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Oceanic evasion fuels Arctic summertime rebound of atmospheric mercury and drives transport to Arctic terrestrial ecosystems

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Mercury (Hg) contamination poses a persistent threat to the remote Arctic ecosystem, yet the mechanisms driving the pronounced summer rebound of atmospheric gaseous elemental Hg (Hg⁰) and its subsequent fate remain unclear due to limitations in large-scale seasonal studies. Here, we use an integrated atmosphere-land-sea-ice-ocean model to simulate Hg cycling in the Arctic comprehensively. Our results indicate that oceanic evasion is the dominant source (~80%) of the summer Hg⁰ rebound, particularly driven by seawater Hg⁰ release facilitated by seasonal ice melt (~42%), with further contributions from anthropogenic deposition and terrestrial re-emissions. Enhanced Hg⁰ dry deposition across the Arctic coastal regions, especially in the Arctic tundra, during the summer rebound highlights the potential transport of Hg from the pristine Arctic Ocean to Arctic terrestrial ecosystems. Arctic warming, with a transition from multi-year to first-year ice and tundra greening, is expected to amplify oceanic Hg evasion and intensify Hg⁰ uptake by the Arctic tundra due to increased vegetation growth, underlining the urgent need for continued research to evaluate Hg mitigation strategies effectively in the context of a changing Arctic.

The Arctic environment, once considered pristine, has shown increasing evidence of mercury (Hg) contamination¹⁻⁵. Elevated Hg concentrations, especially methylmercury (MeHg), have been detected in Arctic fish and marine mammals, often surpassing those found in lower-latitude regions⁶⁻¹⁰. This presents a health risk to Inuit and other Indigenous Peoples who rely on the tissues of these animals as traditional food⁶. Understanding the biogeochemical cycle of Hg in the Arctic environment is, thus, crucial for a more accurate assessment

and effective mitigation of Hg exposure among vulnerable Arctic populations.

Long-term monitoring of surface air gaseous elemental Hg (Hg⁰) concentrations in the Arctic, spanning over three decades, has unveiled distinct seasonal patterns (Fig. 1a). The low atmospheric Hg⁰ concentrations in spring, often referred to as atmospheric mercury depletion events (AMDEs), have been attributed to the oxidation of Hg⁰ by reactive halogen oxidants derived from sea salt during polar

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Fig. 1 | **Seasonal variations in monthly Hg⁰ concentrations in the Arctic atmosphere. a** Monthly average of observed atmospheric Hg⁰ concentrations in surface air from four stations of Arctic coastal regions, including Alert (ALT) (82.5°N,62.5°W; 2006–2017); Zeppelin station at Ny-Ålesund (NYA) (78.9° N, 11.9° E; 2006–2017); Villum Research Station at Station Nord (SND) (81.6° N, 16.6° W; 2012–2017), and Andøya Station (AND) (69.3° N, 16.0° E; 2010–2017). The red line refers to the multi-year average (2006–2017) of simulated atmospheric Hg⁰ concentrations in each station during the same period as the observational data. Details on the origins of observations are referred to Supplementary Table S1. The Arctic map is created using Ocean Data View (https://odv.awi.de/)⁸⁹. The gray shadow is the standard deviation of observations. Red error bar shows the standard deviation of simulation. **b** Comparisons of simulated Hg⁰ concentrations and observations

across various scenario: observations (black solid line with gray shadow as standard deviation), standard simulation (red solid line), no riverine input (blue solid line) and sea-ice dynamics-induced Hg behavior turned off (yellow solid line). Red error bar is the standard deviation of standard simulation. Both the simulations and observations are the multi-year average (2006–2017) of four stations. The bars illustrate the contribution of Hg⁰ from diverse pathways, which include direct atmospheric transport of Hg via air masses and oceanic evasion. The unit of right y-axis is enlarged to avoid overlap. The sources of Hg via atmospheric transport are briefly divided into terrestrial sources and non-terrestrial sources. The oceanic evasion component includes the evasion of Hg⁰ from seawater, which originates from marine Hg pool, river input and sea-ice/snow dynamics.

sunrise and its subsequent deposition^{1.11}. During summer, the Hg⁰ concentrations rebounds, typically exceeding the ambient background concentrations in the Northern Hemisphere. This summertime rebound signifies active Hg cycling in the Arctic during a crucial period of feeding and breeding among many Arctic biota¹², thereby increasing the Hg exposure risk to the Arctic ecosystem and Indigenous Peoples, yet the cause of this rebound remains unclear.

Previous studies suggest that the elevated Hg^0 concentration during summer is caused by the re-emission of springtime AMDE- deposited Hg in snow³ or the ocean¹³, or by anthropogenic emissions via long-range transport¹⁴. However, these sources are mainly prominent in spring due to the frequent northward movement of midlatitude cyclones¹⁵ and fail to account for the rapid increase in atmospheric Hg⁰ concentrations observed during summer. A previous modeling study² attributed the summer rebound to terrestrial processes (riverine and erosion input of Hg), but it appears to overestimate the river input flux to reproduce the summer peak^{16,17}. In addition, the simulated Hg⁰ peak appears one month earlier than

observations^{2,16}, implying the occurrence of other sources to the Arctic Ocean in mid-late summer. The July peak in atmospheric Hg⁰ has been reproduced in earlier atmospheric Hg models^{18,19} by introducing a parameterized evasion flux of Hg⁰ from the Arctic Ocean that increases by latitudes. Recent field studies have also highlighted the role of local oceanic Hg emissions driven by cryospheric^{20,21} processes, such as melting sea ice and snow, as key contributors to the summertime Hg⁰ rebound and also other pollutants^{22,23}. The atmospheric Hg concentrations in the central Arctic were primarily influenced by local oceanic emissions, with minor contributions from direct anthropogenic and terrestrial sources²¹. However, these localized results may not fully represent conditions across broader Arctic regions, particularly at coastal stations where terrestrial inputs could be more prominent. Moreover, oceanic Hg sources are inherently complex, involving both direct atmospheric deposition of anthropogenic and natural Hg, as well as contributions from contemporary and legacy Hg via rivers and coastal erosion. This interaction between cryosphere, atmosphere, land, and ocean creates a highly intricate, multi-spheric phenomenon that drives the Arctic Hg⁰ summer rebound. Achieving a comprehensive, quantitative, and process-based understanding of these interconnected dynamics is essential to accurately trace the origins of the summer rebound and to evaluate the fate of Hg in the Arctic.

Here, we integrate Hg transport models across multiple layers, spanning the atmosphere, sea-ice, land, and ocean. This coupled model encompasses the GEOS-Chem model, global terrestrial mercury model (GTMM), and MITgcm model to simulate specific atmospheric, terrestrial, sea-ice dynamics and oceanic processes that govern Hg cycling in the Arctic (model details are outlined in the Methods section). The GEOS-Chem model covers atmospheric redox chemistry, transport, and deposition of Hg species. The GTMM, coupled with GEOS-Chem, handles inorganic Hg storage and emissions in soil and terrestrial ecosystems. The MITgcm simulates photo- and biological-mediated redox, oceanic transport, and physiochemical processes (i.e., remineralization and particle distribution) of Hg species. The sea-ice model, included in the MITgcm. simulates the Hg cycle in the first-year ice (FYI) and its overlying snow cover²⁴. The dynamic coupling between GEOS-Chem and MITgcm is facilitated by the NJUCPL coupler²⁵. The model outputs are evaluated against the observed Hg⁰ datasets collected from multiple shipbased measurements in the Arctic Ocean and at long-term monitoring stations along the Arctic coast (Supplementary Table S1). With our integrated model, we aim to: 1) Investigate the cause of the Arctic summer Hg⁰ rebound from the perspective of a comprehensive process-based numerical model, 2) Examine the spatial distribution of Hg⁰ in the Arctic atmosphere and ocean during summer, 3) Quantify the sources contributing to summertime atmospheric Hg⁰ concentrations and explore its potential fate.

Results and discussion

Causes of summer elemental mercury rebound in the Arctic atmosphere

To elucidate the specific cause of the summer rebound of atmospheric Hg⁰ concentrations in the Arctic, we conducted a detailed analysis using our integrated model. The summer rebound is well reproduced by our integrated model compared with the observations from four High Arctic stations (Fig. 1a, Supplementary Fig. S1). While there is a slight overestimation in Alert and Zeppelin stations at winter due to the insufficient performance of the wintertime AMDEs, the highly consistence in the atmospheric Hg⁰ concentrations from spring to summer ensure the reliability to further explore the specific mechanism of summer rebound. The transport pathways analyzed include direct atmospheric transport of Hg via air masses and oceanic evasion, as depicted in Fig. 1b. While these two pathways are overlapped–oceanic evasion involves re-emission of Hg deposited on the ocean

surface. Our analysis aimed to explicitly distinguish between these pathways by refining the model so that the atmospheric transport exclusively accounts for direct Hg transport through air masses, eliminating the influence from re-emitted oceanic sources. The atmospheric contributions are predominantly from terrestrial reemission, alongside other non-terrestrial sources, which encompass both anthropogenic and natural emissions. The overall seasonal variability of these atmospheric contributions is minimal, as illustrated in Fig. 1b.

