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Arctic mercury cycling

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68

69

70 **Abstract** Mercury (Hg) levels in Arctic ecosystems, which rose markedly due to global anthropogenic
71 Hg emissions since industrialization, are now being altered as the region warms, with as-yet uncertain
72 ecological consequences. This Review presents a comprehensive assessment of the present-day total Hg
73 mass balance (fluxes and budgets) in the Arctic from a synthesis of published research and atmospheric
74 multi-model simulations, and identifies key future research needs. Arctic atmospheric Hg is primarily
75 emitted outside the region (>98%), and about two-thirds of transported Hg is deposited to terrestrial

76 ecosystems, where it predominantly accumulates in soils via vegetation uptake. Rivers and coastal erosion
77 annually transfer about equal amounts of terrestrial Hg (primarily from above 60° N) to the Arctic Ocean,
78 in approximate balance with modeled net terrestrial Hg deposition in the region, though air-land Hg
79 exchange estimates need further refinement. The revised Arctic Ocean Hg mass balance suggests net
80 atmospheric Hg deposition to the ocean and that Hg burial in inner-shelf sediments is underestimated (up
81 to >100%) and atmosphere-seawater-sediment Hg exchanges are poorly constrained. Specific terrestrial
82 Hg mobilization pathways from the cryosphere (permafrost, ice, snow, and glaciers) remain largely
83 uncertain, and need addressing to anticipate present and future climate impacts on downstream Arctic
84 ecosystems.

85 86 87 **Key points** 88 89

- 90 1. Arctic terrestrial mercury (Hg) emissions from anthropogenic activities (14 Mg y^{-1} ; <1% of
91 global share), wildfires ($8.8 \pm 6.4 \text{ Mg y}^{-1}$), and soil/vegetation re-volatilization ($24(7-59) \text{ Mg y}^{-1}$)
92 are low compared to deposition ($118 \pm 20 \text{ Mg y}^{-1}$). Primarily driven by springtime photochemistry,
93 Arctic Ocean Hg deposition ($65 \pm 20 \text{ Mg y}^{-1}$) exceeds evasion ($32(23-45) \text{ Mg y}^{-1}$), which peaks
94 when snow/ice melt and sea ice retreats.
- 95 2. Large pools of Hg ($\sim 597,000 \text{ Mg}$, 0-3 m depth) have accumulated in permafrost soils,
96 predominantly from vegetation uptake of atmospheric gaseous elemental Hg over millennia.
97 Coastal erosion mobilizes an estimated $39(18-52) \text{ Mg y}^{-1}$ of soil-bound Hg into the Arctic Ocean.
98
- 99 3. Pan-Arctic rivers export $41 \pm 4 \text{ Mg y}^{-1}$ of dissolved and particulate Hg ($\sim 50\%$ each) to the Arctic
100 Ocean predominantly during the spring freshet, likely derived from seasonal snowpacks ($\leq 50\%$)
101 and active-layer surface soils ($\geq 50\%$) of the watershed portion north of 60° N , but specific
102 mobilization pathways are unclear.
103
- 104 4. Revised Arctic Ocean Hg budget ($\sim 1870 \text{ Mg}$) is lower than previous estimates ($2847-7920 \text{ Mg}$),
105 and implies higher sensitivity to changes in climate and emissions. Shelf region particulate Hg
106 settling ($122 \pm 55 \text{ Mg y}^{-1}$) from surface waters is the largest Hg removal mechanism in the ocean,
107 but its annual observations are currently lacking.
108
- 109 5. Combined observation and modeling estimates suggest a balance of Hg mass fluxes in and out of
110 Arctic soils, but air-terrestrial Hg exchange fluxes need further improvement. The revised Arctic
111 Ocean Hg mass balance suggests that Hg burial in shelf sediments ($42 \pm 31 \text{ Mg y}^{-1}$) is
112 underestimated by up to $52.2 \pm 43.5 \text{ Mg y}^{-1}$.
113
- 114 6. Permafrost thaw leads to high concentrations of soil-derived Hg in some streams (up to 1270 ng
115 L^{-1}), but its future impact is presently uncertain. Melt releases $\sim 0.4 \text{ Mg y}^{-1}$ of deposited Hg stored
116 in Arctic glaciers ($2,415 \text{ Mg}$), which is dwarfed by $\sim 40 \text{ Mg y}^{-1}$ of geogenic particulate Hg
117 exported by glacial rivers into adjacent seas.
118
119

120 **Introduction**

121 Mercury (Hg) is a neurotoxic pollutant emitted by both natural and anthropogenic sources, and dispersed
122 globally via atmospheric transport, ocean currents, and rivers. Observations over the past 40 years show
123 that Hg concentrations in Arctic marine mammals and seabirds are elevated compared to lower latitudes
124 ^{1,2}. As a result, Arctic Peoples, who harvest and consume these animals in their traditional diets, are
125 disproportionately exposed to Hg ^{3,4}.

126 Local anthropogenic Hg emissions are negligible in the Arctic, and Hg inputs to Arctic ecosystems are
127 thus largely driven by global Hg emissions ^{1,5}. The existing reservoirs of Hg in Arctic air, soil, snow, ice
128 and water are closely interlinked ^{6,7} (Box 1). Chemical transformations between key Hg species (gaseous
129 and dissolved elemental Hg (Hg(0)), oxidized divalent Hg (Hg(II)) and methylmercury (MeHg)), drive
130 continuous exchanges of these species between reservoirs, influenced by sunlight, organic matter,
131 biological activity, and other key parameters and processes ⁸⁻¹⁰. Climate warming, which is magnified in
132 polar regions ^{11,12}, is anticipated to continue enhancing Hg mobilization due to higher surface
133 temperatures, intensified wildfires ¹³, permafrost thaw ^{14,15}, glacier ¹⁶⁻¹⁸ and sea ice melt ¹⁹, and increased
134 river discharge.

135 The Arctic is a remote region with a paucity of historical Hg observations. However, in recent decades
136 major research initiatives have expanded the spatio-temporal scope of measurements ¹⁹⁻²⁴, used new
137 techniques such as stable Hg isotopes ²⁵⁻²⁹, elucidated redox chemistry ^{30,31} and undertaken process-based
138 modeling ^{6,32-35}. This has greatly improved our understanding of Hg fluxes and processes in the Arctic,
139 underpinning the revision of its mass balance described here. A few global Hg reviews or assessments of
140 different topics ³⁶⁻³⁸ have included some Arctic Hg data in their syntheses. This review represents the first
141 comprehensive re-examination of the Arctic Hg cycle and mass balance since 2011 ¹.

