Article

Assessment of Mercury Concentrations and Fluxes Deposited from the Atmosphere on the Territory of the Yamal-Nenets Autonomous Area

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Abstract: The problem of mercury input and its further distribution in the Arctic environment is actively debated, especially in recent times, due to the observed processes of permafrost thawing causing the enhanced release of mercury into the Arctic atmosphere and further distribution in the terrestrial and aquatic ecosystem. The atmospheric mercury deposition occurs via dry deposition and wet scavenging by precipitation events. Here we present a study of Hg in wet precipitation on the remote territory of the Russian Arctic; the data were obtained at the monitoring stations Nadym and Salekhard in 2016–2018. Mercury pollution of the Salekhard atmosphere in cold time is mainly determined by regional and local sources, while in Nadym, long-range transport of mercury and local fuel combustion are the main sources of pollutants in the cold season, while internal regional sources have a greater impact on the warm season. Total mercury concentrations in wet precipitation in Nadym varied from <0.5 to 63.3 ng/L. The highest Hg concentrations in the springtime were most likely attributed to atmospheric mercury depletion events (AMDE). The contributions of wet atmospheric precipitation during the AMDE period to the annual Hg deposition were 16.7% and 9.8% in 2016/2017 and 2017/2018, respectively. The average annual volume-weighted Hg concentration (VWC) in the atmospheric precipitation in Nadym is notably higher than the values reported for the remote regions in the Arctic and comparable with the values obtained for the other urbanized regions of the world. Annual Hg fluxes in Nadym are nevertheless close to the average annual fluxes for remote territories of the Arctic zone and significantly lower than the annual fluxes reported for unpolluted sites of continental-scale monitoring networks of the different parts of the world (USA, Europe, and China). The increase of Hg deposition flux with wet precipitation in Nadym in 2018 might be caused by regional emissions of gas and oil combustion, wildfires, and Hg re-emission from soils due to the rising air temperature. The 37 cm increase of the seasonally thawed layer (STL) in 2018 compared to the 10-year average reflects that the climatic changes in the Nadym region might increase Hg(0) evasion, considering a great pool of Hg is contained in permafrost.

Keywords: mercury; Arctic; atmospheric wet precipitation; deposition fluxes; AMDE; permafrost thawing

Citation: Eyrikh, S.; Shol, L.; Shinkaruk, E. Assessment of Mercury Concentrations and Fluxes Deposited from the Atmosphere on the Territory of the Yamal-Nenets Autonomous Area. *Atmosphere* **2022**, *13*, 37. <https://doi.org/10.3390/atmos13010037>

Academic Editors: Sae Yun Kwon, Hélène Angot and Yan Wang

Received: 30 October 2021

Accepted: 21 December 2021

Published: 27 December 2021

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1. Introduction

Mercury is a global pollutant that can be released into the environment both from natural sources and processes (emissions from the ocean, geothermal activities and volcanic eruptions, rock weathering, and biomass burning) [1] and anthropogenic activity (coal, oil, and natural gas combustion, mining and smelting activities, gold and silver mining, waste processing, chlor-alkali and cement industry, etc.) [2]. Currently available global emissions inventories are quite complete and accurate for some anthropogenic sources; however, there are still uncertainties in the development of mercury emission inventories

for natural sources and the re-emission of Hg (existing emission estimates vary by a factor of 3) [3,4]. Most mercury polluting the Arctic results from air and ocean routes from sources outside the Arctic: intercontinental transport can cause up to 93% of Hg deposition in the Arctic [3,5,6]. With the existing air circulation system, pollutants are transported from sources thousands of kilometers away from the Arctic region (from the industrially developed regions of Europe, Asia, and North America) in winter, while in the summer, the contribution of meridional transport from medium-distance and local sources of pollution increases [5].

The biogeochemical cycle of mercury in the Arctic is associated with significant risks [7–10]. Once entered in the Arctic environment, Hg is subject to bioaccumulation in the tissues of living organisms, exerting a toxic effect on them. Moreover, biomagnification (a cumulative increase in the Hg concentrations as it moves up the food chain) is observed, causing the highest concentrations in top predators, including humans [11,12].