Our findings indicate that oceanic evasion is the predominant source of atmospheric Hg⁰ during summer, surpassing contributions from atmospheric transport (Fig. 1b). Our simulation closely aligns with observational data, revealing a peak concentration of 1.7 ± 0.1 ng/m³ in July, which is consistent with observed values (Fig. 1b). Atmospheric transport contributes only 36% to the elevated summer Hg⁰ concentrations, despite marked winter intrusion of Hg⁰-rich air masses (approximately 60%) from northern Eurasia²⁶, and during spring from mid-latitude Asia and North America^{27,28}. Conversely, there is a notable shift with oceanic evasion contributing nearly 64% of Hg⁰ during the summer, predominantly influenced by warmer conditions and reduced ice cover. This process is particularly sensitive to the emissions of volatile Hg⁰ in the Arctic, enhanced by the unique meteorological and oceanic conditions prevalent during these months. Further delineating the oceanic sources, our study distinguishes three key routes contributing to the oceanic Hg evasion: river input, sea-ice dynamics (e.g., thinning and melting of snow and sea-ice), and the marine Hg pool within seawater. Although the marine pool contributes nearly 50% throughout the year, it does not explain the notable peak in July. Consequently, our analysis has shifted focus to the contributions of river input and sea-ice dynamics. These routes have been highlighted in literature regarding their roles in the observed summer Hg⁰ peak, as discussed in recent studies^{2,17,20,21}.

The effect of each route is isolated using our model simulation by turning off each route as showed in Fig. 1b. Our results underscore the critical role of sea-ice dynamics in driving the luly peak of atmospheric Hg⁰, consistent with observational data, while river input only show a pronounced influence in the elevating Hg⁰ concentrations from spring to early summer (April to June). Our integrated model indicates an increase in Hg⁰ evasion from both sea-ice surface and seawater, (Supplementary Fig. S2a) associated with melting processes that intensify in June after the disappearance of snow cover (Supplementary Fig. S2b). This melting enhances brine drainage, resulting in nearzero salinity at the ice surface, which hampers Hg stabilization due to a lack of Hg-complexing halides. Deposited divalent Hg (Hg^{ll}) on the bare sea-ice surface undergoes rapid photoreduction, and the resulting Hg⁰ is re-emitted into the atmosphere at a rate of $0.12 \pm 0.04 \,\mu\text{g/m}^2/$ month (Supplementary Fig. S2a) due to weak bonding on the ice surface²⁹. This mechanism is supported by previous observations of higher atmospheric Hg⁰ concentrations over sea-ice³⁰. The increase in seawater Hg⁰ evasion, driven by enhanced meltwater input, affects both the Hg content and its redox chemistry in surface waters. Our simulations show a steady increase in sea-ice Hg released to seawater, peaking in July alongside sea-ice melt due to rising early summer temperatures (Supplementary Fig. S2b). Along with Hg, dissolved organic matter (DOM) trapped within sea-ice is released into surface waters via meltwater. Once in seawater, the Hg from melted ice, primarily in its oxidized form, undergoes reduction through both photochemical³¹ and biotic processes³². The unique phenomenon of the continuous solar radiation during the Arctic summer promotes Hg⁰ production from photoreduction of seawater Hg^{II}, a process that can be further enhanced by the presence of DOM³³.

Elevated Hg⁰ concentrations in the surface ocean enhance air-sea exchange fluxes (0.6 ± 0.1 to $1.2 \pm 0.2 \,\mu g/m^2/month$) into the atmosphere. However, while oceanic Hg⁰ evasion nearly doubles from June to July (Supplementary Fig. S2), atmospheric Hg⁰ concentrations at



Fig. 2 | **Spatial pattern of Hg**⁰ **in the Arctic atmosphere and ocean during summer. a** Simulated and observed spatial pattern of atmospheric Hg⁰ concentrations. The blue dashed line represents the simulated edge of the Marginal Ice Zone (MIZ), while the blue solid line denotes the simulated Perennial Ice Zone (PIZ) edge. Observational data are show in circles. **b** Atmospheric Hg⁰ concentrations and sea-ice concentrations (SIC) across Arctic Ocean latitudes during summer. The correlation coefficient (R) quantifies the relationship between atmospheric Hg⁰ and SIC. Shaded areas indicate the MIZ (gray) and PIZ (pink) during the observational period. Observed atmospheric and seawater Hg⁰ concentrations are depicted by

black solid line, with details provided in Supplementary Table S1. The red solid line indicates the simulated atmospheric Hg⁰ concentrations, and the blue solid line refers to the simulated SIC. **c** Seawater Hg⁰ spatial patterns similar to panel a but for seawater Hg⁰. **d** Seawater Hg⁰ concentrations and SIC across Arctic Ocean latitudes during summer similar to panel b but for seawater Hg⁰ concentrations. **e** Total Hg input fluxes to surface ocean via sea-ice melting during summer. The period of simulated sea-ice edges for panel **a**, panel **c** is corresponding to observation, while simulated edges for panel **e** is a seasonal average in summer.

high Arctic stations remain unexpectedly stable (Fig. 1b). This discrepancy cannot be explained by a simple concentration gradient but instead reflects seasonal shifts in atmospheric stability and transport dynamics. Specifically, the equivalent potential temperature (θ) exhibits a more pronounced increase with height in July compared to June (Supplementary Fig. S3), signaling increased atmospheric stability that limits horizontal transport from oceanic sources to monitoring sites (Jozef et al., 2023). In July, warm, moist air advected from the open ocean over the cooler sea ice induces a stable atmospheric layer, decoupling the surface and reducing vertical mixing and lateral dispersion of Hg⁰ emissions across the region. Simultaneously, wind fields over northern Greenland indicate weaker wind speeds in July (Supplementary Fig. S4), leading to reduced oceanic transport. These findings underscore the limitations of simplistic, back-of-the-envelope estimates for understanding Hg cycling and emphasize the necessity of process-based transport models to accurately capture the complexities of Hg dynamics in the Arctic.

The impact of river input on the summer Hg^0 peak is less pronounced. River input contributes $8.0 \pm 1\%$ to atmospheric Hg^0 during the Arctic spring (April to June), but declines to $4.0 \pm 2\%$ from July to August. The relatively minor influence of riverine Hg on atmospheric Hg^0 compared with previous estimates^{2,17} is attributed to our model's use of a more accurate, observation-based riverine Hg flux inventory³⁴, which is less than half of the previous estimates nearly lower than earlier estimates². The revised inventory establishes empirical relationships between Hg concentrations and various factors, including freshwater discharge, concentrations of suspended sediment and dissolved organic carbon, and present-year anthropogenic Hg emission, unlike the previous inventory that only considered water flow rate^{17,35}. This finding also aligns with a recent study that found a limited impact of terrestrial Hg from pan-Arctic rivers on the summertime Hg⁰ maximum using Hg isotope fingerprints²⁰.

Spatial pattern of Hg⁰ in Arctic atmosphere and ocean

Our model reveals that the sea-ice dynamics is the main cause for the observed spatial distribution of gaseous Hg concentrations in both the Arctic atmosphere and ocean during summer. High resolution measurements of various cruises in Arctic Ocean during summer found consistently elevated trend in seawater Hg⁰ and atmospheric Hg⁰ concentrations from the Marginal Ice Zone (MIZ) to Perennial Ice Zone (PIZ)³⁶⁻³⁸. Our simulation of Hg⁰ concentrations in Arctic air and sea (>60°N) are 1.8 ± 0.1 ng/m³ and 0.16 ± 0.02 pM, respectively, consistent with ship-based observations of 1.7 ± 0.1 ng/m³ for atmospheric Hg⁰ and 0.15 ± 0.05 pM for seawater Hg⁰ (Fig. 2a, c). The model also captures a noteworthy correlation between Hg⁰ concentrations and seaice concentrations, with correlation coefficients (R) ranging from 0.47 to 0.89 (Fig. 2b, d), supporting a close relationship with sea-ice dynamics.

Our results reveal that elevated concentrations of atmospheric Hg⁰ primarily occur along the ice edge between the Marginal Ice Zone



Fig. 3 | **Conceptual model illustrating the sources and transport pathways of Hg**⁰ **in the Arctic atmosphere.** Sources include present-year anthropogenic emissions, present-year natural emissions (biomass burning and geogenic activity), and Hg⁰ re-emissions from Hg reservoirs (ocean and land). Present-year anthropogenic and natural emissions are directly transported to the Arctic atmosphere and deposited, contributing to the Hg reservoirs stored in oceanic and terrestrial compartments. Hg from these reservoirs is subsequently re-emitted through the atmosphere through oceanic evasion (from seawater and sea-ice) and terrestrial processes (soil/vegetation re-emissions and river inputs).

