were averaged from years 1956–1970.

# 2.7. River Hg load calculation

Annual riverine Hg loads were estimated according to the global mean method (Equation (1)) by finding the product of annual flow and average concentration. The method was suggested by Elwan et al. (2018) to be best suited for the monthly frequency of data for the sum of water flow and levels of Hg, assessed in this study.

$$L = mQ_t \left(\sum_{i=1}^n \frac{C_i}{n}\right)$$
 (Equation 1)

where L is annual river load (M/T), m is a conversion factor to convert the calculated values into a specific unit,  $C_i$  is the contaminant concentration (M/L<sup>3</sup>) measured at the ith day,  $Q_t$  is the annual flow (L<sup>3</sup>/T), and n is the number of samples.

# 3. Results and discussion

#### 3.1. Anthropogenic potential sources of Hg

Reviewed potential point sources of Hg are here confined to those located within the study region (Fig. 1). Focus is on releases to surface waters and soils while also including sources of emission to air where local deposition can be expected (i.e., high concurrent particle load). Both ongoing and historic activities are covered. This categorises to 1) mining and metallurgical industry; 2) pulp and paper production (chloralkali production); 3) solid waste handling; 4) burning of oil, gas, and coal; and 5) past military activities.

# 3.1.1. Mineral mining and metallurgical industry

Mining in the Barents region is dominated by nickel (Ni), copper (Cu), cobalt (Co), iron (Fe), and apatite  $(Ca_5(PO_4)_3(OH, F, Cl))$  extraction. The metal ores differ in their content of Hg impurity, depending on the local natural geological formation. The highest content is typically in sulphur (S) ores since S has a strong affinity for Hg (Kerfoot et al., 2004; Romanov et al., 2014). During every stage of mining and metallurgical processing, there is a risk of releasing Hg to the environment (AMAP/UN Environment, 2019; Pirrone et al., 2010). For example, leaching of Hg to nearby waterways has been documented from mine tailings with gangue material from extraction (Wang et al., 2012). Moreover, the high temperatures applied during metal smelting causes elementary Hg to evaporate. Local deposition of the Hg is expected due to the typical high particle content of smelter flue gas. Elevated levels of Hg from these emissions have been detected up to 100 km away from metal smelters in the Kola peninsula (Moiseenko et al., 2018).

3.1.1.1. Copper and nickel. Great reserves of copper (Cu) and nickel (Ni) containing S-deposits forms the basis for large-scale mining operations in Murmansk Oblast. Typically, the Hg content averages at 1 and 0.09 g  $ton^{-1}$  for Cu for Ni ores rich in S, respectively (Romanov et al., 2014). In the north-west, close to the Norwegian border, there were both a large metal smelter (Nikel) as well as a concentrator and briquetting shop (Zapolyarny). The treated mineral ores are of local origin as well as imported. Sources of import cover central Siberia, which is also associated with a very high content of S (Dauvalter, 1994). Due to limited pollution control the emissions of acid oxides and heavy metals from the processing of these S rich Cu and Ni ores has led to extensive environmental damage, such as vegetation death from acid deposition (Gashkina & Moiseenko, 2016) and heavy metal contamination. The nearby River Pasvik is particularly vulnerable due to its proximity to the metal smelter (Amundsen et al., 1997). Further south in Murmansk Oblast, in the town of Monchegorsk, lies one of Europe's largest metallurgical complexes. The town is on the shore of Lake Imandra (Monche Bay, Bolshaya Imandra) which is of environmental concern for receiving waste effluents from the site. Currently, the activities at both two sites in Nikel and Monchegorsk have been stopped. While at Nikel, the smelter has been decommissioned, activities are expected to be reorganised and resumed at Monchegorsk following upgrade of the technical equipment. The upgrade will provide increased capacity, which may also bring with improved pollution control.

In Norway, an active Cu mine is located at the coast in Kåfjord, and two discontinued mines are at the coast in Kvalsund and in the city of Kautokeino, respectively. The latter is located in the upper part of the River Alta.

3.1.1.2. Apatite. Apatite is a group of phosphate (PO<sub>4</sub>) minerals primarily used for the manufacture of mineral fertilizers. In the PO<sub>4</sub> bearing rock on the Kola peninsula, Hg is found averaging 0.01 g ton<sup>-1</sup> (Reta et al., 2018). Often found in association with apatite is nepheline which is a mineral used in e.g., aluminium- or ceramic industry.

One of the world's largest apatite-nepheline ore deposit is located

south of Murmansk Oblast, close to the Khibiny mountain nature reserve (Gerasimova et al., 2018; Lukichev et al., 2017). At each side of the mountain ridge are two different mining operations, facing towards lake Imandra and lake Umbozero, respectively. Various environmental concerns have previously been raised (Didyk et al., 2018). Now, the mining facilities at both sites are under reconstruction, which is expected to improve their environmental standards (AMAP/NEFCO, 2020; Dauvalter et al., 2009).

3.1.1.3. Iron. Iron (Fe) ores from Siberia contain on average 0.06 g ton<sup>-1</sup> Hg (Romanov et al., 2014). Generally, this Hg is bonded to either S or to the Fe oxides (Rytuba, 2003). There are large active iron mining and processing plants in the Murmansk Oblast and in the Republic of Karelia. In Norway there are only several historical Fe mining sites.

In Murmansk, two mining and processing plants are on the shores of Lake Imandra; one south west in the town of Kovdor and one north in the town of Olengorsk. Currently, the site in Kovdor is subject to organisational and technical measures to reduce water use and wastewater discharge (AMAP/NEFCO, 2020). In Karelia, the Kostomuksha iron mine and ore dressing mill represents the largest of its kind in the country (Jukka et al., 2010). Emissions to the air and of wastewater releases from the mine and mill have had documented negative impact on their nearby environment (Tkatcheva et al., 2004). Of major concern regarding Hg contamination to the environment is the dumping of waste effluents and quarry into the local water bodies (Lake Kostamusjärvi and the Kentti water system) (Jukka et al., 2010; Tkatcheva et al., 2004).

In the study region on the Norwegian side there is a halted Fe mine is in the town of Bjørnevatn, situated on the shores of River Pasvik. Extracted material was processed at a plant in the neighbouring coastal city of Kirkenes. While these activities have been paused since 2015, due to economic reasons, the mine is about to resume its activities.