In the atmosphere, Hg exists in three forms: gaseous elemental Hg(0), reactive gaseous (RGM), and particulate-bound mercury (HgP). Oxidized Hg forms have much shorter atmospheric residence times than the most stable and dominant Hg(0) form, with RGM being deposited between hours and days and HgP being deposited within days to weeks. Accordingly, Hg(0) is the long-range transport form, while RGM and HgP are typically deposited locally or regionally [13,14]. Various forms of Hg are removed from the atmosphere with different efficiency by wet or dry deposition. A wet deposition involves the scavenging of gas-phase and aerosol-phase Hg(II), a dry deposition involves surface uptake of both Hg(0) and Hg(II) [13]. The conversion from Hg(0) to Hg(II) plays a key role in atmospheric and biogeochemical Hg cycling. Advances in atmospheric mercury, the dominant oxidation pathways in the atmosphere (O_3 , OH, and Br-induced), were reviewed in the paper [15]. In the Arctic are observed so-called “atmospheric mercury depletion events” (AMDE), a phenomenon discovered in 1995 by Schroeder [16], involving a sharp decrease in Hg concentration in the surface layer of the atmosphere occurring during the polar spring. AMDE chemistry is similar to the tropospheric ozone depletion (ODE) phenomena. The depletion of atmospheric Hg is thought to be caused by Hg(0) oxidation by reactive halogens, namely, Br atoms or BrO radicals and UV radiation. It was shown that this leads to refreezing sea ice forming on open waters and snowpack within 200 km of sea ice, providing a large pool of reactive halogens that drives AMDE [17]. Hg(0) is converted to RGM and/or deposited into the snowpack. Some of the deposited RGM is then reduced to Hg(0) and subsequently re-emitted to the atmosphere, while some of it remains within the snowpack. There is a delicate Hg balance in the Arctic between atmospheric oxidation and deposition on the one hand and snowpack photo-reduction and emission on the other [18]. The study AMDE in an arctic coastal location near Ny-Ålesund, Svalbard (Norway) detected high concentrations of THg deposited onto the snow surface, reaching 373 ng/L and estimated deposition fluxes of 200–2160 ng/m²; however, most of the deposited Hg was re-emitted to the atmosphere via photochemical reactions [19].

Studies of mercury fate in atmospheric air, its wet and dry deposition, and distribution among environmental components in the Arctic were mainly carried out in Canadian Arctic, Greenland, Norway, and Alaska [8,20–24]. EMEP and GMOS monitoring stations, National Atmospheric Deposition Program’s Mercury Deposition Network (NADP-MDN) as well as Canadian Air and Precipitation Monitoring Network (CAPMoN) undertake short- and long-term measurements of Hg in precipitation (wet deposition) in the Polar region in the frame of their monitoring networks [3]. Amderma is the only continuous monitoring station in the Russian Arctic where GEM was measured [25]. Hg studies focused mostly on the European part of the Russian Arctic, where the largest non-ferrous metallurgy complexes, iron ore smelting, and mining enterprises are located [26–29]. For the Siberian part, a mercury problem is observed as well, but studies are limited and are mostly related to the Russian Arctic Seas [30–32]. In the Yamal-Nenets Autonomous Area (YNAA), regional environmental monitoring has revealed the negative impact of mercury on the environment; in particular, the effects of its accumulation in vegetation and animal

organisms [33]. Screening of the mercury content in the blood and hair of the residents of the YNAA detected an excess of the permissible level of mercury, especially among the aboriginal population consuming fish and reindeer meat [34,35]. There are several main possible sources of mercury in the YNAA: transboundary transport with air masses, atmospheric emissions of mercury from fires, and significant regional and local inputs from gas and oil combustion by power plants and factories. Industrial activities in the YNAA are mainly associated with the exploration, development, and operation of oil and gas fields; therefore, the region's territory is subject to technogenic impacts of varying degrees of intensity [36]. There are 4938 million tons of current explored oil reserves and 39,281 billion m³ of current explored gas reserves within the YNAA [37]. The Yamal-Nenets Autonomous Area accounts for 80% of Russia's total natural gas production and about 8% of Russian oil [34]. Arctic landscapes are characterized by low resistance to anthropogenic influences. There are pronounced manifestations of thermal degradation of the permafrost zone in some areas of the Russian Arctic. The nature of permafrost degradation depends on technogenic activities in the region, while climate change enhances their impact [38,39]. Recent studies indicated that the Arctic is a territory of the cycle of mercury with high flows and large reserves [40]. Schuster et al. estimated that soils in permafrost regions contain 1656 ± 962 Gg Hg, of which half (793 ± 461 Gg Hg) is frozen in permafrost [41]. Permafrost contains almost twice as much mercury as all other soils, ocean and atmosphere combined, and it is ready to be released by permafrost thawing over the next century [41,42].