(MIZ) and Perennial Ice Zone (PIZ), which aligns with recent findings by a recent cruise²¹ emphasizing the role of oceanic Hg evasion from the MIZ in contributing to the summertime Hg⁰ peak in the Arctic. Simulated atmospheric Hg⁰ concentrations in the marine boundary layer (MBL) increase by nearly 15%, from 1.6 ng/m^3 over the open ocean to 1.9 ng/m³ at the MIZ-PIZ boundary, consistent with a 16% increase observed in ship-based measurements (~82°N) (Fig. 2b). Mean seawater Hg⁰ concentrations at the MIZ boundary (~71°N) also increase from 0.13 ± 0.02 pM in the lower-latitude open ocean (<71°N) to 0.17 ± 0.01 pM, mirroring ship-based observations of an increase from 0.10 ± 0.03 pM to 0.18 ± 0.05 pM (Fig. 2d). Our simulation suggests that as perennial ice degrades into seasonal sea-ice, there is a prominent release of Hg⁰ from the ocean to the atmosphere, driven by the supersaturation of Hg⁰ beneath the ice, which enhances air-sea exchange³⁷. Additionally, we found an increased input flux of Hg^{II} into the underlying ocean at the MIZ-PIZ boundary, beginning in June and peaking in July (Fig. 2e, Supplementary Fig. S5a, c, e). The released Hg^{II} is predominantly transformed into Hg⁰ through photochemical and biotic reduction processes (Supplementary Fig. S5b, d, f), facilitated by enhanced primary productivity associated with ice-edge blooms. This transformation is supported by the observed correlation between atmospheric Hg⁰ concentrations and chlorophyll-a levels in surface water²¹. Ultimately, this process raises seawater Hg⁰ concentrations in the surface ocean, which is then emitted to the atmosphere through air-sea exchange.

Quantifying the origins of Hg⁰ in the Arctic atmosphere during summer

The sources of atmospheric Hg in the Arctic can be categorized into present-year anthropogenic sources, present-year natural sources, and emissions from Hg reservoirs (ocean and land), as illustrated in Fig. 3. The interconnections between these sources and transport mechanisms are also depicted. Present-year anthropogenic emissions refer to those from current human activities outside the pristine Arctic in the present year. Given the net sink of Hg in the Arctic Ocean (where deposition exceeds evasion)^{5,24}, we classify re-emission from present-year anthropogenic deposition as part of the anthropogenic source. Present-year natural emissions include geogenic activity and biomass burning in the present year. Hg reservoirs consist of historical emissions—both natural and anthropogenic—accumulated in land, ocean, and sea-ice environments, subsequently re-emitted through oceanic (e.g., air-sea exchange) and terrestrial processes (e.g., soil/vegetation

re-emission, river input). To distinguish ocean and land contributions to atmospheric Hg⁰, we further categorize emissions from Hg reservoirs into oceanic and terrestrial re-emissions. Thus, Hg sources are segmented into four categories: present-year anthropogenic emissions, present-year natural emissions, re-emission from ocean reservoirs, and re-emission from land reservoirs. This classification enables precise quantification of each source contribution to the summer increase in atmospheric Hg⁰ concentrations. By selectively deactivating these sources in our integrated model, we quantified their individual contributions to the summertime Hg⁰ rebound in the Arctic.

Our simulation reveals that the spatial variability in summer Hg⁰ concentrations underscores the contributions from four emission sources. During summer, the mass of Hg⁰ in the Arctic boundary layer (Fig. 4a, with uncertainty ranges in Supplementary Table S2) shows that oceanic re-emissions dominate Hg⁰ evasion into the atmosphere, contributing 29.5 ± 7.3 Mg, while atmospheric transport accounts for only about 18% (6.6 \pm 0.2 Mg). The contribution from oceanic sources to Hg⁰ concentrations is prominently higher than that seen at high Arctic coastal stations (Fig. 1b), where nearly 40% of Hg⁰ originates from atmospheric transport. This discrepancy can be attributed to the larger influence of anthropogenic and terrestrial emissions, especially east of Greenland, as illustrated in Fig. 4b. Oceanic Hg primarily originates from previously deposited Hg, including 17.1 ± 4.9 Mg from marine and terrestrial Hg pools and 10.0 ± 1.7 Mg from anthropogenic sources. Figure 4b also illustrates the pronounced impact of oceanic re-emissions over the MIZ near the Greenland Sea, driven by the melting of sea ice (Fig. 2e), which releases Hg stored during AMDEs in spring. This pattern is consistent with recent isotopic analyses that identified distinct Hg isotope signatures in the same region during summer, indicative of re-emission from cryospheric sources²⁰.

Anthropogenic emissions represent the second-largest source for the summer Hg⁰ rebound, primarily through oceanic re-emission after deposition. Direct atmospheric transport accounts for only 2.6 ± 0.1 Mg. Anthropogenic Hg⁰ is predominantly transported to the Arctic via northward-moving mid-latitude cyclones from Asia and North America³⁹, subsequently depositing into the Arctic Ocean. Figure 4b shows that anthropogenic emissions account for nearly 38% of Hg⁰ in the lower Arctic (<70°N) but have a smaller influence at higher latitudes due to the Arctic dome's isolation extending north of 70°N during summer⁴⁰. The overall contribution of anthropogenic emissions exceeds recent estimates, which suggested only a minor (-2%) contribution via long-range transport in summer²¹. This difference is attributed to the explicit representation of atmospheric circulation and the re-emissions of deposited Hg from anthropogenic emissions in our process-based model.

Regarding terrestrial processes, our results show that land reemissions contribute $4.6 \pm 0.8 \text{ Mg}$ of Hg⁰ to the Arctic ground-level air, accounting for over 70% of terrestrial emissions. Figure 4b highlights the prominent role of re-emissions from coastal regions in northern Europe, likely driven by higher Hg⁰ release from snowpack in these areas, where Hg deposition is elevated⁵. During late spring, the photoreduction of Hg in snow is prominently enhanced due to rising snow temperatures⁴¹ and increased grain size⁴², which deepens sunlight penetration, thus boosting Hg reduction⁴³. While river inputs are an important Hg source to the Arctic during the spring freshet, their contribution to atmospheric Hg 0 in summer is minimal (-1.8 ± 0.4 Mg), with the most substantial influence (~6%) seen over the Siberian shelf (Supplementary Fig. S6), where 88% of Hg from rivers and coastal erosion enters the Arctic Ocean¹⁷. This influence is even less pronounced in the Greenland Sea, which is less impacted by Siberian coastal waters, explaining the absence of Hg isotopic signals of terrestrial origin in high Arctic coastal stations during summer²⁰. Despite the total riverine Hg flux across the pan-Arctic reaching ~36 Mg/year^{34,35}, the actual evasion associated with river inputs is dramatically lower, with most Hg being deposited onto Arctic Ocean shelf



Fig. 4 | **Hg mass flows and source contributions during summertime maximum Arctic atmospheric Hg⁰ concentrations. a** Sankey diagram depicting the Hg mass flows during summer, corresponding to the transport pathways and sources outlined in Fig.3. **b** Spatial distribution maps showing the contributions of various Hg⁰

sources to the Arctic ground-level air during summer, highlighting spatial variability across the Arctic region. Oceanic and terrestrial re-emissions are classified as emissions from Hg reservoirs but are presented separately here to clarify their debated impacts on Arctic Hg⁰ dynamics.



Dry Deposition Flux of Hg⁰ (µg/m²/month)



Fig. 5 | Atmospheric dry deposition of Hg⁰ and associated air mass trajectories over the Arctic coastal continents during summer. a–c Spatial distribution of the average atmospheric dry deposition flux of Hg⁰ (μ g/m²/month) across Arctic coastal continents in June, July, and August, respectively. d–f Backward air mass trajectories arriving at the Toolik Field Station (68.6° N, 149.6° W) for the

corresponding months, calculated using 6-hourly Global Data Assimilation System (GDAS) meteorological data from 2015 to 2016. Trajectory clusters are identified using cluster analysis, with the percentages shown representing the relative contribution of each air mass cluster to the total number of trajectories. Panel **d**, **e** were generated using Meteoinfo software (http://www.meteothink.org/)⁶⁹.

sediments²⁰. Natural sources make a relatively minor contribution (-13%) to atmospheric Hg⁰ concentrations, with 2.2 ± 0.4 Mg originating from biomass burning and 2.9 ± 0.2 Mg from geogenic emissions. The influence of natural emissions declines with latitude, with a more prominent impact in northern Eurasia, likely linked to boreal forests in high latitudes of Asia and North America, where frequent wildfires release substantial amounts of terrestrial Hg⁴⁴.