3.1.1.4. Other minerals. Other large mining and metallurgical operations in the study region include aluminium (Al), various rare earth elements, diamond, and graphite. Although several of these sites are associated with environmental concern (Bronder et al., 2014; Bumbulyak et al., 2013), Hg contamination is not typically documented. However, when extracting Al from bauxite, there is a possibility of Hg contamination because bauxite usually contains 0.02–1.5 g ton<sup>-1</sup> Hg (Amano, 2020). There is a large reserve of bauxite in Arkhangelsk Oblast, which is mined in the town of Severoonezhsk, near River Onega. Diamond mining produces large volume of tailings (Rytuba, 2003) which has at other sites been found to contain Hg (Bridge, 2016). There is no known ongoing or previous mining of Hg in the region, nor any active Hg-based artisanal gold mining (Treger et al., 2004). An abandoned gold mine has been identified near the city of Kautokeino in Troms and Finnmark county, Norway.

#### 3.1.2. Pulp and paper production (by the chlor-alkali production)

Pulp and paper plants that produce bleached products are linked to potential large historical releases of Hg (Treger et al., 2004). Until the 1990s, the so-called "Hg cell process" (i.e., the Castner-Kellner process) was used to produce bleach (Baldi & Bargagli, 1984; Maserti & Ferrara, 1991; Treger et al., 2004), using tonnes of elemental Hg. While Hg was intended for reuse in the process (as a cathode), large losses were inevitable through wastewater, ventilation, mechanical losses (spills), and end product (Treger et al., 2004). According to estimates from various facilities, during the process, around 1-4% of the Hg used was lost to air, while about 40-45% was disposed of with wastewater and brine. Additionally, up to 50% was directly deposited onto the facility capital structures and ground (Mihaiescu et al., 2012; Treger et al., 2004). Therefore, the decommissioning of obsolete Hg-based pulp and paper production facilities represent a high pollutant risk unless the large quantities of Hg present in the facilities is treated responsibly (Mihaiescu et al., 2012). Surrounding such facilities, tonnes of Hg is also

likely to reside in the soils, slagheaps, and water bodies (Treger et al., 2004).

Several pulp and paper mills that previously used the Hg-cell process are situated within the study region. The north shore of the great Lake Onega, located in the Republic of Karelia, is home to one of the world's primary newsprint suppliers. Severe pollution of the lake sediments is documented by Tomilina and Grebenyuk (2008), resulting from the mill discharging waste water for 80 years (allegedly for 40 years without treatment) (Podsechin et al., 2009). In Arkhangelsk Oblast, three paper mills are situated along the shores of River Northern Dvina (Ovsepyan and Alina, 2019). Furthest upstream is a mill by the tributary river, Vytsjegda. The second and third mills are located closer to the Northern River outlet: One in the city of Arkhangelsk (which was closed in 2013) (AMAP/NEFCO, 2020), and the other in the city of Novodvinsk.

# 3.1.3. Municipal and industrial solid waste

Levels of Hg in municipal solid waste averages at 2 mg kg<sup>-1</sup> (van Velzen et al., 2002) due to the vast range of Hg-containing consumer products (*e.g.*, lightbulbs, electronics, batteries, automotive parts, thermometers, jewellery, etc.). Municipal solid waste is thus a potential source of Hg pollution unless treated properly. Relevant for the study region is also deposition of end-of-life Hg-containing products used in large scale at industrial sites, such as Hg-containing lamps used to illuminate mines, dressing works, and other industrial facilities (Dauvalter & Kashulin, 2018).

Waste in the study region generally ends up either in landfills (regulated or nonregulated) or is combusted at incineration plants. At non-regulated landfills, Hg can leach to the nearby environment. Moreover, anoxic conditions can develop at the landfill to facilitate bacterial conversion of inorganic Hg to the neurotoxic methyl-Hg. During waste incineration, practically 100% of Hg present in the waste is emitted with the flue gas. The high particle content of the flue gas will likely govern a high local deposition of the emitted Hg (Arctic Council, 2008; van Velzen et al., 2002).

Norway and Russia differ in their waste treatment systems. In Norway, a national waste treatment law came in place in the 1980s (KLD, 1983), that reduced the number of illegal dumpsites (Braaten et al., 2022). In Troms and Finnmark county, there is one regulated dumpsite near the River Alta, treating waste from the two cities of Alta and Hammerfest. Several smaller scale legacy dumpsites are nevertheless expected to exist (Kolarctic, 2021). Recycling stations for Hg-containing products have been established throughout the country, though only a fraction of the Hg-containing waste is handed in by the consumers. Levels of Hg in emissions from incineration plants in Norway are subject to the same strict regulations as for the specialised Hg recycling facilities, in accordance with current BAT (Klif, 2010), and should therefore not pose a risk of Hg pollution.

In Russia, work is underway to create a national waste treatment system (Albrecht et al., 2020; Nikitina, 2021). Currently the administrative regions are as independent as different nations in regard to dealing with this matter (Federal Law No. 89 of 30 June 1998). In the Russian Barents region, most of the municipal waste is disposed at dumpsites or landfills. There are more than 200 registered landfills in the Republic of Karelia, of which about 33% are illegitimate (AMAP/-NEFCO, 2020; Bumbulyak et al., 2013). Similar conditions prevail in Arkhangelsk Oblast. A large new landfill in Arkhangelsk, near the rivers Vytsjegda and Northern Dvina, is under establishment. This landfill will annually store 500 000 tonnes waste from the city of Moscow. The plans have been subject to protests from local inhabitants and environmental groups (Bellona, 2020). In Murmansk Oblast, there is an incineration plant is in the city of Murmansk that burns approximately 22% of the waste from the oblast (Filimonova, 2020). In Murmansk Oblast there are also two specialised facilities for treating Hg-containing solid waste. In the past, these sites were considered problematic because they used obsolete equipment, which led to the emission of Hg, instead of mitigating the problem (Bumbulyak et al., 2013). However, it is claimed that the equipment has now been upgraded (AMAP/NEFCO, 2020). Nevertheless, there is no available information to confirm if any corrective measures have been implemented.