This work aimed to determine the level of Hg concentration and fluxes of wet deposition on the study region to assess seasonal and spatial differences and factors influencing these changes. We hope that this study will partly fill this gap on the map of the Russian Arctic and improve our understanding of the role of wet deposition in the mercury cycle in the Polar Regions.

2. Materials and Methods

2.1. Study Area

Atmospheric wet precipitations (rain, snow) were collected at two monitoring stations of the Scientific Research Centre of the Arctic organized at the urban areas of YNAA: Salekhard and Nadym (Figure 1). The territory of the Yamal-Nenets Autonomous Area is located in the north of Western Siberia, with about 50% of the territory above the Arctic Circle; it belongs to three climatic zones: arctic, subarctic, and temperate continental [37,43]. Situated on the territory of the Nadym lowland, it is bounded by the Polar Urals Mountains from the west and the Siberian Ridges from the south-east. Since there are no significant orographic barriers for the passage of air masses from the north and south, the development of the meridional circulation determines abrupt and rapid changes in synoptic conditions [44]. Nadym ($65^{\circ}32' \text{ N } 72^{\circ}31' \text{ E}$) is a circumpolar city located 100 km south of the Arctic Circle; it is located in the central part of the YNAA in a flat area near the Gulf of Ob with the unimpeded passage of air masses. Salekhard ($66^{\circ}32' \text{ N } 66^{\circ}38' \text{ E}$) is the capital of YNAA; this is the only city in the world that is located directly on the Arctic Circle. The climate of Salekhard is determined by its geographic location, because on the left, it is bounded by the spurs of the Ural Mountains, partially blocking the access of the warm air masses of the Atlantic. Mountain ranges intensify cyclonic activity. The climate of both study sites is subarctic continental, which is the Dfc Code according to Köppen climate classification types [45]. It is characterized by short cool summer and long and severe winter with storms and frequent snowfalls; the air temperature can drop to -60°C . The climate is influenced by permafrost, the proximity of the cold Kara Sea, and the abundance of bays, rivers, swamps, and lakes. Low clouds, a small number of sunny days, and deep soil freezing are observed there. Winter lasts for up to eight months, starting in October-November and ending at the end of May. Snow generally reaches its maximum depth from April to May. The polar day occurs when existing radiation inversions are eliminated; inversions in May are formed largely due to warmer air advection [44]. Total solar radiation is similar in both sites [46]; annual precipitation ranges from 400 to 500 mm, and summer precipitation prevails over

winter [47]. Under current climatic conditions, precipitation deposited on Nadya is formed mainly at an altitude of over 600 m; precipitation deposited on Salekhard, on the contrary, is below 600 m [48].

Figure 1. Study area and location of the monitoring stations.

Wet atmospheric precipitations were sampled from the end of 2016 to 2018: in the Nadya station throughout the year, and in the Salekhard station only during cold periods (owing to technical reasons). The annual distribution of the monthly precipitation and the temperature curve for the monitoring station Nadya is shown in Figure 2.

Figure 2. Mean monthly temperature (red line graph) and total monthly precipitation (bar graph, green color in 2017 and blue color in 2018) over two years for Nadya (warm periods are colored beige; cold periods are colored light blue) based on weather archive data [47].

In our study, the assignment of precipitation to cold and warm periods was justified by the type of precipitation (snow or rain) and air temperature during deposition and also confirmed by an analysis of the water isotopic composition. Under the current climatic conditions, precipitation in cold and warm periods was comparable: 174.1 mm and 241.6 mm (in 2016/2017); 253.8 mm and 211.1 mm (in 2017/2018), respectively. The annual amount of precipitation increased but did not change drastically in two years, amounting to 415.7 mm in 2016/2017 and 464.9 mm in 2017/2018.

2.2. Sample Collection and Analytical Procedure

Event-based wet precipitations (rain and snow) were collected for mercury (Hg) and trace element analyses. Though weekly collections are much more common, event-based precipitation samples provide the best opportunity to study how mercury is scavenging from the atmosphere [49]. The type of precipitation and meteorological conditions (start and end time of precipitation collection, air temperature during precipitation, wind direction, humidity, etc.) were recorded for each precipitation event. Simultaneously samples were taken for Hg, trace metals, and isotopic composition analysis. Details of sample preparation and the determination of stable isotope compositions ($\delta^{18}\text{O}$ and δD) in wet precipitation are described elsewhere [50,51].