The potential fate of summer Hg⁰

To elucidate the fate of Arctic atmospheric Hg⁰ released during summer, we analyzed the dry deposition of Hg⁰. Our results indicate a prominent increase in Hg⁰ dry deposition across the northern Arctic coastal continents (Fig. 5a-c), particularly in the Arctic tundra, coinciding with the summertime rebound in atmospheric Hg⁰. The average dry deposition flux in the tundra (~60°N-75°N) rose from $0.21 \pm 0.20 \,\mu\text{g/m}^2$ /month in spring to $1.35 \pm 0.2 \,\mu\text{g/m}^2$ /month in July, followed by a decline in August (Supplementary Fig. S7). These values are consistent with observed Hg⁰ dry deposition in the Arctic tundra, which ranged from 0.17 ± 2.8 to $1.8 \pm 0.14 \,\mu\text{g/m}^2/\text{month}$ from spring to summer⁴⁵. This trend mirrors the variation in atmospheric Hg⁰ concentrations, suggesting a strong link between air levels and deposition patterns, which may be facilitated by enhanced atmospheric mixing during summer (θ_e difference of 7.3 K vs. 9.8 K in spring; Supplementary Fig. S8). Previous studies have shown that Hg⁰ uptake by vegetation is a key deposition pathway in the Arctic tundra, accounting for approximately 70% of Hg⁰ deposition during the growing season⁴⁶. This reinforces the simulated increase in Hg⁰ deposition, suggesting that oceanic Hg⁰ evasion plays a critical role in supplying atmospheric Hg⁰, which is subsequently deposited onto Arctic terrestrial surfaces.

To further validate this hypothesis, we conducted a cluster analysis of air mass trajectories at Toolik Field Station, consistent with the sampling period of a previous field study, using 736 backward trajectories based on 6-hourly GDAS meteorological data from June to August in 2015 and 2016 (Fig. 5d-f). The analysis revealed that the air masses arriving at Toolik during the summer were predominantly of oceanic origin. Specifically, in June, approximately 46.7% and 23.8% of the trajectories originated from the Chukchi Sea, while the remaining 29.6% of air masses were from inland regions (Fig. 5d). In July, the influence of oceanic sources increased, with 42.3% of air masses coming from the Chukchi Sea and an additional 49.2% from other parts of the Arctic Ocean, while the rest originated from the Laptev Sea (Fig. 5e). By August, air masses of oceanic origin still accounted for a substantial proportion (85.5%), with the remaining 14.5% from terrestrial sources in Siberia (Fig. 5f). This consistent dominance of oceanic air masses, particularly in July, indicates that oceanic emissions are a major contributor to the atmospheric Hg budget over the Arctic tundra, which leads to deposition onto terrestrial surfaces. Thus, marine sources appear to be a key factor in the elevated Hg concentrations observed in Arctic terrestrial ecosystems^{5,46,47}.

Implications for the Arctic Hg cycle

Our study utilizes an advanced process-based model to elucidate the mechanisms driving the controversial summer rebound of atmospheric Hg^0 in the Arctic, which is influenced by diverse re-emissions from the cryosphere^{2,3,13,17–21}. By incorporating complex, Arctic-specific processes—including AMDEs, air-sea exchange, cryospheric interactions, and riverine transport—our model builds upon and addresses key limitations of previous models^{2,16,18,19,39,48}, which may not have fully captured these dynamics. Our findings reveal that the summertime rebound of atmospheric Hg^0 is primarily driven by oceanic evasion (-80%), with contributions from seawater Hg facilitated by sea-ice melting (-42%), anthropogenic deposition (-34%), and terrestrial



Fig. 6 | Integrated modeling of Arctic mercury cycling across air, land, sea ice, and ocean. This schematic illustrates the coupled modeling framework used to simulate Arctic mercury (Hg) cycling, incorporating the atmospheric Goddard Earth Observing System-Chemistry (GEOS-Chem) transport model, the terrestrial Global Terrestrial Mercury Model (GTMM), and the oceanic Massachusetts Institute of Technology General Circulation Model (MITgcm) with its embedded sea ice module. The fully coupled system quantifies the contributions of four Hg sources– oceanic reservoirs (42%), anthropogenic emissions (34%), land emissions (16%), and natural reservoirs (8%)–to the Arctic summer rebound of atmospheric Hg⁰. The model reveals that this summer rebound enhances dry deposition across the pan-Arctic region, with prominent accumulation in the Arctic tundra. This framework provides a comprehensive understanding of summertime Hg dynamics and deposition in the Arctic system.

processes such as re-emission from land and river transport (-16%) as depicted in Fig. 6. The remaining contribution originates from natural emissions. These results align with recent field observations highlighting the critical role of cryospheric sources in summer oceanic evasion^{20,21}. Conversely, re-emissions from riverine inputs contribute less than 5%, challenging the previously held view that pan-Arctic rivers and coastal erosion were dominant sources^{2,17}. This refined understanding of the drivers behind the summer Hg⁰ rebound provides valuable support for ongoing efforts to mitigate Hg pollution in the Arctic.

The Arctic is undergoing rapid warming, characterized by significant sea-ice loss and a shift from multi-year ice (MYI) to FYI at an alarming rate of 9–15% per decade, driven by Arctic amplification^{49,50}. This amplification has resulted in warming rates two to four times the global average^{51,52}. The accelerated transition to FYI is expected to increase oceanic Hg evasion, as FYI facilitates an efficient seasonal exchange between the ocean and atmosphere²⁴. This suggests that the ongoing shift to FYI could play a critical role in enhancing oceanic Hg emissions, thereby contributing to the observed summertime rebound of atmospheric Hg. Furthermore, the increase in oceanic Hg evasion is likely to influence atmospheric transport and subsequent deposition processes, particularly enhancing Hg deposition on the Arctic coastal continent. Arctic warming-induced changes in atmospheric circulation -such as the weakening of the polar dome and altered circulation patterns-have increased north-south airflow⁵³, which might intensify the movement of Hg-enriched air masses from the Arctic Ocean to Arctic coastal regions. This process could elevate Hg deposition onto Arctic terrestrial surfaces, including tundra ecosystems. The ecological impact of Arctic warming on ecosystems, particularly on tundra ecosystem, are of critical concern. Summer warming is driving Arctic greening⁵⁴, characterized by increased plant biomass⁵⁵ as well as a shift in vegetation composition⁵⁶. This greening has the potential to enhance terrestrial Hg uptake, as Arctic tundra are considered a Hg sink⁴⁶, while transforming the Arctic Ocean from a passive receptor of Hg to an active regional source (Fig. 6). Such a transition could pose large ecological and health risks to Arctic terrestrial ecosystems. Nevertheless, the implications of vegetation shift for Hg dynamics remain uncertain, necessitating further investigation to understand these complex interactions.

Despite substantial international efforts to mitigate global Hg pollution, including the adoption of the Minamata Convention, evaluating the effectiveness of these measures in the Arctic remains challenging. Climate change-induced alterations in Arctic Hg transport and deposition processes further complicate this assessment, emphasizing the urgent need for ongoing research. A more comprehensive understanding of how these changes impact the Arctic Hg cycle is essential to accurately evaluate the effectiveness of current mercury reduction strategies and to ensure the protection of vulnerable Arctic ecosystems.

Methods

Model description

The fate and transport of Hg across various environmental compartments-namely the atmosphere, land, and ocean-are effectively captured using a linked Earth system model framework⁵⁷. This comprehensive framework integrates three models: a threedimensional (3D) atmospheric model (GEOS-Chem), a twodimensional (2D) terrestrial model (GTMM), and a 3D ocean Hg dynamic model (MITgcm). These models are interconnected through a two-way online coupling via the NJUCPL coupler⁵⁸. Within this setup, atmospheric Hg concentration and deposition data are exchanged with a frequency of 60 min between GEOS-Chem and the GTMM and MITgcm models, which in turn pass back land re-emission and ocean evasion fluxes, respectively. Initial conditions for these models are derived from established simulations representative of present-day scenarios⁵⁹⁻⁶¹.

This fully integrated framework has been previously applied to assess the influence of both natural biogeochemical cycles and anthropogenic activities on Hg dynamics, demonstrating the robustness and reliability of the coupling mechanism^{25,57,62}. Building on this established model, we have now incorporated sea-ice Hg dynamics to extend our examination of comprehensive Arctic Hg cycling through the interactions of air, land, sea-ice, and ocean as illustrated in Fig. 7. The simulation period spans from 2004 to 2017.



Fig. 7 | Schematic representation of the comprehensive coupled model framework depicting Hg transport across air-land-ice-sea interfaces. The atmospheric Goddard Earth Observing System-Chemistry (GEOS-Chem) transport model, driven by Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA2) meteorological data, provides atmospheric Hg⁰ and Hg^{II} deposition to the terrestrial Global Terrestrial Mercury Model (GTMM), which is integrated within GEOS-Chem model. Landsat Normalized Difference Vegetation Index (NDVI) are obtained from Sea-viewing Wide Field-of-view Sensor (SeaWiFS). GTMM further models the re-emission of Hg⁰ from land, soil, and vegetation. The oceanic model is Massachusetts Institute of Technology General Circulation Model (MITgcm), incorporating ocean state estimates from Estimating the Circulation and Climate of the Ocean version 4 (ECCO v4), with an integrated sea-ice module that includes Hg dynamics. GEOS-Chem model provides Hg deposition and Hg⁰ concentrations to both the ocean and sea-ice modules. The MITgcm outputs Hgo evasion as an oceanic source to GEOS-Chem model. The sea-ice acts as an intermediary between the atmosphere and ocean, regulating Hg transport associated with thermal variations in sea-ice environments. NJUCPL is a previously-developed coupler⁵⁸ employed to couple the atmospheric, land, and ocean models, ensuring consistent data exchange across compartments.