#### 3.1.4. Fossil fuels

Burning of fossil fuels is a common source for heat and power generation in the Russian Barents region, both for industries (Berndt, 2003) and for residential buildings. The flue gasses, especially from the burning of coal, contain a large number of particles that bind Hg, which is thus deposited locally (AMAP/UN Environment, 2019; Pirrone et al., 2010). Globally, the content of Hg in coal, oil, and gas ranges from 0.01 to  $1.5 \text{ mg kg}^{-1}$  (Gaffney, 2014), though the Hg content of Russian coal is found to vary from 0.02 to 0.4 mg kg<sup>-1</sup> (Romanov et al., 2012). Burning of fossil fuels remains a common source for heat and power generation in the Russian Barents region, both for industries (Berndt, 2003) and for residential buildings. The flue gas is high in particles that bind some Hg and is deposited locally (AMAP/UN Environment, 2019; Pirrone et al., 2010). Coal and fuel oils (Pirrone et al., 2010; Romanov et al., 2014) were traditionally used, though there has been a shift towards natural gas and biofuels that contain less Hg (AMAP/NEFCO, 2020; Gaffney, 2014). This transition is implemented at several locations, including two major heat and power plants situated on the shores of Lake Onega in Karelia (Petrozavodskmash and Kondopoga), as well as another facility in the city of Novodvinsk along the River Northern Dvina in Arkhangelsk (AMAP/NEFCO, 2020). Also, in Karelia, there is an ongoing regional programme for the transition towards local biofuels (Kireeva, 2014).

A study of two coal-fired power plant units in Russia (Reftinskaya) showed that the average Hg levels in their flue gases were 3 and 9.5  $\mu$ g m<sup>-3</sup> (Romanov et al., 2012). This is within the range of standards adopted for existing power plants by the European Union and the United States. Overall, it can thus be considered that the coal burning for power generation causes minor Hg pollution in the region. On the other hand, waste from fossil fuels is an environmental concern, such as improper handling of spent oil. In both Murmansk and Arkhangelsk Oblasts work is initiated to establish facilities for processing and decontamination of oil-containing waste (AMAP/NEFCO, 2020; Bumbulyak et al., 2013).

In the Norwegian Troms and Finnmark county, fossil fuels are basically only used for transportation. The major source of energy is from hydropower with an increasing share of wind (www.nve.no). Although not constituting a direct potential release source of Hg, hydropower dams have been found to increase the levels of Hg and methyl-Hg in the water (Pestana et al., 2019).

#### 3.1.5. Past military activities

There are a few former and active military sites within the region that may contribute with Hg releases. The most important legacy source of Hg at these sites is the use of Hg fulminate (Hg(CNO)<sub>2</sub>) as the trigger for blasting in igniting caps and detonators. For example, during WWII, Hg fulminate was used as detonator for the incendiary phosphorous bombs. Production took place in Apatity, Murmansk Oblast (Dauvalter & Kashulin, 2018). Hg fulminate was also used as detonator for the blasting during the mining operations of the apatite-nepheline deposits in Apatity (Dauvalter & Kashulin, 2018).

#### 3.2. Environmental levels of Hg in the Barents region

Levels of Hg in freshwater bodies (rivers and lakes) near the above mentioned potential Hg pollution sources are presented in Table 1 and assessed in relation to national safety limits. Compiled Hg levels in surface sediments and fish, compiled by site (and fish species) are presented in Fig. 2. Sample matrices cover water, sediment, and fish. Most data are from lakes with only a few observations in rivers.

#### 3.2.1. Freshwaters

Studies on the levels of Hg in water are limited, and those that are available often report levels below their method limit of detection

# Table 1

Overview of measured Hg in water, top sediment, and fish muscle in rivers and lakes, and with identified nearby potential release sources. Values exceeding Norwegian \_

Waterbody Location/Type/Name			Environmental compartment, Hg			Potential release sources	
			Water ( $\mu g L^{-1}$ )	**Type of activity	**Type of activity	**Type of activity	Nearby Cit
roms and Finnmark	River	Målselv	$\begin{array}{c} 0.0005 \ (n=11, \\ 0.0002 - 0.0016)^1 \\ 0.0006 \ (n=10, \\ 0.0002 - 0.0016)^2 \\ 0.0010 \ (n=4, \\ 0.0003 \ 0.0011)^3 \end{array}$			5	Bardufoss
		Kautokeino	0.0003-0.0011)		NPi: 0.250 (n = 20, 0.090 - $0.520$ ) <sup>4</sup>	(1, Cu and Ag)	Bidjovagge
		Alta	$\begin{array}{l} 0.0009 \ (n=11, \\ 0.0004 - 0.0015)^1 \\ 0.0008 \ (n=10, \\ 0.0004 - 0.0016)^2 \\ 0.0008 \ (n=4, \\ 0.0008 \ (n=0.010)^3 \end{array}$			3	Alta
		Tana	$\begin{array}{l} 0.0005-0.0012)\\ 0.0012\ (n=11,\\ 0.0005-0.0023)^1\\ 0.0009\ (n=9,\\ 0.0004-0.0027)^2\\ 0.0010\ (n=4,\\ 0.0000\ (n=4,\\ 0.000\ (n=4,\ $			6	
		Pasvik (outlet)	$\begin{array}{c} 0.0004 - 0.0016)^3\\ 0.0010\ (n=11,\\ 0.0003 - 0.0030)^1\\ 0.0006\ (n=11,\\ 0.0003 - 0.0012)^2\\ 0.0008\ (n=4,\\ 0.0002 0.0012)^3\\ \end{array}$			1 (Cu–Ni) 1(Fe)	Nikel Bjørnevatn Zapoljarny
	Lake	Various	0.0004=0.0013) $0.001 (n = 5, 0.0005=0.0016)^{32}$	$\begin{array}{l} 0.16 \; (n=122, \\ 0.002  0.333)^{33} \end{array}$		1 (Cu–Ni)	Nikel Bjørnevatn
		Rayakoski (Pasvik)	<0.01 <sup>5</sup>	0.14 <sup>5</sup>	EWf: 0.020 (n = 14, $0.002-0.036$ ) <sup>5</sup>	6	
		Hestefossdammen (Pasvik) Nyrud (Pasvik) Noatun (Pasvik)	<0.016	0.12 <sup>6 r</sup>	NPi: <b>0.760</b> $(n = 1)^7$ NPi: <b>0.583</b> $(n = 3, 0.500, 0.200)^7$	6 6 6	
		Ruskebukta (Pasvik) Vaggatem (Pasvik)	<0.01 <sup>6</sup> , <sup>33</sup> <0.01 <sup>33</sup>	0.13 <sup>6 f</sup> 0.10 <sup>5</sup> 0.06 <sup>6 f</sup>	B: $0.131 (n = 1)^8$ BT: $0.166 (n = 8, 0.084 - 0.624)^{8,9}$ EG: $0.133 (n = 1)^8$ EPe: $0.112 (n = 63, 0.024 - 0.527)^{8,9}$ EWf: $0.041 (n = 216, 0.007 - 0.058)^{5, 8-9}$ NPi: $0.181 n = 38, 0.051 - 0.667)^{8,9}$ V: $0.046 (n = 25, 0.018 - 0.064)^{8,9}$	1 (Cu–Ni) 1 (Cu–Ni)	Pasvik Pasvik
		Utnes (Pasvik)			EPi: 0.250 $(n = 1)^7$ NPe: 0.170 $(n = 1)^7$	1 (Cu–Ni)	Pasvik
		Bjørnevatn (Pasvik)		0.06 <sup>6 f</sup>	BT: $0.111 (n = 11)^9$ EPe: $0.204 (n = 19)^9$ EWf: $0.059 (n = 30, 0.04-0.087)^9$ NPi: $0.080 (n = 16)^9$	1 (Cu–Ni) 1(Fe)	Pasvik Bjørnevatn Zapoljarny
		Skrukkebukta (Pasvik)	<0.01 <sup>5</sup> , <sup>33</sup>	0.02 <sup>5</sup> 0.03 <sup>6 f</sup>	B: $0.176 (n = 5, 0.036-0.396)^8$ BT: $0.203 (n = 6, 0.082-0.72)^{7.8}$ EG: $0.047 (n = 6, 0.029-0.064)^8$ EPe: $0.307 (n = 53, 0.027-2.622)^{7.8}$ EWf: $0.066 (n = 113, 0.002-0.144)^{5,7.8}$ NPi: $0.458 (n = 13, 0.150-1.531)^{7.8}$ V: $0.056 (n = 22, 0.008)$		