Winter precipitations (snow) were collected into high-density polyethylene plastic bags placed into a barrel equipped with blowing protection. Snow samples were melted in closed plastic containers at room temperature, followed by volume measurement and acidification by ultrapure nitric acid (produced by Savillex DST-1000) to prevent losses and absorption of Hg on the bottle wall ($\text{pH} \leq 2$). Since the general sample was taken to analyze Hg and other elements, we preferred HNO_3 to the HCl that is more conventional for mercury analysis but has an interfering influence on the determination of other trace elements by ICPMS. Rain and mixed (rain and snow) precipitations were collected by a hand-made sampler placed into the barrel with blowing protection about 1.5 m above the ground to avoid contamination of the sample during heavy rains. The collector consisted of a 20 cm glass funnel connected with an HDPE bottle with a silicone tube. The volume of rain samples was measured, and samples were acidified by ultrapure HNO_3 similarly snow samples. Unfiltered acidified samples of wet atmospheric precipitation were transported to the Chemical Analytical Center of the Institute for Water and Environmental Problems of the Siberian Branch of the Russian Academy of Science (IWEP SB RAS) for further analysis.

Handling the laboratory samples was undertaken on a mercury-free clean bench. Five mL/L BrCl solution was added to samples at least 24 h before analysis to determine the total mercury according to Method US EPA 1631 [52]. The sample preparation procedure was controlled by using the “field blank” and “method blank” prepared from MilliQ water (18 M Ω cm quality). The total mercury concentrations were determined by Cold Vapor Atomic Fluorescence Spectrometry using mercury analyzer “Mercur DUO Plus” (Analytik Jena, Jena, Germany) with a detection limit of 0.5 ng/L. Quality control was carried out following the US EPA 1631 and fully complied with the accepted quality criteria of the method (Table 1).

Table 1. QA/QC quality criteria of the US EPA 1631 method.

Parameter	Methodological Criteria	Results
Initial and Ongoing Precision and Recovery (IPR), %		
Residual Standard deviation (RSD)	21	<5
Recovery	79–121	96–101
Matrix Spike/Matrix Spike Duplicate (MS and MSD), %		
Relative Percent Difference (RPD)	24	<10
Recovery	71–125	87–118

2.3. Calculation of Hg Volume-Weighted Concentration and Wet Deposition Flux

It should be noted that values below the detection limit were included in the calculation of average values estimated as half a detection limit value [53].

For a representative assessment of the level of mercury pollution in atmospheric precipitation, it is advisable to use volume-weighted concentrations (VWC) calculated using the Equation (1) for each precipitation event, taking into account the contribution of each event to the annual water equivalent:

$$C_{VWC} = \frac{\sum C_i \times P_i}{\sum P} \quad (1)$$

where C_i is Hg concentration in the i -th sample (ng/L); P_i —the amount of i -th sample in water equivalent (mm); P —the total amount of precipitation for the season or year in water equivalent (mm).

A quantitative assessment of the mercury deposition on the underlying surface was carried out by calculating deposition fluxes (2) for different years and seasons.

$$F = \frac{C \times P \times 0.1}{S} \quad (2)$$

where F is the Hg flux ($\mu\text{g}/\text{m}^2$) with wet atmospheric precipitation; C —the concentration of Hg in the sample ($\mu\text{g}/\text{L}$); P —the amount of precipitation in water equivalent (mm); and S —sampler area (dm^2).

Data were processed using Microsoft Excel 2010 software. Student's t -test was applied to evaluate VWCs and Fluxes differences for different monitoring stations and seasons. Differences were assessed as statistically significant at $p < 0.05$.

3. Results and Discussion

3.1. Hg Concentrations in Wet Atmospheric Precipitation of Yamal-Nenets Autonomous Area

The results of total mercury determination in wet atmospheric precipitation in the cities of Nadym and Salekhard are given in Table 2. Mercury concentration in Nadym precipitation varied from 1 ng/L to 29.4 ng/L in a warm period and from <0.5 ng/L (less than the detection limit) to 63.3 ng/L in a cold one, with the maximum concentration in the springtime (middle of May). In Salekhard, the Hg concentration range in a cold period is smaller, possibly because late spring samples were not collected; therefore, the highest Hg concentrations may have been missed.

Table 2. Average values, ranges, and volume-weighted concentrations (VWC) of Hg in wet atmospheric precipitation of Salekhard and Nadym in different seasons and years.