GEOS-Chem

The GEOS-Chem model (Version 13.3.0) is utilized to simulate the atmospheric transport of mercury (Hg), along with its dry and wet deposition processes. This version integrates updated chemical mechanisms for atmospheric Hg chemistry, following the latest advances outlined in a recent study⁶³. These mechanisms encompass the oxidation of Hg⁰ by bromine (Br) and the hydroxide radical (OH), followed by the oxidation of monovalent mercury (Hgl) by ozone and various radicals, as well as the photolysis of Hg^{II} in both gas and aqueous phases. The bromine chemistry over Arctic sea-ice was modeled using an updated formulation, where the concentrations of bromine oxide (BrO) in the boundary layer are regulated by air temperature and BrO_x radicals $(BrO_x = Br + BrO)^2$. This process was specifically conducted in its native resolution grid squares when there is an adequate sea-ice cover and exposure to incident shortwave radiation, representing the characteristic spring AMDEs observed in polar regions. The Hg emission inventory utilized in this model is a modified version of the EDGARv4.tox2. This adaptation aggregates emissions both by country and by sector⁶². The GEOS-Chem model is driven by atmospheric forcing data provided by NASA/GMAO's MERRA2 reanalysis, which offers a native resolution of 0.5° latitude × 0.625° longitude. The model operates on a 10-min timestep and is conducted at a horizontal resolution of $4^{\circ} \times 5^{\circ}$ with 47 vertical layers extending to the mesosphere.

GTMM

The dynamics of Hg within terrestrial ecosystems, including the deposition and subsequent re-emissions from components such as leaves, litter, and soil, are simulated using the GTMM. This model encompasses a single-layer representation of the top 30 cm of soil across a spatial resolution of $1^{\circ} \times 1^{\circ}$. GTMM employs a mechanistic approach, based on the Carnegie-Ames-Stanford Approach (CASA)

terrestrial biogeochemical cycle model, to simulate the storage and emissions of inorganic mercury from soil and other terrestrial ecosystem components. Hg fate within the soil pool and its exchange between the land and atmosphere are modeled with a monthly timestep within GTMM. The implementation of GTMM as a module within the GEOS-Chem model adheres to methodologies established in a prior study⁶¹, ensuring a comprehensive integration of terrestrial and atmospheric Hg processes.

MITgcm-sea-ice-Hg model

The oceanic chemistry and transport of Hg, including Hg redox chemistry⁶⁴, partitioning onto particulate Hg⁶⁰, sinking to the ocean floor, and riverine Hg input, are simulated using the MITgcm model¹⁶. The model has a horizontal resolution of $1^{\circ} \times 1^{\circ}$ with 50 vertical layers, and a higher resolution over Arctic regions (approximately 30 km× 30 km) based on the Estimating the Circulation & Climate of the Ocean (ECCO v4) framework, which provides ocean state estimates⁶⁵. The MITgcm was run from 2004 to 2017, constrained by the availability of ocean forcing data from ECCO V4, with a time step of 3600 s. The first two years were treated as spin-up to stabilize the model. This simulation period matches the timeframe of most ship-based measurements of Arctic Hg detailed in the following Observation Data Sets section. The sea-ice-Hg model, integrated as a module of MITgcm, is based on a variant of the viscous-plastic dynamic-thermodynamic sea-ice model^{66,67}. This model incorporates transport processes of Hg in the sea-ice environment such as freeze rejection, snow flooding, melting, and brine drainage²⁴. The processes of brine drainage and ice melting represent distinct mechanisms affecting sea-ice. Brine drainage involves the downward movement of dense, salty brine through ice channels due to gravity, occurring predominantly during the formation and growth stages of sea-ice. This mechanism is a main contributor to the Hg output from sea-ice environments in the spring. In contrast, ice melting is driven by thermal variations, primarily occurring in the summer due to rising temperatures. The photochemical transformation between Hg⁰ and Hg^{II} in the sea-ice environment is also accounted for, with the rate coefficients of photo redox determined by radiation intensity⁶⁸. Re-emissions from snow are calculated using the parameterization of molecular diffusion transport of Hg⁴⁸. It is important to note that the sea-ice-Hg model primarily represents FYI and does not include a detailed representation of MYI. The air-ice-sea exchange of Hg, including atmospheric dry and wet deposition of Hg^{II}, oceanic Hg⁰ evasion, and re-emissions from snow and sea-ice²⁴, is handled by the NJUCPL²⁵. The input of Hg from river freshwater runoff to coastal oceans is derived from the latest observation-based riverine Hg flux inventory³⁴. This updated inventory improves empirical relationships between Hg concentrations and freshwater discharge, suspended sediment concentration, DOC, and present-day anthropogenic Hg emissions, unlike previous inventories that only considered water flow rate impacts on river Hg flux^{17,35}.

Cluster analysis of air mass trajectories

To investigate the origins and transport pathways of air masses contributing to atmospheric Hg⁰ concentrations during summer, we performed a cluster analysis of air mass trajectories at Toolik Field Station (68.6° N, 149.6° W). The analysis was conducted using MeteoInfoMap (http://www.meteothink.org/)⁶⁹, employing a 168-h (7-day) backward trajectory calculation⁷⁰ with 6-hourly GDAS meteorological data. GDAS, produced by the National Centers for Environmental Prediction (NCEP), assimilates global meteorological observations to generate comprehensive gridded datasets, providing essential inputs for trajectory modeling. The analysis focused on the summer months–June, July, and August–for both 2015 and 2016, corresponding to the sampling timeframe of a previous field study that measured stable Hg isotopes at the same location⁴⁶. The backward trajectories were generated for each day across these three months, resulting in 736 air mass trajectories in total. The cluster analysis was then applied to group the air mass trajectories based on their similarity, enabling us to identify the major source regions contributing to atmospheric Hg⁰ observed at Toolik Field Station during the summer.

Observation data sets

We collected gaseous Hg⁰ concentrations from both the air and ocean across various Arctic expeditions to assess the uncertainty arising from model coupling. Atmospheric Hg⁰ data were obtained from several cruises conducted in 2004, and 2020. In 2004, atmospheric Hg⁰ concentrations were measured during a summer expedition from Bremerhaven, Germany, to the Arctic Ocean and North Atlantic³⁰, spanning June 16 to August 29, 2004. For 2020, atmospheric Hg⁰ concentrations were collected during the Multidisciplinary Drifting Observatory for the Study of Arctic Climate (MOSAiC) expedition²¹, conducted from June 9 to September 30, 2020, within a geographical range of 78.34°–90°N and 40.93°W–175.7°E.

Seawater Hg⁰ observations were also collected during several expeditions. In 2004, seawater Hg⁰ data were obtained during the Oden icebreaker expedition from July 13 to September 25, 2004³⁶. Seawater Hg⁰ concentrations in the Canadian Arctic were also measured aboard the Canadian Coast Guard Ship (CCGS Amundsen)⁷¹ between August 16 and October 13, 2005. Additionally, dissolved Hg⁰ were measured with five-minute temporal resolution aboard the United States Coast Guard Cutter (USCGC Healy) during the U.S. GEO-TRACES Arctic (GN01) cruise³⁷, covering the Western Arctic Ocean from August 9 to October 12, 2015, from Dutch Harbor, Alaska, to the North Pole and back. Corresponding Hg⁰ evasion fluxes were collected to validate our simulated air-sea exchange fluxes of Hg⁰.

In addition to these cruise datasets, long-term atmospheric Hg^0 data were gathered from four Arctic coastal monitoring stations: Alert, Canada (82.5°N,62.5°; 2006–2017), Zeppelin Mountain, Norway (78.9° N, 11.9° E; 2006–2017), Villum Research Station, Greenland (81.6° N, 16.6; 2012–2017), and Andøya Station (69.3° N, 16.0° E, 2010–2017). More details about the observation datasets are provided in Supplementary Table S1. It is important to note that several relevant expeditions in the Arctic have measured additional species of Hg^{72-76} , which were not included in this model evaluation, as our focus was primarily on air-sea surface interactions.

Model evaluation

To ensure the reliability of our coupled model, we present the evaluation of our model's performance in simulating atmospheric Hg⁰ concentrations, surface ocean Hg⁰, and the air-sea Hg⁰ flux using both model-observation comparisons and statistical analysis. The model outputs are systematically compared against corresponding observational data across various spatial locations as listed in Supplementary Table S1. Our analysis evaluates the accuracy of the model's standard configuration (C1) and assesses the impact of omitting critical processes and emission sources, including air-sea exchange, sea-ice dynamics, river input, present-year anthropogenic emissions, present-year natural emissions, and re-emissions from Hg reservoirs (C2-C8) as illustrated in Supplementary Fig. S9. The results are summarized based on statistical indices, including the coefficient of determination (R²) and root mean square error (RMSE), as illustrated in Supplementary Fig. S10. Additionally, Kruskal-Wallis statistical tests highlight the significance of various environmental processes and emission sources on Hg cycling in the Arctic (Supplementary Fig. S11). The details of the assessment of various key processes are elaborated in the Supplementary Information.