142 In this Review, an updated understanding of the total Hg mass balance in the Arctic is developed from
143 literature synthesis and modeling conducted for the Arctic Monitoring and Assessment Programme
144 (AMAP) 2021 Hg Assessment. Other parts of the AMAP 2021 Assessment concerned Hg
145 methylation/demethylation, and Hg concentrations and trends in biota, which are not covered here. Arctic
146 mercury fluxes (emissions and depositions, oceanic inflow/outflow, particle settling and sediment burial,
147 river export, coastal erosion, sea ice export) and reservoir mass budgets (atmosphere, permafrost soils,
148 seasonal snowpacks, glaciers, sea ice, and ocean) are developed for the land area north of 60° N and the
149 Arctic Ocean (AO) - defined as the central basin and the Barents, Kara, Laptev, East Siberian, Chukchi
150 and Beaufort Seas (Fig. S1). The significance of these estimated fluxes to Hg cycling in Arctic
151 ecosystems is examined, and future research areas are prioritized based on current uncertainties. This

152 Review benchmarks contemporary total Hg pathways and levels in the Arctic, and supports the
153 effectiveness evaluation objective of the *Minamata Convention on Mercury*.

154
155 **Atmospheric mercury**

156
157 Each year 6,000-9,000 Mg of Hg is emitted to the atmosphere⁴⁴ through anthropogenic activities (2,000–
158 3,000 Mg)^{39,40}, biomass burning (400-700 Mg)⁴¹⁻⁴³, and geogenic degassing and legacy emissions from
159 land (1,000–1,600 Mg)³⁷ and oceans (2,700–3,400 Mg)³⁷, mainly as Hg(0) (0.5-1 year lifetime) with some
160 as Hg(II) (1-2 weeks lifetime). Atmospheric Hg is transported to the Arctic from global sources, driven by
161 synoptic-scale pressure systems. Hg transport is enhanced from northern Eurasian sources in winter,
162 influenced by the Arctic polar dome, and from mid-latitude Asian and North American sources in spring,
163 facilitated by convective lifting⁴⁴⁻⁴⁸.

164 Changes in air circulation patterns due to warmer temperatures and lower sea ice concentrations with a
165 warming climate^{49,50} are impacting contaminant transport to and within the Arctic⁵¹ including Hg
166 transport and deposition^{5,35,52}. Following sections describe atmospheric Hg emission sources, transport
167 and deposition pathways, and concentrations and deposition patterns in the Arctic.

168 ***Anthropogenic and wildfire emissions***

169 Less than 1% of estimated global annual anthropogenic Hg emissions to air (~14 Mg, 2015) are emitted
170 from sources within the Arctic^{39,53}. Main sources of Hg emissions in the Arctic are point sources in
171 northern Russia, including non-ferrous metal smelters and coal-fired power plants⁵³. Other sources of
172 anthropogenic Hg emissions in the Arctic include mining, oil and gas activities, and uncontrolled disposal
173 of waste that might contain Hg-added products. The methodological assumptions made in estimating
174 anthropogenic Hg emissions and their geospatial distribution, especially from non-point sources in areas
175 of sparse population, mean that they are subject to large uncertainty.

176 In the Arctic, Hg emissions from open biomass burning are primarily related to natural (wildfire) sources,
177 predominantly boreal forest fires. Hg in biomass is almost exclusively derived from atmospheric
178 deposition and is released through the combustion of living and ground litter, and soil heating during
179 wildfires^{54,55}. Annual Hg emissions estimates from open biomass burning vary widely, ranging from ~20
180 Mg^{54,56} to 200 Mg⁴², with large uncertainties, particularly for boreal forest fires. The uncertainty is
181 associated with high interannual variability in the burned area of boreal forests and use of non-biome
182 specific Hg emissions ratios (enhanced concentration ratio of Hg and a co-pollutant) or emissions factors
183 (Hg mass emitted per area burned) with assumed carbon species emission enhancements⁴¹⁻⁴³. Further,
184 these empirical emission ratios or factors are often measured at sites distant from fire sources, which

185 likely lead to overestimation of Hg emissions due to the shorter atmospheric lifetime of carbon monoxide
186 (CO) and variability in CO emissions based on vegetation-type and fire intensity⁵⁶. Following the
187 empirical emission factor-based method (EEM3)⁵⁶, an improved mean atmospheric Hg emission from
188 Arctic fires (>60°N) is estimated as 8.8±6.4 Mg y⁻¹ for the period 2001-2019. This emission estimate uses
189 the mean emission factor from two near source boreal forest aircraft studies^{54,56}, mean burned area based
190 on three separate algorithms⁵⁷⁻⁵⁹, mean total fuel consumption (including soil carbon releases) of
191 Canadian Arctic biomes (2.31±0.81 kg m⁻²)⁶⁰, complete (100±5%)^{54,56} release of Hg during fires, and
192 atmospheric Hg fractions of 96.2% elemental and 3.8% particulate.

193 The current push for natural resource development in the Arctic (oil and gas exploration, mining) could
194 lead to greater anthropogenic Hg releases within the region. Additionally, human development in the
195 Arctic is projected to increase due to climate change, raising the potential for increasing anthropogenic
196 Hg sources. Boreal peatlands are estimated to contain ten times greater Hg stocks than in boreal forests
197 due to a thicker organic layer⁶¹, and likely release more Hg during fires. The Hg speciation of peat fire
198 emissions might differ due to more dominant smoldering (vs. flaming) combustion⁶². Global fire
199 emissions databases indicate a larger increasing trend in wildfires (frequency, intensity, and burning
200 season length) north of 60° between 2005-2018 than 50-60° N, partly attributed to human activities⁶³⁻⁶⁶.

201 *Air-surface Hg exchange*

202 Hg exchange between the Earth's surface and the atmosphere occurs via several pathways⁶⁷: Hg(0)
203 exchange with vegetation, soils, snow, ice and waters; and atmospheric oxidation of Hg(0) and
204 subsequent wet and dry Hg(II) deposition in gaseous or particulate phase. Vegetation uptake of
205 atmospheric Hg(0) is the dominant deposition pathway in vegetated Arctic ecosystems (~70% of total
206 deposition), when compared to the Hg(II) deposition^{37,68}. Hg oxidation and deposition processes intensify
207 during springtime (known as atmospheric mercury depletion events (AMDEs)) in coastal and marine
208 Arctic environments⁶⁹, facilitated by excessive photochemical release of gaseous bromine to air from
209 snowpacks on sea ice, wind-blown snow particles and sea-salt aerosols⁷⁰⁻⁷³. However, 40-90% of Hg
210 deposited during AMDEs is photo-reduced and reemitted⁶⁶. Hg(0) evasion rates are low in the interior
211 tundra in all seasons^{68,74} and high from marine waters during spring/summer sea ice retreat¹⁹.