City	Hg Concentration, Ng/L				Period
	Min	Max	Average	VWC	
Cold period					
Salekhard	1.5	26.1	6.6	5.4	5 October 2016–01 May 2017 *
	1.2	20.4	5.5	5.6	1 November 2017–18 April 2018 *
Nadym	<0.5	27.5	6.3	6.6	26 October 2016–29 May 2017
	<0.5	63.3	14.8	8.6	14 October 2017–03 July 2018
Warm period					
Nadym	3.7	22.6	8.6	6.3	30 May 2017–13 October 2017
	1.0	29.4	12.2	10.5	4 July 2018–12 October 2018

* incomplete cold period.

The minimal and maximal Hg concentrations differ insignificantly between periods, excluding short-term high increases in springtime. The comparison of average concentrations in Nadym and Salekhard in the same cold periods showed that Hg concentrations

were similar in 2016/2017 and differed almost three times in 2017/2018, which might be explained by the incomplete sampling period in Salekhard. Moreover, average concentrations are often overestimated due to the disproportional contribution of weak short-term precipitation with high concentrations of pollutants. At the same time, VWC represents the pollution level more objectively, accounting for the real contribution of each event in total annual precipitation [49]. VWCs of mercury in atmospheric precipitation in both sites are similar, though VWCs in Salekhard are lower than in Nadym for both years; however, the difference is statistically significant only in the cold period 2017/2018. Besides the reason related to incomplete sampling, the orographic features of the city's location and differences in the cloud height where precipitations were formed might influence the levels of atmospheric pollution of Nadym and Salekhard in the winter period. In Nadym, precipitation is formed mainly at an altitude of over 600 m; in Salekhard, on the contrary, below 600 m [48]. This difference is confirmed by the excess sulfates (ex-SO_4^{2-}) analysis previously conducted at the same locations, which are indicators of anthropogenic pollution of the atmosphere. The content of ex-SO_4^{2-} (% of total sulfates) in the cold period was 37 and 19 in 2016/2017, and 34 and 8 in 2017/2018 for Salekhard and Nadym, respectively [48]. Thus, the pollution of the Salekhard atmosphere is determined mainly by regional and local sources throughout the year, while in Nadym, in the cold period, the main source of pollutants is long-range transport and local fuel combustion; in the warm season, on the contrary, internal regional sources make a greater contribution.

3.2. Seasonal and Interannual Changes of Hg Concentrations in Atmospheric Precipitation in Nadym

Most studies report that wet mercury deposition is higher in warm seasons, and this has been attributed to more precipitation [54]. However, it should be noted that most studies compare the calendar periods of the year (winter, spring, summer, and autumn), i.e., the Hg concentrations over the corresponding three-month periods [37,54]. Indeed, summer precipitation is 1.8–2.3 times greater than the precipitation in winter (depending on the study year). However, in fact, winter in Siberia lasts much longer than three months; therefore, we consider it possible to divide the periods according to the type of precipitation (rain and snow). In such a case, precipitation in cold and warm periods is quite comparable: 174.1 mm and 241.6 mm (in 2016/2017); 253.8 mm and 211.1 mm (in 2017/2018) in cold and warm periods, respectively. The seasonal dynamic of VWCs of mercury in atmospheric precipitation in Nadym is shown in Figure 3. In 2016/2017 VWCs were at the same level in the cold and warm periods (differences weren't statistically significant). An increase was observed in the cold period, and an even larger increase was observed in the warm period (differences were statistically significant), indicating the evident rise of Hg concentration in 2017/2018.

Hg pollution in Nadym in cold periods is related to long-range transport and local emissions from fuel and waste combustion. The region's sources of atmospheric moisture precipitated in the Nadym Lowland during the cold period of 2016–2017 were determined based on the joint analysis of synoptic, trajectory, and isotopic data: the Atlantic Ocean (35.7%), the North Atlantic Ocean, and the Arctic Ocean (30.4%), the Black Sea-Caspian region (20%), and inland regions (about 10%) [51].

In a warm period of the year, when the meridional transport of air masses is enhanced, the mercury level is determined by its regional background and emissions from the medium-distance intercontinental sources. Therefore, in the warm season, elevated Hg concentrations may be associated with wildfires in the tundra; also, it is worth considering such a possible Hg source as the release of mercury during the warming up of the tundra surface [41,42], which will be discussed in more detail below, in Section 3.3.

Figure 3. Seasonal volume-weighted concentrations of mercury (ng/L) in wet atmospheric precipitation in Nady in the 2016–2018 years, red and blue colors correspond to warm and cold periods, respectively.