In simulating ground-level atmospheric Hg⁰ concentrations (top row of Supplementary Fig. S10), the standard case (C1) achieves an R² of 0.50 and RMSE of 0.12. This indicates a moderate correlation with observations, suggesting the model effectively captures the general spatial patterns of Hg⁰ in the atmosphere, though it overestimates concentrations in high-latitude regions. The Kruskal–Wallis test reveals significant differences (p < 0.05) between the observation and various sensitivity tests (Supplementary Fig. S11), particularly for scenarios excluding various emissions. These emissions substantially impact atmospheric Hg⁰ concentrations, as seen in the large deviations from baseline ranks. The exclusion of air-sea exchange (C2) results in a lower R² of 0.47, and the RMSE jumps to 1.01, reflecting a severe underestimate of atmospheric Hg⁰ concentrations. The statistical analysis further supports this, with air-sea exchange showing a significant departure from observational ranks (p < 0.05). River input (C3) and sea-ice dynamics (C4) also influence Hg⁰ concentrations, though their effects are more localized^{17,21}, as evidenced by the moderate shifts in ranks in the Kruskal–Wallis test.

For surface ocean Hg⁰ concentrations, the model's standard configuration (C1) shows reasonable accuracy with an R² of 0.52 and RMSE of 0.04. When air-sea exchange is excluded (C2), the simulated surface ocean Hg⁰ concentrations increase dramatically, with R² reduced to 0.46 and RMSE increased to 0.26, underscoring the critical role of this process in regulating surface Hg⁰ levels. The Kruskal-Wallis test (Supplementary Fig. S11) corroborates this finding, showing significant rank shifts for the air-sea exchange scenario, reflecting its importance in modulating surface ocean Hg dynamics. Interestingly, the omission of sea-ice dynamics (C4) also shows a significant impact on surface ocean Hg⁰ concentrations, with a notable reduction in model accuracy ($R^2 = 0.07$) and an increased RMSE (0.09). This is further supported by the Kruskal-Wallis test, which shows a significant difference (p<0.05) for sea-ice dynamics, reflecting its influence on Hg⁰ cycling in polar regions³⁷. In contrast, the effect of excluding river input and emissions is less significant, as indicated by a more modest shift in ranks and a smaller change in model performance. While Hg reservoirs significantly influence seawater Hg⁰ concentrations, the present-year anthropogenic and natural emissions have a comparatively minor impact on these concentrations (Supplementary Fig. S11). This suggests that the large-scale oceanic Hg dynamics exhibit a relatively longer response time to recent emission sources⁷⁷, highlighting the lag effect in oceanic Hg⁰ adjustments to changes in anthropogenic and natural inputs. For the air-sea Hg⁰ flux, the model captures broad flux patterns with an R² of 0.34 and an RMSE of 0.42 in the standard case (C1). The exclusion of air-sea exchange (C2) has the most dramatic impact. The absence of sea-ice Hg dynamics also shows a low model performance ($R^2 = 0.23$), indicating that it plays a key role in modulating Hg⁰ fluxes to the atmosphere.

The Kruskal–Wallis test results across all three variables (atmospheric Hg⁰, seawater Hg⁰, and Hg⁰ flux) indicate that air-sea exchange, and sea-ice dynamics processes exert significant control over Hg cycling in the Arctic, as shown by significant rank differences (p < 0.05). Hg⁰ re-emission from Hg reservoirs also emerge as critical factors influencing Hg distributions and fluxes, particularly in the atmospheric contexts. This analysis highlights the importance of accurately representing key processes, such as air-sea exchange and sea-ice dynamics, to fully capture the complexities of Hg cycling in the Arctic. The statistical results provide further evidence that omitting these processes results in substantial deviations from observed data, underscoring their importance in Arctic Hg models.

Model uncertainty

Our study faces uncertainties primarily arising from model parameterizations and emission inventories, which are critical to accurately simulating the spatial and temporal dynamics of atmospheric Hg⁰. Firstly, while the GEOS-Chem model used in this study has known limitations in underestimating atmospheric Hg^{II} at mid-latitude sites⁷⁸, its representation of Arctic bromine chemistry over sea ice has been refined², leading to improved accuracy. Our previous validation against observed total Hg in surface snow²⁴ also supports the reliability of the model in representing Arctic atmospheric Hg^{II}. However, substantial uncertainties still exist regarding air-sea transport fluxes of Hg and emission sources. The uncertainty in the simulated air-sea fluxes of Hg⁰ is particularly sensitive to the parameterization of air-sea exchange, which depends on the exchange velocity (k) as a function of wind speed. Various parameterizations-including linear, quadratic, and cubic forms of wind speed-are derived from in-situ field surveys. While this study adopts a cubic form parameterization⁷⁹, we also examined linear⁸⁰ and quadratic⁸¹ forms (Supplementary Fig. S12). The linear form notably underestimated Hg⁰ flux, suggesting it may inadequately capture the dynamics of Hg⁰ transfer velocity, whereas the quadratic form showed slight deviations from the standard scheme. Recent measurements indicate that a cubic dependence on wind speed is more appropriate, particularly in high-wind-speed regions⁸². This suggests that our model might still underestimate Hg⁰ emissions driven by high wind speeds, such as those associated with wave breaking and bubble formation, further contributing to uncertainties in Hg⁰ evasion. The uncertainty in Hg⁰ evasion is also influenced by sea-ice dynamics. Our sea-ice model employs a simplified thermodynamic approach⁶⁷, which does not fully account for the complex thermo-dynamic interactions that govern sea-ice behavior. Although our model captures the seasonal variability of sea ice, the rapid variations in sea-ice concentrations may amplify Hg⁰ evasion during warmer seasons due to an overestimated open ocean area. Additionally, the model limitations in reproducing the characteristics of MYI result in a younger simulated MYI than observed, reducing the retention time of Hg in sea-ice environments and leading to larger Hg input through FYI melting²⁴. Despite these limitations, the boundary between FYI and MYI is reasonably captured, indicating a credible spatial distribution of seawater Hg⁰ in the Arctic. Nevertheless, the representation of Hg dynamics within the sea-ice model remains insufficient, underscoring the need for further improvements to better capture the contribution of sea ice to Hg cycling.

The simulation period represents a limitation of our study, as it was restricted by the availability of ocean state estimation data from ECCO v4, which is currently only updated to December 2017⁸³. This constraint limits our ability to fully capture recent and potentially critical changes in Arctic dynamics, such as accelerated sea-ice loss and alterations in oceanic circulation due to climate change⁵¹. Extending the simulation period with up-to-date datasets could provide a more comprehensive understanding of recent trends in Hg cycling and their interaction with rapidly evolving Arctic environmental conditions. Despite this limitation, our integrated process-based model represents an important attempt to study Arctic Hg cycling and provides a valuable foundation for future assessments. The model framework could be transplanted to more comprehensive Earth system models to better evaluate the effects of climate change on Hg cycling⁸⁴. Another notable limitation arises from the emission inventories used to quantify the contributions of anthropogenic and natural emissions. The anthropogenic emission inventory, although relatively recent^{62,85}, lacks future scenario-specific emissions, which may affect our capacity to project and evaluate the effectiveness of mitigation measures. Moreover, the natural emission inventory used in our study did not include contributions from hydrothermal venting, an important geogenic source of Hg⁸⁶. Incorporating these emissions in future modeling efforts would enhance the accuracy of simulated Hg budgets and provide a fuller understanding of natural contributions to Arctic Hg dynamics.

Data availability

The observational Arctic atmospheric gaseous Hg⁰ concentrations during the summer expedition from Bremerhaven, Germany, to the Arctic Ocean and North Atlantic³⁰ are provided by Dr. Katrine Aspmo with prior permission. The measured atmospheric gaseous Hg⁰ concentrations during the U.S. GEOTRACES Arctic (GN01) cruise are available through the Biological and Chemical Oceanography Data Management Office (BCO-DMO) (https://www.bco-dmo.org/dataset/). Atmospheric Hg⁰ concentrations collected during the Multidisciplinary Drifting Observatory for the Study of Arctic Climate (MOSAiC) expedition are accessible through Scientific Data (https:// doi.org/10.18739/A2C824G3G)⁸⁷. Data for seawater Hg⁰ concentrations are also available through BCO-DMO database. Oceanic Hg⁰ evasion fluxes in the surface Arctic Ocean were sourced from previously published studies (Table S1) with the permission from the respective authors. Atmospheric gaseous Hg⁰ concentrations from four monitoring stations were obtained from the European Monitoring and Evaluation Programme (EMEP) database (https://ebas-data.nilu.no). The GDAS meteorological data used in the analyses were downloaded from NOAA's archive (https://www.ready.noaa.gov/data/archives/ gdas1/). Simulated Arctic atmospheric Hg⁰ concentrations at Arctic stations, generated as part of this study, are provided in the Source data file. Source data are provided with this paper.