(continued on next page)

#### Table 1 (continued)

Waterbody Location/Type/Name			Environmental compartment, Hg			Potential release sources	
			Water ( $\mu g L^{-1}$ )	**Type of activity	**Type of activity	**Type of activity	Nearby City
Murmansk	Lake	Kuetsjärvi (Pasvik) Saraslaki	< 0.01 <sup>5</sup>	$\begin{array}{c} 0.16^5 \\ 0.05^6 \ f \\ 0.400 \ (n=12, \\ 0.100-0.420)^{10} \\ 0.41^{11} \\ 0.51^{11} \\ 0.42^{11} \end{array}$	B: 0.216 $(n = 4)^9$ EPe: 0.064 $(n = 7)^9$ EWf: 0.027 $(n = 83, 0.002-0.15)^{5,9}$ NPi: 0.080 $(n = 9)^9$		
		Various, Kola penninsula	$0.003-0.005 (n = 3)^{31}$	0.13	BT: 0.009 (n = 5, 0.0033-0.0173) <sup>31</sup> EWf: 0.013 (n = 5, 0.0067-0.0356) <sup>31</sup>		
	River	Kola		0.11 (0.009–0.2) <sup>12</sup>	-	3, 4	Murmansk
		Tuloma	$0.0007 (n = 4, 0.0006-0.0008)^{13}$			3 (Hg waste)	Kolsky district
		Niva	$0.0004 (n = 1)^{13}$			1 (Al)	Kandalaksha
	Lake	Imandra, Bolshaya	< 0.01 <sup>14-15</sup>	0.335 (0.079 <b>-1.000</b> ) <sup>16</sup>	EWf: 0.01 (n = 18, 0.001–0.027) <sup>14-15</sup>	1 (Ni -Cu) 1 (Fe, mica, vermiculite)	Monchegorsk Olenegorsk
		Imandra, Yokostrovskaya	$< 0.01^{14-15}$	0.154 (0.018–0.305) <sup>16</sup>	EWf: 0.025 (n = 18, $0.002-0.038$ ) <sup>14-15</sup>	1 (Apatite, nephline)	Kirovsk
						4 3 (Hg waste)	Apatity Kirovsk
		Imandra, Babinskaya	$< 0.01^{14-15}, \frac{17}{7}$	0.097 (0.058–0.126) <sup>16</sup>	EWf: 0.029 (n = 8, $0.016-0.033$ ) <sup>15</sup>	1 (Fe, mica)	Kovdor
		Bolshoy Vudyavr	$< 0.05^{18}$	<b>0.52</b> (n = 10, 0.26- $0.78$ ) <sup>18</sup>	Ac: 0.067 $(n = 10)^{18}$	1 (apatite- nephline)	Kirovsk
		Umbozero		$0.079 (n = 5, 0.049 - 0.106)^{19}$	EWf: 0.039 $(n = 8)^{19}$	1 (rare metals)	Lovozero district
Karelia	Lake	Poppalijarvi Koivasjärvi Kento Lomjärvi Keski-Kuittijärvi		$\begin{array}{c} 0.125^{20} \\ 0.055^{22} \ (10 \ {\rm cm}) \\ 0.085^{20} \\ 0.038^{22} \ (10 \ {\rm cm}) \\ 0.013^{22} \ (10 \ {\rm cm}) \end{array}$	EPe: $0.322 (n = 10)^{21}$ EPe: $0.151 (n = 10)^{21}$ EPe: $0.109 (n = 19)^{21}$	1 (Fe)	Kostomuksha
		Onega, Kondopoga		$0.03^{23}$		2, 4	Kondopoga
		Onega, Petrozavodsk	$\begin{array}{l} 0.0013 \ (n=9, \\ 0.0004-0.0027)^{24, \dagger} \end{array}$	27	76	3	Petrozavodsk
Arkhangelsk	River	Northern Dvina	$\begin{array}{l} 0.0050 \ (n=10,\\ 0.0022-0.0113)^{13}\\ 0.042 \ (n=54,\\ 0.008-0.270)^{25}\\ <0.01^{26}\\ 0.0800^{27}\\ 0.0128 \ (n=34,\\ 0.00120 \ (n=0210)^{20}\\ \end{array}$	0.135 <sup>27</sup> 0.195 (n = 4, 0.0200- <b>0.8000</b> ) <sup>28</sup>	B: $0.016 (\ge 5)^{26}$ NPi: $0.075 (\ge 5)^{26}$ EWf: $0.169 (\ge 5)^{26}$ Various: $0.038$ , (n = 50, $0.002-0.127)^{27}$	2, 4 2 2 4	Arkhangelsk Novodvinsk Koryazhma Severodvinsk
		Mezen	0.0031-0.0212)		B: 0.150 (n - 357) <sup>29</sup>	6	

\*Ac = Arctic Charr (*Salvelinus alpinus*); B = Burbot (*Lota lota*); BT = Brown Trout (*Salmo trutta*); EG = Eurasian Grayling (*Thymallus thymallus*); EPe = European Perch (*Perca fluviatilis*); EWf = European Whitefish (Coregonus lavaretus); NPi = Northern Pike (*Esox lucius*); V = Vendace (*Coregonus albula*).