Annual Hg VWCs in wet precipitation in Nady were 6.7 and 10.0 ng/L, demonstrating an increase of about 56% from 2016/2017 to 2017/2018. This result is at odds with a downward trend of mercury concentrations in the air and atmospheric deposition observed in recent years at many monitoring stations in the United States, Canada, and Europe that is explained by a decrease in anthropogenic emissions [23,24,55–57]. Significant long-term temporal trends in precipitation mercury concentrations were found at 34 of the 135 MDN sites investigated, with ~88% of significant trends showing decreases in VWCs [58]. However, if there was a downward trend in the eastern part of the United States, the changes were insignificant or increased in the western part, despite the absence of an increase in local mercury emissions [23]. For remote areas of the Canadian Arctic and Alaska, it was shown that interannual variability during the studied period is insignificant [53]. In the last ten years, only slight changes in mercury deposition fluxes were observed in high northern latitudes (above 60° N)—from −0.9 to +0.1% per year [55]. The study of heavy metals and mercury content in the atmosphere and atmospheric precipitation of the Arctic territories has been actively carried out in different years by scientists from other countries, including Russia [22,25,54,59,60]. Comparison of Hg concentrations in atmospheric precipitation in the city of Nady with remote areas of the Arctic showed that both the median and the two-year average VWC of mercury in Nady are at a higher level, whereas the annual fluxes are pretty comparable (Table 3).

Table 3. Concentrations and fluxes of mercury in wet atmospheric precipitation in Nady and other remote Arctic locations.

Location	Hg Concentration, ng/L		Annual Flux, µg/m ²	Study Period
	Median	Annual VWC		
Nady, Russia [this study]	6.7	8.3	3.6	2016–2018
Dalniye Zelentsy, Russia [29]	–	6.0	3.0	2001–2002
Ny-Ålesund, Norway [54]	–	4.5	1.1	2012–2015
Pallas, Finland [54]	–	6.1	2.1	2011–2014
Nome, USA [53]	3.5	6.2	2.3	2013–2015
Glacier Bay National Park, USA [53]	1.8	1.9	3.0	2010–2013
Dutch Harbor, USA [53]	2.3	2.9	4.5	2009–2015
Gates of the Arctic National Park, USA [53]	3.6	6.0	2.1	2008–2015

Due to the limited available data, we compare the values obtained in different years, realizing that this is not quite objective. We need more data for a reliable assessment of mercury's spatial and temporal input with wet deposition. The average VWC of mercury in atmospheric precipitation in Nadym, calculated over two years, is comparable with values reported for other urbanized areas [61–66] (Figure 4), although it is significantly lower than those reported for heavily air-polluted cities, such as Shanghai, China (median 113, range 1–987 ng/L) [67].

Figure 4. Annual volume-weighted concentrations of Hg in wet precipitation in various urbanized areas, ng/L (the intensity of the colour is proportional to the Hg concentration).

3.3. Wet Deposition Flux

Most of the Hg deposition (93%) in the Arctic is thought to be caused by intercontinental transport from the global Hg pool [6]. The annual wet deposition fluxes were $2.7 \mu\text{g}/\text{m}^2$ and $4.4 \mu\text{g}/\text{m}^2$ in 2016/2017 and 2017/2018, respectively. It was shown in the Northern Hemisphere that interannual differences in THg wet deposition are mostly linked with precipitation volume [54]. Annual precipitation increased slightly (session 2.1), whereas Hg wet deposition increase was more pronounced. The main contributor to atmospheric pollution in the Arctic in 2018 was Russia (52%), including internal sources located in the Arctic [68]. Sources from Kazakhstan take the second position in pollution of the Arctic by heavy metals (12%) despite the long distance from the Arctic. The global character of Hg pollution explains the high contribution of sources of other world (23%). The average annual deposition flux for the studied period was $3.6 \mu\text{g}/\text{m}^2$. A North-South Hg deposition gradient changes from $0.2 \mu\text{g}/\text{m}^2$ in the northern tundra Alaska (68.6°N) [69] to $4.8 \mu\text{g}/\text{m}^2$ in the south (57.7°N) [53], and Nadym located at 65.3°N fits well here. Hg deposition flux in Nadym is comparable to the remote territories of the Arctic zone (see session 3.2, Table 3), but it was found to be significantly lower than averaged assessments in other regions (North America— $9.5 \mu\text{g}/\text{m}^2$, Europe— $6.8 \mu\text{g}/\text{m}^2$, Australia— $5 \mu\text{g}/\text{m}^2$, China— 4.8 and $12.6 \mu\text{g}/\text{m}^2$ in remote and urban sites, respectively) [3,70–72].