Code availability

In adherence to principles of transparency and reproducibility, we have made our code accessible to facilitate the replication and validation of our findings. The GEOS-Chem (GTMM included), MITgcm, and the coupler are provided here at our research group website: (https://www.ebmg.online/mercury/)⁸⁸. Any additional information required to reanalyze the data reported in this paper is available from the lead contact (yzhang127@tulane.edu).

References

- 1. Schroeder, W. H. et al. Arctic springtime depletion of mercury. *Nature* **394**, 331–332 (1998).
- 2. Fisher, J. A. et al. Riverine source of Arctic Ocean mercury inferred from atmospheric observations. *Nat. Geosci.* **5**, 499–504 (2012).
- 3. Lindberg, S. E. et al. Dynamic oxidation of gaseous mercury in the arctic troposphere at polar sunrise. *Environ. Sci. Technol.* **36**, 1245–1256 (2002).
- 4. Segato, D. et al. Arctic mercury flux increased through the Last Glacial Termination with a warming climate. *Nat. Geosci.* **16**, 439–445 (2023).
- 5. Dastoor, A. et al. Arctic mercury cycling. *Nat. Rev. Earth Environ.* **3**, 270–286 (2022).
- 6. AMAP. AMAP Assessment 2011: Mercury in the Arctic. Assessment (AMAP, 2011).
- Clarke, T. Mercury falling into food chain. Nature https://doi.org/10. 1038/news010906-2 (2001).
- Dietz, R. et al. What are the toxicological effects of mercury in Arctic biota? Science of the Total Environment. 443, 775–790 (2013).
- Owens, B. Seafood diet killing Arctic foxes on Russian island. Nature https://doi.org/10.1038/nature.2013.12953 (2013).
- Wang, K. et al. Subsurface seawater methylmercury maximum explains biotic mercury concentrations in the Canadian Arctic. Sci. Rep. 8, 14465 (2018).
- Steffen, A. et al. A synthesis of atmospheric mercury depletion event chemistry in the atmosphere and snow. *Atmos. Chem. Phys.* 8, 1445–1482 (2008).
- Laidre, K. L. & Regehr, E. V. Arctic marine mammals and sea ice. In Sea Ice, 3rd ed. https://doi.org/10.1002/9781118778371.ch21 (2016).
- Hirdman, D. et al. Transport of mercury in the Arctic atmosphere: Evidence for a springtime net sink and summer-time source. *Geophys. Res. Lett.* 36, 174–178 (2009).
- Durnford, D., Dastoor, A., Figueras-Nieto, D. & Ryjkov, A. Long range transport of mercury to the Arctic and across Canada. *Atmos. Chem. Phys.* **10**, 6063–6086 (2010).
- Fuelberg, H. E., Harrigan, D. L. & Sessions, W. A meteorological overview of the ARCTAS 2008 mission. *Atmos. Chem. Phys.* 10, 817–842 (2010).

Article

- Zhang, Y. et al. Biogeochemical drivers of the fate of riverine mercury discharged to the global and Arctic oceans. *Global Biogeochem. Cycles* 29, 854–864 (2015).
- 17. Sonke, J. E. et al. Eurasian river spring flood observations support net Arctic Ocean mercury export to the atmosphere and Atlantic Ocean. *Proc. Natl. Acad. Sci. USA.* **115**, E11586–E11594 (2018).
- Dastoor, A. P. & Durnford, D. A. Correction to Arctic Ocean: Is It a Sink or a Source of Atmospheric Mercury? *Environ. Sci. Technol.* 53, 9966–9966 (2019).
- Angot, H. et al. Chemical cycling and deposition of atmospheric mercury in polar regions: review of recent measurements and comparison with models. *Atmos. Chem. Phys.* **16**, 10735–10763 (2016).
- Araujo, B. F. et al. Mercury isotope evidence for Arctic summertime re-emission of mercury from the cryosphere. *Nat. Commun.* 13, 4956 (2022).
- Yue, F. et al. The Marginal Ice Zone as a dominant source region of atmospheric mercury during central Arctic summertime. *Nat. Commun.* 14, 4887 (2023).
- Boyer, M. et al. A full year of aerosol size distribution data from the central Arctic under an extreme positive Arctic Oscillation: insights from the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) expedition. *Atmos. Chem. Phys.* 23, 389–415 (2023).
- 23. Schmale, J., Zieger, P. & Ekman, A. M. L. Aerosols in current and future Arctic climate. *Nat. Clim. Chang.* **11**, 95–105 (2021).
- Huang, S. et al. Modeling the Mercury Cycle in the Sea Ice Environment: A Buffer between the Polar Atmosphere and Ocean. *Environ. Sci. Technol.* 57, 14589–14601 (2023).
- Zhang, Y. et al. An updated global mercury budget from a coupled atmosphere-land-ocean model: 40% more re-emissions buffer the effect of primary emission reductions. One Earth 6, 316–325 (2023).
- Hirdman, D. et al. Source identification of short-lived air pollutants in the Arctic using statistical analysis of measurement data and particle dispersion model output. *Atmos. Chem. Phys.* **10**, 669–693 (2010).
- Monks, S. A. et al. Multi-model study of chemical and physical controls on transport of anthropogenic and biomass burning pollution to the Arctic. *Atmos. Chem. Phys.* **15**, 3575–3603 (2015).
- Willis, M. D., Leaitch, W. R. & Abbatt, J. P. D. Processes Controlling the Composition and Abundance of Arctic Aerosol. *Rev. Geophys.* 56, 621–671 (2018).
- Amyot, M., Mierle, G., Lean, D. R. S. & McQueen, D. J. Sunlightinduced formation of dissolved gaseous mercury in lake waters. *Environ. Sci. Technol.* 28, 2366–2371 (1994).
- Aspmo, K. et al. Mercury in the Atmosphere, Snow and Melt Water Ponds in the North Atlantic Ocean during Arctic Summer. *Environ. Sci. Technol.* 40, 4083–4089 (2006).
- Luo, H., Cheng, Q. & Pan, X. Photochemical behaviors of mercury (Hg) species in aquatic systems: A systematic review on reaction process, mechanism, and influencing factor. *Sci. Total Environ.* **720**, 137540 (2020).
- Mason, R. P., Rolfhus, K. R. & Fitzgerald, W. F. Methylated and elemental mercury cycling in surface and deep ocean waters of the North Atlantic. *Water Air Soil Pollut.* **80**, 665–677 (1995).
- Schartup, A. T., Ndu, U., Balcom, P. H., Mason, R. P. & Sunderland, E. M. Contrasting effects of marine and terrestrially derived dissolved organic matter on mercury speciation and bioavailability in seawater. *Environ. Sci. Technol.* 49, 5965–5972 (2015).
- Liu, M. et al. Rivers as the largest source of mercury to coastal oceans worldwide. Nat. Geosci. 14, 672–677 (2021).
- 35. Zolkos, S. et al. Mercury Export from Arctic Great Rivers. *Environ. Sci. Technol.* **54**, 4140–4148 (2020).