212
213 For this Review, the contemporary atmospheric Hg cycle (year 2015) was simulated (Fig. 1) using an
214 ensemble of Hg models^{33,75} (GLEMOS⁷⁶, GEOS-Chem^{34,77}, GEM-MACH-Hg^{52,78-80}, DEHM^{81,82}; see
215 details of the models in the Supplementary Information Text S1, Table S1). Arctic atmospheric mercury
216 concentrations are shaped by transport patterns from lower latitudes and deposition processes in the

217 Arctic¹. Air Hg(0) concentrations in the Arctic (1.4 ng m⁻³, modeled and measured⁸³⁻⁸⁵ domain annual
218 average) are characterized by a latitudinal gradient with concentrations greater than 1.4 ng m⁻³ occurring
219 across most of the terrestrial Arctic and concentrations less than 1.4 ng m⁻³ over the Canadian Arctic
220 Archipelago (CAA), Greenland and the AO (Fig. 1a). In the Arctic, the strongest Hg(0) seasonal cycle is
221 observed in coastal and marine regions (1.1-1.6 ng m⁻³, monthly mean range)³³. Minimum Hg(0)
222 concentrations, driven by AMDEs, are observed in spring, and maximum concentrations occur in summer
223 that are attributed to snow and sea ice melt and oceanic Hg re-emissions^{33,78,86}. Total oxidized Hg
224 concentrations at high Arctic coastal sites are an order of magnitude higher in springtime (>150 pg m⁻³, 10
225 years mean observed at Alert, Canada) than the remainder of the year (<25 pg m⁻³, Alert)⁸⁷. Arctic
226 atmospheric Hg levels are less dynamic in fall and winter, and primarily reflect northern hemispheric
227 background Hg⁸⁶. The models accurately simulate the seasonal cycle of atmospheric mercury species³³,
228 but underestimate the amplitude of the seasonal variation due to a lack of real-time mechanistic modeling
229 of the production of bromine species and sea ice dynamics^{78,88}.

230

231 The Arctic is characterized by low wet deposition fluxes (<5 µg m⁻² y⁻¹, modeled; <4 µg m⁻² y⁻¹,
232 observed^{76,89}), especially in arid areas of Greenland, the CAA, and Siberia, compared to lower latitudes
233 (up to 30 µg m⁻² y⁻¹)⁹⁰⁻⁹² (Fig. 1b). While precipitation Hg concentrations in the Arctic are low most of the
234 year, springtime snowfall Hg concentrations can be anomalously high during AMDEs, especially in
235 coastal and marine locations (for example, median total Hg snowfall concentrations of 79-388 ng L⁻¹ in
236 March, Utqiagvik, Alaska)⁹³. Models overestimate Hg wet deposition flux by up to 100% compared to
237 observations^{8-10,94}. However, measured Hg wet deposition fluxes are likely significantly underestimated in
238 polar regions due to measurement challenges. Wet deposition collectors have lower collection efficiencies
239 for snow than the recommended precipitation gauges (by ~30%)⁹⁵. However, precipitation gauges also
240 underestimate snowfall amounts by 20-50% in windy environments⁹⁶⁻⁹⁸, and are subject to the
241 uncertainties of measuring trace precipitation prevalent in the Arctic^{99,100}.

242

243 Model ensemble annual average total Hg deposition flux rates in the Arctic are 6.8±1.2 µg m⁻² y⁻¹ (5.2±1.2
244 µg m⁻² y⁻¹, north of 66.5° N) over land and 7.4±1.6 µg m⁻² y⁻¹ over the AO with about half of seasonal
245 contributions occurring in summer over land and in spring over ocean (Table S2; Fig. 1c). Highest
246 modeled Hg deposition rates (up to 20 µg m⁻² y⁻¹) occur in the regions characterized by: presence of local
247 Hg emissions (Fennoscandia); efficient trans-Pacific transport (northwestern North America); high net
248 primary productivity (boreal forests); and higher precipitation amounts (ocean around Greenland, and
249 coastal northwestern Europe and North America) (Fig. 1c). Over land, Hg deposition rates decline from
250 south to north and west to east in Eurasia and North America, driven by proximity to Hg emission

251 sources. Compared to the measured⁶⁸ Hg deposition flux of $9.2 \mu\text{g m}^{-2} \text{y}^{-1}$ at a tundra (Alaska) site, the
252 model ensemble-estimated deposition flux is $7.2 \pm 2.2 \mu\text{g m}^{-2} \text{y}^{-1}$ (~22% lower). GEM-MACH-Hg model-
253 simulated median dry deposition Hg flux in boreal forests ($8.3 (3.5\text{--}15.1) \mu\text{g m}^{-2} \text{y}^{-1}$)³⁷ is consistent with
254 the litterfall-based estimate ($8.6 (1.3\text{--}19.8 \mu\text{g m}^{-2} \text{y}^{-1})$)⁹⁷, but underestimated in Arctic tundra ($4.2 (1.1\text{--}$
255 $8.8)$)⁴⁴ $\mu\text{g m}^{-2} \text{y}^{-1}$, model; $6.0 (1.4\text{--}17)$)¹⁰¹ $\mu\text{g m}^{-2} \text{y}^{-1}$, litterfall-based). Modeled deposition compares well
256 with lake sediment-inferred average Hg deposition rates in the Canadian high Arctic^{102,103} and sub-
257 Arctic^{102,104,105} of $3.5 \mu\text{g m}^{-2} \text{y}^{-1}$ (~ $3.6 \mu\text{g m}^{-2} \text{y}^{-1}$ ($0.5\text{--}5 \mu\text{g m}^{-2} \text{y}^{-1}$), modeled) and $7.5\text{--}10.0 \mu\text{g m}^{-2} \text{y}^{-1}$
258 ($\sim 7.65 \mu\text{g m}^{-2} \text{y}^{-1}$ ($5\text{--}10 \mu\text{g m}^{-2} \text{y}^{-1}$), modeled), respectively. Model ensemble wintertime (September-May)
259 deposition is also consistent with observation-based^{29,106,107} deposition at the three high Arctic sites ($\sim 2\text{--}4$
260 $\mu\text{g m}^{-2}$).

261 Model ensemble estimates Arctic atmospheric total Hg burden of 330 Mg (290-360 Mg, seasonal
262 variance), and total deposition of $243 \pm 41 \text{ Mg y}^{-1}$ (~51% deposited to the ocean) north of 60°N . Model
263 ensemble total deposition estimate of $133 \pm 31 \text{ Mg y}^{-1}$ north 66°N in 2015 is in line with the literature
264 range of $110\text{--}131 \text{ Mg y}^{-1}$ ^{76,78,81}. Modeling estimates of Hg(0) evasion flux from soils and vegetation is 24
265 ($7\text{--}59$) Mg y^{-1} (model ensemble), and from the AO are 23.3 (GEM-MACH-Hg) and 45.0 Mg y^{-1} (GEOS-
266 Chem). Air-sea Hg exchange fluxes are highly dynamic in spring and summer as a result of AMDEs¹⁰⁸,
267 snow/sea ice melt and river discharge^{34-36,80}, but open ocean measurements representing all seasons are
268 lacking to confirm model estimated marine Hg fluxes^{19,109,110}.

269 Long-term Hg observations at high Arctic sites show seasonally variable and overall neutral (Ny-Ålesund,
270 Svalbard)¹¹¹ to decreasing ($\leq 1.0\%$ per year, Alert⁸³ and Villum Research Station, Greenland⁸²) air
271 concentrations of Hg(0), and increasing springtime Hg(II) concentrations ($9\text{--}17\%$ per year, Alert⁸³).
272 Climate warming led changes in AMDEs-related oxidation chemistry⁸³, air-surface Hg exchanges^{5,52} and
273 increasing precipitation⁵² with consequences to Hg deposition in the Arctic are suggested to be
274 responsible for the changing atmospheric mercury cycling. Vegetation Hg deposition is projected to
275 increase with increasing vegetation cover and density^{32,112}, but Hg evasion from soils is projected to
276 increase in response to accelerating wildfires and permafrost thaw-led microbial reduction and release of
277 stored Hg in soils¹⁵.