Seasonal Hg wet deposition fluxes are presented in Figure 5, along with atmospheric wet precipitation. Fluxes of Hg varied from 1.14 to $2.22 \mu\text{g}/\text{m}^2$ and increased in 2017/18. The marked increase in wet Hg deposition during the warm season of 2018 cannot be attributed to increased precipitation, since precipitations were lower in the warm period than in the cold one in 2017/2018. Transboundary transport of Hg with the western and southwestern air masses prevails only in winter, and this did not change drastically from 2017 to 2018 [68]. Assessment of pollution emitted to the atmosphere from stationary sources of pollution in the YNAA (enterprises of the oil industry, mining, and processing

of minerals, the fuel and energy complex) amounted to 786 thousand tons in 2018 that is 9% less than in 2017 [36]. Local pollution from gas and oil combustion is also possible. Although Russia has a 95% standard for using associated petroleum gas and high fines for excess flaring, this is still happening. As a result of the flaring of associated gas in Russia, on average, one ton of produced oil accounts for about 8 kg of emissions of harmful substances, including mercury [73]. For example, in 2018, Tekhneftinvest was fined RUB 1.2 million for flaring more than 358 thousand cubic meters of associated petroleum gas in the YNAA. Wildfires also make a certain contribution to atmospheric Hg emissions every year. In summer, air masses entered from the side of the continent where the tundra and forests were burning mostly in June and July [74]. The annual area of wildfires in Siberia is about five million hectares; according to existing estimates, the annual atmospheric emission of mercury from fires in Siberia may amount to 2.7 tons [75]. Terrestrial lichens and mosses might be largely saturated with mercury in the northern regions. However, it should be noted that the annual area of wildfires did not rise substantially in 2018 [76]; moreover, the location of fires was closer to the study area in 2017.

Figure 5. Seasonal atmospheric wet precipitation in Nadym (mm, green triangles) and fluxes of Hg wet deposition ($\mu\text{g}/\text{m}^2$, red and blue colors correspond to warm and cold periods, respectively).

One more hypothesis is that the increase of Hg flux reflects regional input caused by high temperatures in this period. In general, in the Northern Hemisphere, the spring and summer periods of 2018 were the second warmest periods since 1958. In the Arctic, 2018 was very warm (the second in a row since 1936), the anomaly of the average annual air temperature in the latitudinal zone of 60–70° N (to which Nadym belongs) was 2.1 °C [77]. The empirical model developed on six mercury flux datasets from different terrestrial sites predicted the impact of climate change on terrestrial mercury air-surface exchange. Hg flux is expected to increase by between 15% and 43% for a 1–2 °C temperature rise by 2050 and 15–96% for a temperature rise between 1–3.7 °C by 2100 [78]. Re-emission of Hg from soils is shown to be higher at higher temperatures: soil warming and winter freeze-thaw cycles increased soil Hg(0) evasion by 31% and 35%, respectively, relative to the control plots [79]. Mercury in permafrost soils represents an environmental risk as a great reservoir of Hg containing nearly twice as much Hg as in all other soils, the ocean, and the atmosphere combined [41]. There was a 30-year lag between the start of large-scale permafrost thaw in the 1990s and the increase in Hg(0) evasion after 2020 [42]. The indicator of the state of

permafrost soils reflecting the climatic changes is the thickness of the seasonally thawed layer (STL). In the Nadym region in 2018, the STL thickness increased substantially to 30 cm compared to 2017 and 37 cm compared to the 10-year average STL being the most significant in Western Siberia [77]. Since permafrost is continuing to thaw, the increase in Hg(0) evasion might be expected.

3.4. Spring Increase of Mercury Concentrations in Wet Precipitation

Atmospheric mercury depletion events (AMDEs) are observed in springtime throughout the Arctic.