\*\*Type of activity: 1 = mining and metallurgical industry, 2 = historic pulp and paper production, 3 = municipal and industrial solid waste, 4 = fossil fuels, 5 = past military activities; 6 = no identified potential source of Hg.

 $^\dagger\,$  Filtered (0.45  $\mu m$ ); f value is read from figure and should be considered proximate.

- <sup>1</sup> (Braaten et al., 2020);
- <sup>2</sup> (Kaste et al., 2021);
- <sup>3</sup> (Kaste et al., 2022);
- <sup>4</sup> (Skotvold et al., 1997);
- <sup>5</sup> (Kashulin et al., 2011);
- <sup>6</sup> (Stebel et al., 2007);
- <sup>7</sup> (Berglen et al., 2018);
- <sup>8</sup> (Amundsen et al., 2023);
- <sup>9</sup> (Amundsen et al., 1997);
- <sup>10</sup> (Dauvalter, 2003);
- <sup>11</sup> (Dauvalter, 1994);
- <sup>12</sup> (Fedorov et al., 2018, and references therein);
- <sup>13</sup> This present study;
- <sup>14</sup> (Moiseenko et al., 2020);
- <sup>15</sup> (Gashkina et al., 2020);
- <sup>16</sup> (Dauvalter & Kashulin, 2018);
- <sup>17</sup> (Moiseenko et al., 2018);
- <sup>18</sup> (Pavlova et al., 2019);
- <sup>19</sup> (Dauvalter et al., 2009);
- <sup>20</sup> (Tkatcheva et al., 2004);
- <sup>21</sup> (Tkatcheva et al., 2000);

- <sup>22</sup> (Jukka et al., 2010);
- <sup>23</sup> (Tomilina & Grebenyuk, 2008);
- <sup>24</sup> (Efremova et al., 2019);
- <sup>25</sup> (Fedorov et al., 2010a);
- <sup>26</sup> (Moiseenko & Gashkina, 2020);
- <sup>27</sup> (Ovsepyan et al., 2014);
- <sup>28</sup> (Fedorov et al., 2010b);
- <sup>29</sup> (Castello et al., 2014);
- <sup>30</sup> (Sonke et al., 2018);
- <sup>31</sup> (Moiseenko & Kudryavtseva, 2001);
- <sup>32</sup> (Braaten et al., 2014);
- <sup>33</sup> (Muladal, 2020, and references therein).

#### Table 2

Information on the study rivers covering location, size, and annual Hg flux and water flow. Hg flux and water flow is "new to this study".

River name	Point of discharge (UTM)*			Riverize		New data from "this	New data from "this study"	
	East	North	Zone	Length, km	Catchment area, km <sup>2</sup>	Hg flux, kg $yr^{-1}$	Water flow, $m^3 yr^{-1}$	
Målselv	406570	7660047	34	145	3239	1.6	2.7e+9	
Alta	586586	7759686	34	242	7373	2.8	3.6e+9	
Tana	543964	7791926	35	366	16 389	6.0	6.0e+9	
Pasvik	386937	7709634	36	388	18 404	3.9	4.9e+9	
Tuloma	510151	7662955	36	64	21 700	4.9	6.7e+9	
Northern Dvina	586854	7149552	37	744	357 052	653	1.4e+11	

\* WGS84.

(LOD), which is generally at 0.01  $\mu$ g L<sup>-1</sup> (Table 1). Quantification of Hg below this level is challenging as it requires specialised and costly instrumentation. It is worth noting that several studies report Hg for filtered water samples (<0.45  $\mu$ m), and thereby excluding Hg bound to particles that typically constitute 50% of the total Hg in Arctic rivers (Dastoor et al., 2022).

The site with the highest reported levels of Hg in was the Russian River Northern Dvina ( $\overline{X}$ =0.029 µg Hg L<sup>-1</sup>, min-max: 0.002–0.270 µg Hg L<sup>-1</sup>, Table 1), which does not exceed the national drinking water limits (Russia: 0.5 µg Hg L<sup>-1</sup>, Norway: 1.0 µg Hg L<sup>-1</sup>) (Dudarev et al., 2019; FAF, 2010; NIPH, 2018). The elevated Hg levels of this river can likely be explained by the relatively high abundance of industrial activities along the river shores, comprising two pulp and paper mills, several sites with fossil fuel combustion, and a municipal solid waste site.

Elevated Hg levels were also found in lakes from the Kola peninsula of Murmansk Oblast (min-max: 0.003–0.005  $\mu$ g Hg L<sup>-1</sup>) (Moiseenko & Kudryavtseva, 2001) and in water from the Petrozavodsk Bay ( $\overline{X}$ =0.0286  $\mu$ g Hg L<sup>-1</sup>, min-max: 0.0004–0.0027  $\mu$ g Hg L<sup>-1</sup>) (Efremova et al., 2019), located northwest of Lake Onega in the Republic of Karelia (Efremova et al., 2019). At the Kola peninsula, primary sources for this Hg contamination include Cu–Ni metallurgical activities in the cities of Severonickel and Pechenganickel, and the combustion of fossil fuels, as well as municipal solid waste from the city of Murmansk (Table 1). In contrast, the Petrozavodsk Bay has no local industrial Hg sources, though a city with a population of about 300 000 is situated by the river outlet. Thus, the Hg levels, which were higher in the bay area than in the open water of the lake, can be assumed to result from urban activities (Efremova et al., 2019; Moiseenko & Sharov, 2019).