278 Current and projected future Arctic warming trends¹¹³ towards an increase in thinner first year sea ice,
279 more open sea ice leads, a longer open water season, and increased ice dynamics, might favor conditions
280 that stimulate AMDE-related Hg deposition by enhancing reactive halogen sources. A 22-year (1996-
281 2017) trend analysis of Arctic atmospheric bromine monoxide (BrO) satellite measurements¹¹⁴ found
282 moderate spatiotemporal relationship ($r = 0.32$, $p < 0.05$) between springtime BrO concentrations and the

283 first-year sea ice extent in the AO, both having increased over this period. Higher temperature is
284 suggested to decrease the Hg oxidation via Br-initiated reactions owing to the thermal dissociation of
285 HgBr^{115,116}, but the transformation of HgBr to BrHgO via ozone reaction likely dominates over the
286 thermal dissociation¹¹⁷ resulting in weaker temperature dependence of Hg oxidation. Convective mixing
287 over more prevalent open leads can increase Hg(0) supply in near-surface air to be oxidized and deposited
288 to surrounding snowpacks^{118,119}.

289 **Terrestrial mercury**

290
291 The terrestrial system plays a dual role in Arctic Hg cycling as a sink of atmospheric Hg and as a source
292 of Hg to surface waters⁷. The importance of terrestrial Hg exports to the AO is supported by soil-like Hg
293 stable isotope signatures in marine sediments and biota^{120,121}. Hg lifetime in Arctic terrestrial reservoirs
294 ranges between ~10³ (soils), 10²-10³(glaciers) and 1 (snowpack) year, which might be reduced in future
295 due to Arctic warming led large perturbations of the terrestrial system³⁸. In the following, we discuss Hg
296 distribution and storage in terrestrial compartments (vegetation, soils, snowpacks and glaciers) and
297 transfer of terrestrial Hg to the AO via riverine transport and coastal erosion.

298 ***Vegetation Hg***

299
300
301 Vegetation uptake of atmospheric Hg(0) is a dominant source of Hg in vegetation^{68,122} and soils^{25,122}
302 (through transfer when plants die) in the Arctic tundra. Pronounced Hg concentration differences exist
303 both within and among different vegetation functional groups (lichen, mosses, and vascular plants) in the
304 Arctic (Table S3): lichen (mean: 62±41 ng g⁻¹) and mosses (mean: 61±39 ng g⁻¹) generally show highest
305 Hg concentrations, and lower Hg levels have been reported in vascular plants (mean: 10±5 ng g⁻¹). Arctic
306 vascular plants show about three-times lower Hg concentrations than globally averaged vascular foliar
307 (leaves and needles) concentrations (mean: 34±21 ng g⁻¹)¹²³. Arctic vegetation Hg data are still relatively
308 sparse and large uncertainties exist in regard to spatiotemporal variability. Cryptogamic vegetation
309 (which include lichen and mosses) show substantially higher Hg concentrations than vascular plants, and
310 high variability is reported in Arctic lichen Hg concentrations (19–186 ng g⁻¹) (Table S3) due to
311 differing deposition sources and pathways¹²⁴. Lichens, as important forage substrates, can be potentially
312 important Hg sources in caribou and other Arctic herbivores¹²⁴. Hg concentrations ranging from 20-195
313 ng g⁻¹ are reported^{125,126} in mosses. Lichen and mosses have a large representation in Arctic vegetation
314 communities, which leads to relatively high bulk vegetation Hg concentrations (mean: 49±37 ng g⁻¹) in
315 the Arctic¹²² (Table S3). One study¹²² calculated tundra to contain aboveground standing Hg biomass

316 pools of $29 \mu\text{g m}^{-2}$ and a boreal forest study¹²⁷ showed a foliar Hg pool of $21 \mu\text{g m}^{-2}$ (Pallas, Finland).
317 These northern standing biomass Hg pools are surprisingly similar in magnitude to foliar Hg biomass of
318 lower-latitude forests with much longer growing seasons ($15\text{-}45 \mu\text{g m}^{-2}$)^{68,122,127}.

319 **Soil Hg**

320 Arctic soils, a globally important Hg reservoir, have accumulated atmospheric Hg over millennia with
321 anthropogenic Hg mostly confined to the surface layer¹³⁰. Soil Hg can be released to surface waters via
322 river runoff and coastal erosion or re-emitted to the atmosphere^{68,128,129}. Higher Hg concentrations in
323 active-layer (seasonally thawing and freezing) surface soils (65 ng g^{-1} , median) than deeper mineral
324 (permafrost) horizons (48 ng g^{-1} , median) are primarily reported from soil profiles in Alaska¹³⁰,
325 Svalbard¹³¹ and the Western Siberian Lowlands¹²⁹ (Fig. 2a-b, Table S4). The Hg enrichment of surface
326 soils highlights atmospheric Hg deposition as a major source^{25,68,122}. Hg stable isotopes reveal that
327 atmospheric Hg(0) vegetation uptake is the major deposition pathway in the Arctic Coastal Plain of
328 Alaska, accounting for 70% (56-81%, interquartile range (IQR)) of Hg in organic surface O-horizons,
329 54% (43-62%, IQR) in A mineral horizons, and 24% (14-34%, IQR) in B horizons^{25,68}. Geogenic source
330 contributes 20% (A horizons) to 40% (B horizons)^{25,68} Hg in mineral soils of Alaska. Oxidized Hg
331 deposition during AMDEs is a minor contribution (<5%) to all soil horizons^{68,130}.

332
333 The Northern Hemisphere permafrost region 0-1 m soil Hg pool has been estimated to be 184 Gg (136–
334 274 Gg, 25% confidence interval (CI))¹³⁰, 240 Gg (110–336 Gg, IQR)¹²⁹, and 755 ± 301 Gg (95% CI)¹²⁸.
335 Along a Western Siberian Lowland transect, the 0-1 m soil Hg pool increases with latitude from 0.8 mg
336 m^{-2} (56° N) to 13.7 mg m^{-2} (67° N), suggesting significant re-emission losses of soil Hg in the sub-
337 Arctic¹²⁹. The 0-3 m soil Hg pool estimates range from 597 Gg (384–750 Gg, IQR)¹²⁹ to 1656 ± 962 Gg
338 (95% CI)¹²⁸. Alaska observations represent the largest share (50%) of the soil Hg data collected, but only
339 cover 7% of the Northern Hemisphere permafrost region (Fig. 2a-b). To overcome the lack of spatially
340 distributed Hg soil data, studies extrapolate limited observations of mercury to carbon ratios (Hg/C) in
341 Arctic soils¹²⁸⁻¹³⁰ to the entire Arctic region using high-resolution soil carbon maps¹³². The highest
342 permafrost soil Hg estimate¹²⁸ is likely biased high; the application of elevated deeper mineral permafrost
343 soil Hg/C ratio ($1,600 \text{ ng g}^{-1}$) observed in northern Alaska (due to higher geogenic Hg levels) than
344 elsewhere^{68,130} to the entire Arctic¹²⁸ led to systematic overestimation of organic surface soils Hg
345 content^{129,130}. Best estimates for the Arctic soil Hg pool are ~49 Gg for 0-0.3 m (surface soils), ~212 Gg
346 for 0–1 m (active layer soils), and ~597 Gg for 0–3 m, averaging the two Arctic soil Hg pool inventories
347 based on whole-Arctic literature surveys and western Siberia data^{129,130}.