This process is considered to be the main factor in the discharge of mercury from the atmosphere, which leads to its deposition on the surface of snow and ice in the coastal zone of the Arctic seas and can lead to additional input in the Arctic from tens to hundreds of tons of Hg annually [18,19]. Currently, deposition of Hg during AMDEs is typically inferred from concentrations of THg in the snowpack and not primarily through direct measurements of dry or wet deposition [22]. Research undertaken between 2011 and 2015 showed that AMDE is evenly distributed from April to May (38% each) [80]. On the territory of the Russian Arctic, a similar study was carried out at the Amderma station (near the coast of the Kara Sea), where, since 2010, AMDE has also been recorded in the winter seasons, when there is no direct solar radiation [81]. It was shown that for the spring season, the number of AMDEs doubled from 23.2 to 40.4%, and for the winter season, the number of AMDEs increased 10-fold from 0.2 to 26.9% from 2001 to 2013 [32].

In our study, the maximum values in wet precipitation samples were found in the spring, most likely associated with the AMDE phenomenon. Figure 6 shows the Hg concentrations change in atmospheric precipitation in Nadym, demonstrating a considerable influence of AMDE on Hg deposition with wet precipitation.

Figure 6. Mercury concentrations in wet precipitation in Nadym during AMDE.

Deposition fluxes were calculated to assess the contribution of atmospheric precipitation during the AMDE period to the total annual mercury deposition. The contribution of wet precipitation during the AMDE in the total annual wet precipitation was 16.7% in 2016/2017 and 9.8% in 2017/2018 (Figure 7). This is higher than the values calculated by Obrist and colleagues [69], which demonstrated minor contributions from the deposition of Hg(II) from precipitation or AMDEs (<5%), and most of the Hg (about 70%) in the interior Arctic tundra is derived from Hg(0) deposition. It should be emphasized that this study was based on the comprehensive Hg deposition mass balance using isotope data; whereas, we calculated the contribution without taking into account dry Hg deposition.

Figure 7. AMDE contribution (dark blue) in total annual Hg wet deposition in Nadym (light blue), where the ratio of the circle sizes corresponds to the ratio of the annual Hg wet deposition values.

4. Conclusions

This study has shown that regional differences in Hg concentration and deposition in the Arctic can be substantial on temporal and spatial scales. Mercury concentrations in atmospheric wet precipitation in Nadym are higher than in Salekhard because the pollution of the Salekhard atmosphere is determined mainly by regional and local sources. In contrast, the main source of pollutants in Nadym is long-range transport in cold periods that is confirmed by the results of excess sulfates (ex-SO_4^{2-}) analysis as indicators of anthropogenic pollution of the atmosphere.

The highest Hg concentrations observed in the Nadym station in the springtime are most likely attributed to Atmospheric Mercury Depletion Events (AMDE). The contributions of atmospheric precipitation during the AMDE period to the annual Hg wet deposition were 16.7% and 9.8% in 2016/2017 and 2017/2018, respectively.

The average annual VWC in wet atmospheric precipitation in Nadym is comparable with the values obtained for other urbanized regions of the world; however, it is much higher than the values reported for remote Arctic places. However, Hg annual deposition flux in Nadym is comparable to remote territories of the Arctic zone and less than annual fluxes in continental-scale monitoring networks of other parts of the world (USA, Europe, and China).

At many monitoring stations in the United States, Canada, and Europe, a downward trend of mercury concentrations in the air and atmospheric deposition were observed in recent years. In the Nadym region, the increase of Hg concentrations and deposition fluxes in wet atmospheric precipitation in 2017/2018 comparing 2016/2017 was observed. This might be explained by the possible regional atmospheric mercury emissions from fires, gas and oil combustion. Another likely hypothesis is that mercury released from the soils caused warming in 2018, since the re-emission of Hg from soils is known to be higher when the temperature rises. The increase in the thickness of the seasonally thawed layer from 2017 to 2018 reached 30 cm in the Nadym region. Further study is required to confirm this hypothesis and identify the trend of Hg deposition with wet atmospheric precipitation on the territory of the Russian Arctic.

Since the wet deposition of mercury is an important part of its overall balance, the results of field observations can be used for model calculations and understanding the atmospheric circulation of Hg at high latitudes.

Author Contributions: Conceptualization, E.S.; methodology, E.S. and S.E.; investigation, L.S. and E.S.; writing—original draft preparation, E.S. and L.S.; writing—review and editing, E.S.; visualization, L.S.; resources and funding acquisition, E.S. and S.E. All authors have read and agreed to the published version of the manuscript.

Funding: This study was carried out within the framework of the State assignment of the Institute for Water and Environmental Problems SB RAS.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The authors are grateful to Biol.Sci. Agbalyan Elena and colleagues of the Sector for Ecological and Biological Research of the State Institution of the YNAA for the sampling and cooperation.

Conflicts of Interest: The authors declare no conflict of interest.

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