In Troms and Finnmark county, water Hg levels were consistently low in both lakes ( $\overline{X}$ =0.001 µg Hg L<sup>-1</sup>, min-max: 0.0002–0.0016 µg Hg L<sup>-1</sup>) (Braaten et al., 2014) and rivers ( $\overline{X}$ =0.0029 µg Hg L<sup>-1</sup>, min-max: 0.0002–0.0030 µg Hg L<sup>-1</sup>) (Braaten et al., 2020; Kaste et al., 2021, 2022). The Pasvik river course is the most exposed to Hg releases due to its proximity to the metallurgical activities in Nikel and Zapolyarny (Table 1). Interestingly, the highest Hg levels were measured in the nearby River Tana, which has no identified point sources of Hg in its catchment. This could indicate that the Hg released from the metallurgical activities in Pasvik does not reach the river outlet and/or is insignificant compared to the background Hg levels from legacy Hg. These elevated levels of Hg may also partly be due to increased leaching of Hg from the thawing of permafrost (Schaefer et al., 2020) in northern Norway (Borge et al., 2017).

#### 3.2.2. Freshwater sediments

Contaminated sediments represent both a present and a future source of Hg to the waterways. Levels in surface sediment layers can be used to assess the current ecological risk, while levels in deeper layers reflect legacy loadings (Tkatcheva et al., 2004), and levels at deeper depths presents the natural background levels. According to EU classification (Miljødirektoratet, 2020), Hg polluted sediment in Norway is defined as having a concentration of 0.52  $\mu$ g Hg g<sup>-1</sup> d.w., and can be categorized into moderate (0.52–0.75  $\mu$ g Hg g<sup>-1</sup> d.w.), strong (0.75–1.45  $\mu$ g Hg g<sup>-1</sup>, d.w.) pollution levels. There is currently no corresponding limit for Hg in sediments in Russia.

In Troms and Finnmark county, lake and river sediments were generally low in Hg ( $\overline{X}$ =0.022, min-max: 0.002–0.333 µg Hg g<sup>-1</sup>, Table 1 and Fig. 2). Christensen (2008) report on slightly higher Hg levels in sediments from the coastal- and north-eastern regions compared to the rest of the county. This was attributed to enhanced deposition of long-range transported Hg, by the typical higher coastal precipitation, and influence from the metallurgical activities near Pasvik, respectively. The Pasvik watercourse can be divided into "upstream" (Rayakoski, Nyrud, Noatun, Hestefossdammen, Ruskebukta, and Vaggatem) and "downstream" (Utnes, Bjørnevatn and Skrukkebukta), relative to the location of the Nikel point source. From the relatively scarce data available, listed in Table 1, there appears to be no difference in sediment Hg levels between upstream and downstream sites. An exception is Lake Kuetsjärvi, located along a tributary to the main watercourse, in close vicinity to the smelter. This lake has allegedly received wastewater outflow from the ore fields, smelters, and stockpiled tailings for more than 60 years (Dauvalter, 2003). Here, Hg sediment levels were up to four times higher (max: 0.45  $\mu$ g Hg g<sup>-1</sup>) than at the other Pasvik sites (Christensen, 2008; Kashulin et al., 2011; Skotvold et al., 1997). Lake Kuetsjärvi has allegedly received wastewater outflow from the ore fields, smelters, and stockpiled tailings for more than 60 years (Dauvalter, 2003).

Lake Imandra in Murmansk Oblast showed elevated Hg sediment levels resulting from the combination of industrial activities and natural high background levels (Moiseenko et al., 2018). The lake has a complex



**Fig. 2.** Compiled average concentration and range (min-max) of Hg in fish muscle (top,  $\mu g g^{-1}$ , w.w.) and in surface sediment (bottom,  $\mu g g^{-1}$ , w.w.) from lakes and rivers in the Barents study region. Averages are weighted by the number of observations. Illustrated by the horizontal dotted line is, for fish, the recommended safety limit (0.50  $\mu g g^{-1}$ , w.w.) and for sediments, the minimum concentration classifying as polluted (0.52  $\mu g g^{-1}$ , w.w.) See Table 1 for details.

shoreline with three main basins (north: Bolshaya, south-east: Yokostrovskaya, and south-west: Babinskaya) connected by narrow passages. The highest Hg levels were in the northern Bolshaya Imandra basin ( $\overline{X}$ : 0.335, min-max: 0.079–1.000  $\mu$ g Hg g<sup>-1</sup>, Table 1) (Dauvalter & Kashulin, 2018), where the sediments can be classified as unpolluted to strongly polluted. This contamination is likely due to the metallurgical operations in the town of Monchegorsk, where wastewater from the Cu-Ni processing is allegedly discharged into the lake. Hg levels decreases at further distances from Monchegorsk, providing additional evidence for the origin of the pollution (Dauvalter & Kashulin, 2018). In the Yokostrovskaya bay, surface sediment Hg levels were low ( $\overline{X}$ : 0.154, min-max: 0.018–0.305  $\mu$ g Hg g<sup>-1</sup>) despite its vicinity to apatite mining operations in the town of Apatity. However, the most substantial concentrations of Hg were found in the deeper sediment layers at the bay, reaching up to 2.3  $\mu$ g Hg g<sup>-1</sup> (Dauvalter & Kashulin, 2018), classified as very strongly polluted. The depth (7–9 cm) of these sediments coincides with the time period of 1930-1940s, when Hg-fulminate was used both for the mine blasting of apatite and in the production of phosphorous bombs. At the south-western Babinskaya bay, Hg sediment levels were low ( $\leq 0.126 \ \mu g \ Hg \ g^{-1}$ ) in correspondence with the limited anthropogenic activities in the region.

Lake Umbozero is neighbouring to the east of Lake Imandra, with the two lakes separated by the Khibiny mountains. Lake Umbozero is less polluted than Lake Imandra (Jernström et al., 2010). This is also reflected by lower levels of Hg in the sediment ( $\leq 0.106 \ \mu g \ Hg \ g^{-1} \ d.w.$ , Table 1 (Dauvalter et al., 2009)). The apatite-nepheline mining operations present are associated with pollution of metals other than Hg (Dauvalter et al., 2009). The large size of the lake Umbozero may further reduce the contamination problem. In the nearby smaller lake, Bolshoy Vudyavr, higher Hg sediment levels were prevalent, categorizing as polluted or strongly polluted ( $\overline{X}$ : 0.52, min-max: 0.26–0.78  $\mu g \ Hg \ g^{-1}$ ) (Pavlova et al., 2019).