348

349 *Snowpack Hg*

350 With winter lasting about two-thirds of the year (September-May), the snowpack is a major substrate for
351 atmospheric Hg exchange with Arctic ecosystems¹. Unique physical and chemical cryospheric processes
352 enrich Hg concentrations in polar snow (1-1,000 ng L⁻¹ in Arctic snow; Table S7)¹³³ compared to lower
353 latitude locations (1~40 ng L⁻¹)¹³⁴⁻¹³⁷. Frost flowers on sea ice^{119,138,139}, surface hoar crystals¹⁴⁰, and
354 diamond dust¹⁴¹ have elevated Hg concentrations (commonly 50-200 ng L⁻¹ with values up to 1,200 ng L⁻¹)¹⁴¹. Sea ice is a major source of reactive halogens to snowpacks and ambient air^{70,142} that oxidize Hg(0)
355 in air during AMDEs and can stabilize Hg in snow^{106,108,133}. Hg stabilization in snow can arise from
356 suppressed Hg(II) photo-reduction¹⁴³ and enhanced Hg(0) re-oxidation¹⁴⁴ by halogens in snowpacks,
357 including in interstitial air⁸⁸ and at the stage of snowmelt^{106,143}. Consequently, higher Hg concentrations
358 are measured in snow at coastal locations (5~200 ng L⁻¹; Svalbard, Alert, Utqiagvik, and
359 Greenland)^{86,106,108,107,145,146} than inland sites (0.5-5 ng L⁻¹; Toolik Lake, Alaska, Mt. Oxford and Agassiz,
360 Canada, and Summit Station, Greenland)^{68,74,147}.

362 A large fraction (40-90%)¹³³ of snowpack Hg is re-emitted by photo-reduction, which depends on factors
363 like halide and particulate matter content of snow, snowfall frequency, snow temperature, solar radiation,
364 snowpack ventilation and upward latent heat flux^{133,146,148-151}. However, due to relatively elevated
365 snowpack Hg deposition rates in otherwise pristine locations a significant amount of Hg is retained,
366 especially in coastal and marine regions^{80,124}. A snowpack study from interior Alaska showed that the
367 tundra snowpack is less prone to Hg(0) emissions than previously estimated⁷⁴. During the spring freshet,
368 Hg accumulated in snow and ice is: re-emitted during snow metamorphism and melt, exported to
369 freshwaters or the marine system (40-80%, coastal Alaska), or incorporated into ecosystems by sorption
370 to soil or vegetation surfaces¹⁰⁶. Early snow melt includes an “ionic” pulse of major elements and elevated
371 Hg concentrations (3-30 ng L⁻¹)^{106,145}. GEM-MACH-Hg simulation estimated an Arctic terrestrial
372 seasonal snowpack Hg loading of 39 (35–42) Mg, and mean meltwater ionic pulse Hg concentrations that
373 are generally lower in North America (< 10 ng L⁻¹) than in Eurasia (> 20 ng L⁻¹) due to differences in Hg
374 deposition rates⁸⁰.

375 376 *Glacial Hg*

377 Glaciers mobilize Hg (dissolved and particulate) downstream via seasonal thaw and net ice mass wastage.
378 The Hg in glacier-fed streams comes from anthropogenic and natural sources, including atmospheric
379 deposition to snow, and erosion from bedrock. High-latitude glacier ice formed in the 19-20th centuries is
380 enriched in Hg by factors of 2–15 relative to pre-industrial ice^{28,152-154}. Upscaling an ice core-based
381 estimate of the median Hg concentration (0.8 ng L⁻¹)^{28,152-154} and using current estimates of glacier ice

382 volume¹⁵⁵, the mean size of the Arctic glacial Hg pool is estimated at 2415±22 Mg (~97% in Greenland).
383 Based on spaceborne, gravity-based assessments of mass wasting rates^{156,157} and ice-core Hg data, melting
384 Arctic glacier ice releases 400±7 kg y⁻¹ of Hg (~60% from Greenland). While total Hg levels in en- or
385 supra-glacial ice are typically <1 ng L⁻¹, concentrations in sub- or pro-glacial streams fed by subglacial
386 drainage can be 10-100s times larger, depending on catchment geology, owing to high particulate Hg
387 loads (Table S5). A new study¹⁵⁸ reporting dissolved Hg levels of 44-351 ng L⁻¹ in a west Greenland river
388 presently stands as an outlier, calling for further confirmation. Typical Hg concentrations in bulk
389 suspended particles carried by glacial streams are 5 to >500 ng g⁻¹^{16,159-161}, and estimated total Hg yields
390 from most glacier-fed catchments are 0.03-3.88 g km⁻² y⁻¹ in basins with 21-82% glacier coverage, but a
391 few have yields >10 g km⁻² y⁻¹ (Fig. 2d, Table S5). For Greenland, multiplying riverine particulate Hg
392 concentrations¹⁶² by estimated sediment exports yields¹⁶³ a total annual output of ~40 Mg y⁻¹ Hg, far in
393 excess of Hg releases from melting ice. Run off from Greenland mostly discharges into adjacent seas
394 (>90%), with the remainder into the AO. Most glacierized catchments are in Greenland, the CAA and
395 Svalbard¹⁶⁴, and thus outside the large mainland watersheds for which Hg fluxes are monitored^{7,165}.
396 Presently, the limited number of glacial rivers sampled (~10% in Table S5) and wide range of Hg levels
397 and fluxes in these rivers make glacial Hg flux estimates uncertain.

398 ***Riverine Hg***

399 Rivers mobilize and carry Hg from soils, snow, ice and released from permafrost and bedrock to
400 freshwater ecosystems and the oceans. A strong coupling between Hg export and both water discharge
401 and particle loadings suggests that hydrology and soil erosion are key drivers of Arctic riverine Hg
402 export^{7,165,166}. Over 90% of annual Hg export from large Arctic rivers occurs in spring/early summer,
403 attesting to the importance of snowpack melt to riverine Hg^{7,165,167-169} (Fig. 2). Hg:C ratio was observed to
404 be 2-3 times higher in spring waters from western Siberia rivers compared to summer and fall¹⁶⁷,
405 reflecting runoff of wintertime accumulated Hg deposition in snow. During the spring freshet, the ground
406 is still frozen. Percolation through soil is thus not a major Hg source, but surface soil entrainment by
407 overland flow and river bank erosion are¹⁰⁶. Terrestrial contributions increase through summer as the
408 ground thaws, leading to a minor second peak in Hg exports from some rivers during late summer or
409 fall^{165,167}. Mercury in snowmelt and surface soils released during spring likely originates mainly from
410 recent atmospheric deposition, while later releases have larger, as yet unquantified, shares from legacy Hg
411 deposition and natural sources^{68,74,93,130,167}. Groundwater is important for some Arctic rivers (such as
412 Yukon¹⁷⁰), but its magnitude as a Hg source is unknown.