- Andersson, M. E., Sommar, J., Gårdfeldt, K. & Lindqvist, O. Enhanced concentrations of dissolved gaseous mercury in the surface waters of the Arctic Ocean. *Mar. Chem.* **110**, 190–194 (2008).
- DiMento, B. P., Mason, R. P., Brooks, S. & Moore, C. The impact of sea ice on the air-sea exchange of mercury in the Arctic Ocean. *Deep. Res. Part I Oceanogr. Res. Pap.* 144, 28–38 (2019).
- Temme, C. et al. Air/water exchange of mercury in the north atlantic ocean during arctic summer. In XIII International Conference on Heavy Metals in the Environment, Rio Janeiro (2005).
- 39. Dastoor, A. et al. Arctic atmospheric mercury: Sources and changes. Sci. Total Environ. **839**, 156213 (2022).
- Klonecki, A. et al. Seasonal changes in the transport of pollutants into the Arctic troposphere-model study. J. Geophys. Res. Atmos. 108, 8367 (2003).
- Ferrari, C. P. et al. Snow-to-air exchanges of mercury in an Arctic seasonal snow pack in Ny-Ålesund, Svalbard. *Atmos. Environ.* **39**, 7633–7645 (2005).
- 42. Libois, Q. et al. Influence of grain shape on light penetration in snow. *Cryosphere* **7**, 1803–1818 (2013).
- Mann, E. A. et al. Photoreducible Mercury Loss from Arctic Snow Is Influenced by Temperature and Snow Age. *Environ. Sci. Technol.* 49, 12120–12126 (2015).
- Fraser, A., Dastoor, A. & Ryjkov, A. How important is biomass burning in Canada to mercury contamination? *Atmos. Chem. Phys.* 18, 7263–7286 (2018).
- Jiskra, M., Sonke, J. E., Agnan, Y., Helmig, D. & Obrist, D. Insights from mercury stable isotopes on terrestrial-atmosphere exchange of Hg(0) in the Arctic tundra. *Biogeosciences*. 16, 4051–4064 (2019).
- 46. Obrist, D. et al. Tundra uptake of atmospheric elemental mercury drives Arctic mercury pollution. *Nature* **547**, 201–204 (2017).
- Olson, C. L., Jiskra, M., Sonke, J. E. & Obrist, D. Mercury in tundra vegetation of Alaska: Spatial and temporal dynamics and stable isotope patterns. *Sci. Total Environ.* 660, 1502–1512 (2019).
- Durnford, D. et al. How relevant is the deposition of mercury onto snowpacks?-Part 2: A modeling study. *Atmos. Chem. Phys.* 12, 9251–9274 (2012).
- Comiso, J. C. A rapidly declining perennial sea ice cover in the Arctic. Geophys. Res. Lett. 29, 1956 (2002).
- 50. Comiso, J. C. Large decadal decline of the arctic multiyear ice cover. J. Clim. **25**, 1176–1193 (2012).
- Serreze, M. C. & Barry, R. G. Processes and impacts of Arctic amplification: A research synthesis. *Glob. Planet. Change* 77, 85–96 (2011).
- 52. Rantanen, M. et al. The Arctic has warmed nearly four times faster than the globe since 1979. *Commun. Earth Environ.* **3**, 168 (2022).
- Overland, J. E. et al. Nonlinear response of mid-latitude weather to the changing Arctic. *Nature Clim Change* 6, 992–999 (2016).
- Berner, L. T. et al. Summer warming explains widespread but not uniform greening in the Arctic tundra biome. *Nat. Commun.* 11, 4621 (2020).
- Jia, G. J., Epstein, H. E. & Walker, D. A. Greening of arctic Alaska, 1981-2001. *Geophys. Res. Lett.* **30**, 2067 (2003).
- Forbes, B. C., Fauria, M. M. & Zetterberg, P. Russian Arctic warming and 'greening' are closely tracked by tundra shrub willows. *Glob. Chang. Biol.* 16, 1542–1554 (2010).
- 57. Zhang, Y. et al. Global health effects of future atmospheric mercury emissions. *Nat. Commun.* **12**, 3035 (2021).
- Zhang, Y. et al. A Coupled Global Atmosphere-Ocean Model for Air-Sea Exchange of Mercury: Insights into Wet Deposition and Atmospheric Redox Chemistry. *Environ. Sci. Technol.* 53, 5052–5061 (2019).

Article

- Horowitz, H. M. et al. A new mechanism for atmospheric mercury redox chemistry: Implications for the global mercury budget. *Atmos. Chem. Phys.* 17, 6353–6371 (2017).
- Zhang, Y., Soerensen, A. L., Schartup, A. T. & Sunderland, E. M. A Global Model for Methylmercury Formation and Uptake at the Base of Marine Food Webs. *Global Biogeochem. Cycles* 34, e2019GB006348 (2020).
- 61. Smith-Downey, N. V., Sunderland, E. M. & Jacob, D. J. Anthropogenic impacts on global storage and emissions of mercury from terrestrial soils: Insights from a new global model. *J. Geophys. Res. Biogeosci.* **115**, G03008 (2010).
- 62. Xing, Z. et al. International trade shapes global mercury-related health impacts. *PNAS Nexus* **2**, 128 (2023).
- 63. Shah, V. et al. Improved Mechanistic Model of the Atmospheric Redox Chemistry of Mercury. *Environ. Sci. Technol.* **55**, 14445–14456 (2021).
- Soerensen, A. L. et al. An improved global model for air-sea exchange of mercury: High concentrations over the North Atlantic. *Environ. Sci. Technol.* 44, 8574–8580 (2010).
- Forget, G. et al. ECCO version 4: an integrated framework for nonlinear inverse modeling and global ocean state estimation. *Geosci. Model Dev.* 8, 3071–3104 (2015).
- Hibler, W. D. A Dynamic Thermodynamic Sea Ice Model. J. Phys. Oceanogr. 9, 815–846 (1979).
- Zhang, J. & Hibler, W. D. III On an efficient numerical method for modeling sea ice dynamics. *J. Geophys. Res. Ocean.* **102**, 8691–8702 (1997).
- Mann, E. A., Mallory, M. L., Ziegler, S. E., Tordon, R. & O'Driscoll, N. J. Mercury in Arctic snow: Quantifying the kinetics of photochemical oxidation and reduction. *Sci. Total Environ.* **509–510**, 115–132 (2015).
- Wang, Y. Q. An Open Source Software Suite for Multi-Dimensional Meteorological Data Computation and Visualisation. J. Open Res. Softw. 7, 21 (2019).
- Stein, A. F. et al. NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System. *Bull. Amer. Meteor. Soc.* 96, 2059–2077 (2015).
- Kirk, J. L. et al. Methylated mercury species in marine waters of the Canadian high and sub arctic. *Environ. Sci. Technol.* 42, 8367–8373 (2008).
- Kohler, S. G. et al. Distribution pattern of mercury in northern Barents Sea and Eurasian Basin surface sediment. *Mar. Pollut. Bull.* 185, 114272 (2022).
- 73. Petrova, M. V. et al. Mercury species export from the Arctic to the Atlantic Ocean. *Mar. Chem.* **225**, 103855 (2020).
- Sommar, J., Andersson, M. E. & Jacobi, H. W. Circumpolar measurements of speciated mercury, ozone and carbon monoxide in the boundary layer of the Arctic Ocean. *Atmos. Chem. Phys.* **10**, 5031–5045 (2010).
- Tesán Onrubia, J. A. et al. Mercury Export Flux in the Arctic Ocean Estimated from 234Th/238U Disequilibria. ACS Earth Sp. Chem. 4, 795–801 (2020).
- Yu, J. et al. High variability of atmospheric mercury in the summertime boundary layer through the central Arctic Ocean. *Sci. Rep.* 4, 1–7 (2014).
- Zhang, Y., Jaeglé, L., Thompson, L. & Streets, D. G. Six centuries of changing oceanic mercury. *Global Biogeochem. Cycles* 28, 1251–1261 (2014).
- Gustin, M. S. et al. Observations of the chemistry and concentrations of reactive Hg at locations with different ambient air chemistry. Sci. Total Environ. **904**, 166184 (2023).

- 79. McGillis, W. R., Edson, J. B., Hare, J. E. & Fairall, C. W. Direct covariance air-sea CO2 fluxes. *J. Geophys. Res. Ocean.* **106**, 16729–16745 (2001).
- Liss, P. S. & Merlivat, L. Air-Sea Gas Exchange Rates: Introduction and Synthesis. *Role Air Sea Exchange Geochem. Cycl.* 185, 113–127 (1986).
- Nightingale, P. D. et al. In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers. *Global Biogeochem. Cycles* 14, 373–387 (2000).
- 82. Osterwalder, S. et al. Critical Observations of Gaseous Elemental Mercury Air-Sea Exchange. *Global Biogeochem. Cycles* **35**, e2020GB006742 (2021).
- 83. ECCO Consortium et al. Synopsis of the ECCO Central Production Global Ocean and Sea-Ice State Estimate (Version 4 Release 4). https://doi.org/10.5281/zenodo.4533349 (2021).
- Yuan, T. et al. Potential decoupling of CO2 and Hg uptake process by global vegetation in the 21st century. *Nat. Commun.* 15, 4490 (2024).
- 85. Muntean, M. et al. Evaluating EDGARv4.tox2 speciated mercury emissions ex-post scenarios and their impacts on modelled global and regional wet deposition patterns. *Atmos. Environ.* **184**, 56–68 (2018).
- Torres-Rodriguez, N. et al. Mercury fluxes from hydrothermal venting at mid-ocean ridges constrained by measurements. *Nat. Geosci.* 17, 51–57 (2024).
- 87. Angot, H. et al. Year-round trace gas measurements in the central Arctic during the MOSAiC expedition. *Sci. Data* **9**, 723 (2022).
- Huang, S. Fully-coupled atmosphere, land, sea-ice, and ocean model for Mercury. Available at: http://www.ebmg.online/ mercury/ (2025).
- 89. Schlitzer, Reiner, Ocean Data View (2021). Available at: odv.awi.de.

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Author contributions

S.H., Y.Z., T.Y., and Z.S. conceptualized the study. Methodology was developed by S.H., Y.Z., P.W., and R.C. The investigation was carried out by S.H., T.Y., Z.S., R.C., D.P., P.Z., L.L., and Y.Z. Visualization was performed by S.H., Z.S., and Y.Z. Funding acquisition, project administration, and supervision were managed by Y.Z. The original draft was written by S.H. and Y.Z., with all authors contributing to the review and editing process, including S.H., Y.Z., T.Y., Z.S., L.L., F.W., Z.X., F.Y., and G.Z.

Competing interests

The authors declare no competing interests.

Additional information

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