The Kentti River system in Karelia, in the vicinity of an Fe mine, show elevated levels of sediment Hg (Table 1) (Jukka et al., 2010). The river starts by the Lake Kostamusjärvi, which has been used as a tailing bed, receiving effluents from the mine. While the waterflow from the lake has been blocked, some leakage has apparently occurred. The levels of Hg in the surface sediment were found to decrease when moving downstream: i.e., 0.055  $\mu$ g Hg g<sup>-1</sup> in Lake Koivasjärvi (20 km downstream); 0.38  $\mu$ g Hg g<sup>-1</sup> in Lake Lomjärvi (40 km downstream); and 0.013  $\mu$ g Hg g<sup>-1</sup> in Lake Keski-Kuittijärvi (60 km downstream) (Jukka et al., 2010).

Hg levels in the sediments from the Kondopoga bay of Lake Onega, located further south in Karelia, were  $0.03 \ \mu g \ g^{-1}$  (Table 1). The bay hosts a large pulp and paper mill, previously employing the Hg-cell process. Industrial wastewater containing toxic pollutants (and nutrients) have likely been discharged into the lake. Although this is not

reflected by the measurements, the level presented is from one single study. It is on the other hand noteworthy that the potential Hg pollution at this bay has not received more attention.

River Northern Dvina in Arkhangelsk Oblast has been subject to multiple studies documenting Hg sediment levels near its outlet (Table 1). While most observations were below the limit classified as polluted ( $<0.52 \ \mu g \ Hg \ g^{-1} \ d.w.$ ) there were a few alarmingly high levels: Sediments classified as strongly polluted, at 0.8  $\ \mu g \ Hg \ g^{-1} \ d.w.$ , were reported near the city of Arkhangelsk (Fedorov et al., 2010b). This is in accordance with the elevated Hg levels reported for river water, and thus is likely due to the emissions from the same industrial and urban sources as mentioned for the freshwater in section 3.2.1.

### 3.2.3. Freshwater fish

Consumption of fish contaminated with Hg is the major source for human uptake of Hg (Ovsepyan et al., 2014). Both Norway and Russia operate with recommended dietary limits, which are at 0.5  $\mu$ g Hg g<sup>-1</sup> w. w. in Norway (NIPH, 2020) and at 0.3 and 0.6  $\mu g$  Hg  $g^{-1}$  w.w. for non-predatory and predatory freshwater fish, respectively, in Russia (SanPiN 2.4.1.3049–13, 2013). In addition, both countries have special dietary recommendations for vulnerable groups such as pregnant women. Hg level in fish depends on several factors in addition to the level of Hg in the water (McKinney et al., 2022; Thomas et al., 2016). Fish species, size and age, food preferences, and migration patterns are important governing factors. For example, an older (carnivorous) fish that stays close to a release source can be expected to show higher Hg levels than a younger fish migrating away from the pollution source (Amundsen et al., 2023). Inside of the fish, the highest levels are in the tissue of the internal organs, while the level in muscle is most relevant for human consumption (Dauvalter & Kashulin, 2018; Dudarev et al., 2015) and will therefore be discussed here.

Within the study region, the highest Hg levels in fish muscle were at sites within the Pasvik watercourse (Table 1 and Fig. 2). Here, Hg levels in fish muscle exceeded the national dietary recommendations in individuals of northern pike (Esox Lucius), brown trout (Salmo trutta), and European perch (Perca fluviatilis). Perch, pike, and trout are all (at a certain age) carnivore species that bioaccumulate Hg and contribute to biomagnification in the food web (Amundsen et al., 2023; Thomas et al., 2016). Similar to what was found in the sediments in the Pasvik river there was no significant differences in fish Hg levels between the "downstream" and the "upstream" sites (Amundsen et al., 2023; Amundsen et al., 1997). Fish species tended to be a stronger explanatory factor than location of sampling (Fig. 2). Note that important explanatory factors such as fish size were not considered due to limited information available. Fish from the Lake Kuetsjärvi, located close to the metallurgical complex in Nikel, exhibited pathological modifications in organs and tissues, as well as other issues that were linked to pollution incidents (Stebel et al., 2007).

At several of the sites with documented high Hg levels in water and/ or sediments, the levels were not correspondingly high in fish. However, the measurements of Hg in fish were limited and usually only reported for European whitefish (Coregonus lavaretus). Although whitefish is widely distributed in the region, it is not recommended for studying pollution levels due to its low bioaccumulation potential being a relatively short-lived invertivorous fish(Amundsen et al., 2023; Thomas et al., 2016). For example, despite the very high levels of Hg in the sediments in the Bolshaya bay of Lake Imandra, the levels in whitefish were low ( $\leq$ 0.027 µg Hg g<sup>-1</sup> w.w., Table 1) (Gashkina et al., 2020; Moiseenko & Kudryavtseva, 2001). Similarly, in the polluted waters of River Northern Dvina only low levels of Hg have been reported for whitefish ( $\leq 0.169 \ \mu g \ g^{-1}$  w.w.), in addition to burbot ( $\leq 0.016 \ \mu g \ g^{-1}$  d. w.) and pike ( $\leq$ 0.016 µg g<sup>-1</sup> d.w.). In contrast, a relatively high level of Hg was measured in perch (0.322  $\mu$ g Hg g<sup>-1</sup> d.w.) taken from the Lake Poppalijärvi, near an iron mine in Karelia. Fish from the site moreover showed symptoms of liver damage (Tkatcheva et al., 2000). This contrasts to the low levels of Hg documented for the sediments of the lake

# (0.125 $\mu$ g Hg g<sup>-1</sup>) (Tkatcheva et al., 2000).

# 3.3. Relevance for the Minamata Convention

Despite the increasing focus of the international community in recent years on defining and abating sources of Hg release, there remains a significant gap in knowledge regarding a comprehensive overview of Hg release sources to the Arctic rivers and ocean. Empirical data that bridges the potential release sources with the Hg water levels and river fluxes are scarce. Also lacking is a comprehensive understanding of the potential impact these point sources of Hg contamination have on human health and the local ecosystems, both under the current and future scenarios.

Under the Minamata Convention, guidelines on the methodology for preparing inventories of releases sources for Hg was adopted in 2022 (Minamata Convention on Mercury, 2022a), almost five years after its entry into force. One of the causes for this delay in addressing releases to land and water could be the lack of a robust knowledge-basis on the characteristics, occurrence, and volume of Hg from the different release source categories. This resulted in that when the treaty was adopted there was no agreement nor compromise achieved on a definite list of release sources to be regulated, unlike the Annex D of the convention listing emission source categories regulated under the treaty. Arguably, another reason for rendering releases to water and land less priority has been their anticipated localized impact compared to emissions into air, bringing the whole discussion closer to contaminated sites and catchments, yet not to the global Hg pool and transboundary pollution.