413

414 Local soil permeability, organic carbon content, glacial history, and vegetation cover all affect Hg storage
415 and retention, making permafrost thaw contributions to riverine Hg exports difficult to quantify^{171,172}.
416 About 5% of Hg in Arctic soils is stored in regions with ice-rich permafrost susceptible to developing
417 hillslope thermokarst features, like retrogressive thaw slumps, which can quickly mobilize vast amounts
418 of particulate Hg into rivers¹⁴. Gradual thaw may also enhance particulate Hg export in streams,
419 especially in the sporadic permafrost zone¹⁶⁷.

420 Total Hg concentrations vary by four orders of magnitude across Arctic watercourses (0.17–1,270 ng L⁻¹;
421 Supplementary Table S5). The highest reported Hg levels are in streams affected by retrogressive thaw
422 slumps (max. 1,270 ng L⁻¹)¹⁴, in ephemeral snowmelt-fed streams from coastal Alaska (max. 80 ng L⁻¹
423)^{93,106} where halogens likely drive enhanced Hg oxidation in air and retention in snow^{108,144}, and in turbid
424 glacier-fed rivers (max. 74 ng L⁻¹) during peak discharge^{16,160}. In rivers with large watersheds (>10⁵ km²),
425 Hg concentrations are between 0.4 and 27 ng L⁻¹ (median: 4.80 ng L⁻¹; Supplementary Table S5) with
426 above 10 ng L⁻¹ concentrations reported in northwestern North America (Yukon and Mackenzie
427 watersheds) and Russia (Severnaya Dvina and Katun rivers). While tributaries to the Yukon and
428 Mackenzie rivers are relatively pristine, the Katun River (Ob' watershed; max. 300 ng L⁻¹) flows over Hg-
429 enriched bedrock¹⁷³, and the Severnaya Dvina River (5-21 ng L⁻¹) is locally impacted by Hg
430 contamination around Archangelsk¹⁷⁴. The share of particulate Hg varies between 29 and 60% across
431 most Arctic rivers, but exceeds 85% in some glacier-fed streams^{16,17}.

432
433 Annual riverine Hg yields (average: 2.81 µg m⁻²; median: 1.76 µg m⁻²) in the Arctic exhibit large
434 variations between watersheds (0.03–98.5 µg m⁻²; Table S5), with 68% estimates between 0.9 and 4.4 µg
435 m⁻². Higher river Hg yields are generally observed for rivers in western Eurasia and North America, with
436 lower yields in Greenland, the CAA and western Hudson Bay (Fig. 2d). This pattern is broadly consistent
437 with atmospheric Hg deposition across the Arctic (Figs. 1c and 3c). Watersheds on the AO coast are
438 estimated to have the highest riverine Hg yields, driven by their high AMDE-led snowpack Hg
439 loadings^{7,80,93,106} (Fig. 2d).

440 Comparison between observation-based Hg exports from major Arctic rivers (Yenisei, Lena, Ob',
441 Pechora, Severnaya Dvina, Kolyma, Mackenzie and Yukon draining about 70% of pan-Arctic watershed
442 extending up to 45° N) and modeled (ensemble) Hg deposition reveals that river Hg exports are strongly
443 positively correlated (r = 0.87) with their watershed Hg deposition components north of 60° N (Fig. S2;
444 Table S6). Correspondingly, river Hg yields increase with increasing watershed proportions north of 60°
445 N (Fig. 1d; Table S6). These results suggest greater re-emission¹²⁹ and/or sedimentation occurring of
446 deposited Hg at latitudes south of 60° N. Additionally, river Hg yields are higher from the watersheds
447 with greater Hg deposition rates (> 60° N), driven by proximity to Hg emission and presence of dominant

448 deposition pathways (Figs 1c and 2c; Table S6). The observation-based^{7,165} total Hg export from the eight
449 largest Arctic rivers (23.6 Mg y⁻¹) is about half of the modeled annual Hg deposition (47.6 Mg y⁻¹) and
450 comparable to summertime (25.2 Mg y⁻¹) or snow cover period (22.4 Mg y⁻¹) Hg deposition in these
451 watersheds north of 60° N (Table S6).

452 Observation-based estimates of annual Hg export from pan-Arctic rivers have converged on the range
453 between 37¹⁶⁵ and 44⁷ Mg y⁻¹, with eight major rivers exporting about 50% of the Hg. Much of this Hg is
454 discharged directly to the AO (41±4 Mg y⁻¹), with 2-3 Mg y⁻¹ entering Hudson and Baffin bays. Applying
455 measured snowpack Hg export rate of ~50% (40-80%)¹⁰⁶ to the modeled end of season snowpack Hg
456 loading (39 Mg y⁻¹)⁸⁰ in the Arctic (north of 60° N) suggests that snowpacks supply up to half of the pan-
457 Arctic river Hg export, the balance being derived from active layer surface soils. This finding is in line
458 with observations of roughly equal amounts of dissolved and particulate Hg in river runoff to the ocean⁷.

459
460 Future Arctic riverine Hg exports will be impacted by Arctic warming through accelerated permafrost
461 thaw, glacier melt, increased rainfall, shorter snow cover seasons, and wildfire-related soil erosion.
462 Glacial discharge is forecasted to increase across much of the Arctic until the mid-21st century; Greenland
463 will become the dominant source thereafter, while outflow from smaller glaciers and ice cap will
464 decline^{175-177,178}. These trends will likely be reflected in glacier-fed riverine Hg exports. Permafrost will
465 also likely contribute more Hg to rivers via enhanced water percolation through a deepening active layer,
466 and slump development along stream banks^{167,171,179}. With unconstrained fossil fuel burning, terrestrial Hg
467 model simulations for the Yukon River project a doubling of Hg(II) export by 2100, and a tripling by
468 2200¹⁵. However, thaw processes vary widely across ice-rich and ice-poor permafrost, making pan-Arctic
469 projections more difficult and highlighting the need to quantify thaw-induced Hg releases in different
470 settings. Understanding the sensitivity of small watershed Hg fluxes to change is especially important
471 given their prevalence along Arctic coastlines.

472

473 *Coastal erosion Hg export*

474 Arctic coastal erosion rates are among the highest in the world because of long reaches of unlithified
475 glacial drift in elevated bluffs, rapid sea-level changes, and exposed ground ice susceptible to the action
476 of wind, water, and thermoerosion¹⁸⁰⁻¹⁸². The Hg mass entering the ocean can be estimated from Arctic
477 soil Hg concentrations in permafrost soils including the active layers compiled from the literature (REF
478 ^{130,183,184} and references therein; REF¹²⁹, excluding a southern, non-permafrost site; Table S4), and eroding
479 soil mass based on soil volumes from the Arctic Coastal Dynamics Database¹⁸⁵ (Table S7). Most soil Hg
480 data are from inland tundra sites; additional sampling of coastal soils is needed to confirm their similarity
481 to inland soils. Using the mean soil Hg concentration of 66.1±52.3 ng g⁻¹ dry weight, the estimated annual

482 erosional Hg flux is $39\pm 30 \text{ Mg y}^{-1}$ (median 32 Mg y^{-1} ; $18\text{-}52 \text{ Mg y}^{-1}$, IQR) (Table S7). The Eurasian
483 Arctic, where erosion rates are highest, contributes 89% of the Hg flux. Calculated coastal erosion flux
484 uncertainty only reflects the variability in soil Hg concentrations, not erosion mass.

485 Coastal erosion is increasing in many Arctic areas and is now higher than at any time since observations
486 began 50-60 years ago because of interacting climatic, oceanographic and geomorphological factors^{180,186}.
487 A projected future increase in rates of coastline erosion^{180,185} will contribute more Hg to the ocean.

488 **Marine mercury**

489 The Hg cycle in the AO is the key link between anthropogenic emissions and releases, Arctic marine
490 biota and human exposure. Although efforts to understand the AO Hg cycle were historically hindered by
491 a lack of data¹⁸⁷⁻¹⁹⁰, coordinated efforts (such as GEOTRACES) since 2015 have made the AO one of the
492 most sampled ocean basins with ~2000 total Hg seawater observations^{19-22,191-194} (Fig. 3a).

493 The AO has unique and complex hydrography, distinguishing its Hg cycle from that of other oceans. The
494 AO is covered by sea ice, affecting the air-sea Hg exchange. Almost completely surrounded by land and
495 over half underlain by continental shelf, the AO receives a larger river discharge relative to its area than
496 any other ocean. The freshwater inputs, traceable in the central AO, result in strong stratification^{20,192,194}.
497 Mercury enters the AO via rivers and coastal erosion, atmospheric deposition, and through Atlantic and
498 Pacific Ocean inflows, and is removed from the Arctic water column by evasion, sedimentation, and
499 outflow to the Atlantic Ocean.

500 Overall, Hg concentrations measured in Arctic waters are somewhat higher than in the North Atlantic¹⁹³
501 and global ocean¹⁹⁵ (Fig. 3a). The AO is the only ocean where the highest water Hg concentrations reside
502 in the surface waters ($0.24\pm 0.12 \text{ ng L}^{-1}$, 0-20 m, $n=159$)¹⁹⁵. North Atlantic-sourced waters flowing into
503 the Arctic are lower in Hg ($0.15\pm 0.07 \text{ ng L}^{-1}$, 200–500m, $n=164$) as are Arctic deep waters below 500 m
504 ($0.13\pm 0.06 \text{ ng L}^{-1}$, $n=369$). There is no significant difference in Hg concentrations between the Nansen,
505 Amundsen, Makarov and Canadian Basins. Higher Hg levels were observed in the CAA^{21,188} and on the
506 North-Eastern Greenland Shelf, but not at the Barents Sea Opening²⁰. Mercury concentrations in the East
507 Siberian and Chukchi shelf (< 200m) averaged $0.22\pm 0.08 \text{ ng L}^{-1}$ ($n=74$)^{22,196}.

508 Using all available mean seawater total Hg concentration data^{21,22,24,187,191,192,194}, the water column Hg
509 budget is revised to 1870 Mg (distributed by depth as: 44 Mg (0-20m), 228 Mg (20-200m), 224 Mg (200-
510 500m) and 1375 Mg (500m-bottom)). This estimate is lower than reported in previous AO Hg mass
511 budget studies ($2847\text{-}7920 \text{ Mg}$)^{6,197}, primarily as a result of being based on more high latitude water
512 measurements.

513 While there is no long-term temporal dataset for AO seawater Hg concentrations, some studies suggest
514 that AO Hg concentrations and distribution will likely be affected by climate change. Climate change-
515 induced loss of sea ice (5–10% decrease/decade in annual mean sea ice extent^{198,199}) might result in a net
516 increase in evasion flux of Hg from water to air^{5,6}, through enhanced net Hg reduction¹⁹ and higher open
517 water surface area⁵. In parallel, higher primary productivity²⁰⁰ could enhance the removal of Hg from
518 surface waters with the particle settling flux.

519 *Air-sea Hg(0) exchange*

520 Measurements of Hg(0) concentrations in the AO are limited^{19,22,109} but show elevated concentrations in
521 upper ocean waters especially under sea ice (Fig. 3b). Air-sea exchange of Hg(0) is a diffusion process
522 largely controlled by surface water Hg(0) and water turbulence. High resolution measurements of surface
523 water Hg(0) indicate supersaturation in summer and fall resulting in a net sea to air flux^{19,109}. Hg(0)
524 evasion is higher in coastal areas and the CAA ($\sim 144 \text{ ng m}^{-2} \text{ d}^{-1}$)¹⁸⁹ than in the ice-free open ocean (< 24
525 $\text{ ng m}^{-2} \text{ d}^{-1}$)^{19,109,110}, and higher in summer than in fall^{19,109}.

526 Sea ice acts as a barrier to evasion^{19,109}, allowing Hg(0) to build up likely by biologically-mediated and
527 dark redox processes ($\sim 10^{-7} \text{ s}^{-1}$)^{8,101,196,201-203} linked to organic carbon cycling at the ice-water interface.
528 Reaching a steady state under sea ice likely takes months assuming no other losses. The elevated
529 concentrations suggest the potential for a substantial release of Hg(0) during spring/summer sea-ice
530 melt¹⁰⁹. As a result, there is likely enhanced Hg evasion in spring/summer during ice melt, and models
531 suggest that this and snowpack melt contribute to the elevated atmospheric Hg concentrations measured at
532 coastal locations in late spring/early summer^{34,48,78,80}. The timescale for surface water Hg(0) to return to
533 open water concentrations is several weeks¹⁹.

534 A net Hg(0) evasion (diffusion) flux of 24.9 Mg y^{-1} from sea to air is estimated for the AO based on
535 measurements of open ocean water flux estimates ($77 \text{ ng m}^{-2} \text{ d}^{-1}$)^{19,109}, and an open water area ranging
536 from 10% in winter to 65% in summer. Further, release of Hg(0) from under sea ice during melt is added
537 based on seasonal sea ice changes and under ice Hg(0) concentrations (2.9 Mg y^{-1}), reaching a total
538 evasion of 27.8 Mg y^{-1} . This is comparable to estimates of 45.0 and 23.3 Mg y^{-1} from GEOS-Chem and
539 GEM-MACH-Hg models, respectively, providing a best estimate of 32 Mg y^{-1} (range $23\text{--}45 \text{ Mg y}^{-1}$)
540 based on measurements and models.