The experts' work on the BAT/BEP guidelines for identifying and abating Hg releases to water and land is still ongoing, and it will likely take more time before the COP can adopt it. Meanwhile, this present study summarizes the data and information available for the Barents region, providing a valuable input to the background assessment and identifying needs for techniques and practices that are required to reduce the Hg release impact in the region.

#### 3.4. Seasonality and loads of riverine mercury

Monthly Hg concentrations in the five monitored Barents rivers correlated strongly to the water discharge (Fig. 3, all rivers:  $r^2 = 0.99$ , p < 0.05, without Northern Dvina:  $r^2 = 0.73$ , p < 0.05). Peak Hg levels occurred during spring snow melt when the levels were up to five times higher than during base flow. This supports the thesis of most transport occurring during spring, as described by others, e.g., (AMAP, 2021; Fisher et al., 2012; Sonke et al., 2018). Moreover, this illustrates the importance of seasonality when assessing Hg risk exposure. A second peak of Hg export during autumn rainstorms, which has been reported by others (Sonke et al., 2018), was not clearly observed here.

Annual Hg loads from the five large rivers (Table 2) adds up to 729 kg Hg y<sup>-1</sup>. This represent about 2% of the total panarctic riverine Hg export, based on the estimates by Dastoor et al. (2022). The largest river, Northern Dvina contributed with the most Hg (653 kg Hg y<sup>-1</sup>). Sonke et al. (2018) reported almost twice the Hg export for the this river (1200 kg Hg y<sup>-1</sup>, alternate name S. Dvina). This deviation could result from the combination of their more frequent sampling scheme, likely covering multiple peak level measurements, and year-to-year variation which can be substantial (Sonke et al., 2018). For the remaining rivers, lower annual Hg loads prevailed (1.6–6.0 Kg Hg y<sup>-1</sup>), which coincides with the smaller size of these catchments (Table 2 and Fig. S1).

To enable comparison between rivers of different sized catchments, the Hg load and water discharge can be normalised to catchment area, providing Hg yield ( $\mu$ g Hg m<sup>-2</sup> y<sup>-1</sup>) and water yield (runoff, cm y<sup>-1</sup>). This relationship has been used e.g., by Sonke et al. (2018) to extrapolate Hg export from a few rivers to the panarctic total. They operated with different ratios: one for the for rivers draining to the Hudson Bay (y = 0.0323x - 0.118) and another for the remaining North American-Eurasian rivers (y = 0.143x - 1.12). The large difference was



**Fig. 3.** Bars illustrating the concentration of mercury (Hg, ng  $L^{-1}$ , x-axis) measured by month in the water of Rivers Målselv, Alta, Tana, Pasvik, and Northern Dvina. The solid lines illustrate monthly measurements of water discharge (m<sup>3</sup> s<sup>-1</sup>, y-axis). Note that the scale on the axes varies.

rationalized to a higher precipitation creating a greater dilution at the former. Our estimate for Northern Dvina is comparable to the latter (Fig. S5), while the remaining smaller Barents rivers located in Norway can be described by a new and even lower relationship (Fig. 3, y = 0.0048x + 0.12,  $r^2 = 0.8$ ). A higher precipitation is prevalent int the Norwegian part of the Barents regions although not as much as in the Hudson Bay (Lammers et al., 2001). This could, in addition to the lack of point sources discharging Hg directly into the watercourses explain the discrepancy.

From the presented data, it was not possible to identify elevated Hg export from rivers that could be allocated to identified potential release from point sources. Among the studied rivers, Northern Dvina and Pasvik had the highest concentration levels and largest number of potential release sources. Surprisingly, River Pasvik showed the lowest Hg yield among the rivers (Fig. 3). This could indicate that the poorly dissolved Hg is not efficiently transported from the release site to the outlet and/or that the background level from deposited Hg (Dastoor et al., 2022) masks the contribution from the local source.

#### 4. Conclusions and future recommendations

To tackle the ongoing Hg pollution in the Arctic, it is pivotal to gain a better understanding of its sources and transport pathways. Here,

several potential Hg release sources in the Barents region have been identified, covering both historic and ongoing anthropogenic activities. The facilities at several of the active sites are currently undergoing modernisation, likely providing improved pollution control. However, limited information is available on remediation work of the anticipated contaminated sites. As summarised herein, elevated levels of Hg have been documented in waters, sediments, and fish across the region. Stores of Hg in sediments can be suspended to the waterways, and thereby pose a risk to the local population and wildlife, as well as to the Arctic Ocean. In the future, such suspensions of Hg are expected to increase because of climate change. Our new data on riverine Hg loads represent export from Barents catchments, for which data was not previously available. Strong seasonality was shown, with Hg concentrations being up to five times higher during spring snow melt compared to base flow conditions during winter. This demonstrates the importance of addressing seasonality, both for assessing riverine transport to the Arctic Ocean and for the risk of exposure. Future work calls for more measurements of Hg to be conducted at sites near the potential release sources highlighted in this review. This will serve to quantify the links between releases from point sources and Hg in fish, including the effects of stressors such as climate change. This will supply stakeholders and resource managers with information necessary to be able to select the optimum and sustainable mitigative measures, important both for the Barents (and Arctic)

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environment and the health of the regional population.

#### Author statement

Cathrine Brecke Gundersen: Conceptualization; Formal analysis; Investigation; Writing - original draft; Visualization. Evgeniy Yakushev Funding acquisition; Investigation; Resources; Project administration. Petr Terentjev: Investigation; Resources; Project administration. Nikolai Kashulin: Supervision. Vladimir Korobov: Investigation. Natalia Frolova: Investigation; Resources, Data curation; Project administration. Alexander Romanov: Writing - review & editing. Una Jermilova: Visualization. Alexey Lokhov: Investigation. Igor Miskevich: Supervision; Investigation. Ekaterina Kotova: Supervision; Investigation; Project administration. Eirik Hovland Steindal: Writing - review & editing; Conceptualization. Hans Fredrik Veiteberg Braaten: Conceptualization; Supervision; Writing - review & editing; Funding acquisition; Project administration.

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#### Data availability

Data will be made available on request.

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# Appendix A. Supplementary data

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