541 *Sea ice Hg exchange*

542 Arctic sea ice is central to the Arctic Hg cycle, as it regulates the air-sea exchange of volatile Hg
543 species¹⁹, and controls the amount of sunlight that penetrates the water column and thus the photo-

544 mediated transformations of Hg²⁰⁴. Moreover, sea ice traps, accumulates^{23,205,206}, and transports Hg across
545 the AO¹⁹⁴. When seawater freezes, it traps Hg dissolved in the water or bound to particles in the newly
546 formed ice. Sea ice matures by rejecting salts and dissolved Hg into brine, a halide and Hg-rich, dense,
547 cold phase^{205,208}. This brine has a low freezing temperature, remains liquid, and pools into channels. Thus,
548 brine expulsion can move Hg from the bulk ice to the underlying seawater, enriching it (1.05±0.52 ng
549 L⁻¹)²³ relative to the polar mixed layer (~0.3 ng L⁻¹)¹⁹². The amount of particulate Hg is highly variable
550 and depends on the turbidity of waters where the sea ice originally formed. For example, “dirty ice”
551 formed near the coast in resuspended sediment-rich waters²⁰⁷, is enriched in particulate Hg²⁰⁶. Particle-
552 bound contaminants are less likely to move into the brine and remain in the ice²⁰⁶. As sea ice ages, it loses
553 Hg through brine expulsion or move to the surface and form frost flowers^{23,208,119}, but it can also gain Hg
554 from atmospheric deposition or adsorption on snow and frost flowers^{139,140}. Mercury can re-enter brine
555 channels from underlying waters and sediment. All these processes affecting sea ice Hg result in the wide
556 range of concentrations reported, from 0.1-12 ng L⁻¹^{19,23,205,206,209}. However, when only Central AO sea
557 ice^{19,23} is considered, the overall concentration and the variability within cores and across sites are
558 consistent (0.61±0.29 ng L⁻¹). Using this mean Hg concentration and the mean sea ice volume for 2015
559 (1.54x10⁴ km³), the AO sea ice Hg reservoir is estimated as 9.2 Mg (range 3.5-14.6 Mg). Annually,
560 1.4±0.4 Mg y⁻¹ of sea ice Hg is exported out of the AO (through Fram Strait), assuming an approximate
561 sea ice export of 2400±640 km³.

562 *Ocean currents Hg exchange*

563 Mercury enters the AO through ocean currents from the Atlantic Ocean (43±9 Mg y⁻¹, Fram Strait²⁰; 6±4
564 Mg y⁻¹, Barents Sea Opening²⁰), and the Pacific Ocean (6²⁰ Mg y⁻¹, 1–14²² and 3.7-7.6⁶ Mg y⁻¹, Bering
565 Strait), and exits via the Fram Strait (54±13 Mg y⁻¹)²⁰ and Davis Strait (19±8 Mg y⁻¹)²⁰ into the Atlantic
566 Ocean. Improved data²⁰ have well-constrained the total AO oceanic Hg inflow (55±7 Mg y⁻¹) and outflow
567 (73±8 Mg y⁻¹), compared to previous estimates (46¹⁹⁷ and 53 (40–62)⁶ Mg y⁻¹, inflow; 68¹⁹⁷ and 79 (49–
568 122)⁶ Mg y⁻¹, outflow). Observations consistently suggest a net Hg export from the AO to the North
569 Atlantic Ocean (18-26.2 Mg y⁻¹)^{6,20,197}, due to the relatively high Hg concentrations in the outflow
570 (0.26±0.09 ng L⁻¹) compared to the inflow (0.16±0.06 ng L⁻¹)²⁰.

571 While a slightly increasing trend in water inflow from Bering Strait has been observed, the Fram Strait
572 water exchange shows strong seasonal variability but small long-term changes²¹⁰, which suggests no
573 future trend for ocean Hg exchange due to low water input from Bering Strait²¹¹.

574 *Water-sediment Hg exchange*

575 Rivers and coastal erosion, ocean primary production and wind-blown dust deposition provide
576 organic/inorganic particles to the AO that accumulates in marine sediments (490 and 134 Gg/y in shelf
577 and deep basins, respectively^{6,212,213}). The high affinity of Hg to particles and the drawdown of particles
578 from the surface ocean provide an important mechanism for Hg settling and burial in the marine
579 sediments on millennial time scales²¹⁴.

580 The highest particulate Hg concentrations in the AO are observed over the shelf (0.031 ± 0.01 ng L⁻¹)
581 followed by Eastern (0.019 ± 0.008 ng L⁻¹) and Western (0.014 ± 0.010 ng L⁻¹) basins, which are about 3
582 times higher than in the North Atlantic^{22,191,214}. When normalizing to particle weight, the Hg concentration
583 of the particles increases from the shelf (211 ± 69 ng g⁻¹) to the central Arctic Ocean ($1,484 \pm 467$ ng g⁻¹)¹⁹¹
584 compared to up to 90 ng g⁻¹ (36 ± 27 ng g⁻¹)²¹⁵ near the Mackenzie River and 240-1,080 ng g⁻¹ in the
585 offshore North Atlantic Ocean²¹⁵. The AO sediments Hg concentrations are also higher in the deep basin
586 (60.4 ± 44.5 ng g⁻¹, n=70) compared to the shelf (28.9 ± 22.0 ng g⁻¹, n=70)¹⁹¹.

587 Using suspended particulate Hg concentrations and Th-234 radionuclide tracer measurements, Hg settling
588 fluxes from surface waters to below 100 m of 34.7 ± 15.6 ng m⁻² d⁻¹ (shelf) and 9.2 ± 43.3 ng m⁻² d⁻¹ (deep
589 basin) are estimated¹⁹¹. The central AO Hg settling flux has a high uncertainty because of very low
590 particle export fluxes. Total Hg settling fluxes from the surface ocean waters (100 meters depth) of
591 122 ± 55 (shelf) Mg y⁻¹ and 7.2 ± 17 Mg y⁻¹ (deep basin) are estimated for the AO area considered in this
592 Review.

593 Marine sediments exchange Hg with overlying seawater by sedimentation/resuspension and bi-directional
594 diffusion of Hg species⁶. A recent Th-230 radionuclide tracer approach refined the deep basin Hg burial
595 flux²¹⁶ to 3.9 ± 0.7 Mg y⁻¹, compared to previous estimates (8 Mg y⁻¹ and 3.5 ± 3 Mg y⁻¹)^{6,191}. Higher Hg
596 burial rates are estimated in shelf sediments (30 Mg y⁻¹ and 20 ± 14 Mg y⁻¹)^{6,191}. However, the shelf Hg
597 burial flux is likely underestimated due to a lack of measurements in the inner shelf, where burial is
598 expected to be higher²¹⁷. New observations on the Siberian shelf suggest Hg burial rates of up to 75 Mg y⁻¹
599 and that earlier estimates are biased low because of underestimated sediment density²¹⁷. Using all
600 available data^{191,216,217} gives a revised shelf Hg burial flux of 42 ± 31 Mg y⁻¹ (n=114). Net benthic Hg
601 diffusion flux from the AO sediments to the overlying water column is estimated at 5 Mg y⁻¹ for both
602 shelf and deep basins based on limited data⁶, calling for further measurements especially in the deep
603 basin.

604 **Mercury mass balance**

605 In the last decade, observations and modeling have advanced the understanding of AO Hg mass
606 balance^{6,7,20,34,78,191}. This review provides a revised AO and first terrestrial system Hg mass balance
607 assessment in the Arctic (Fig. 4; Table S9).

608 The Hg mass balance developed in this Review suggests that presently annual atmospheric Hg deposition
609 of $118 \pm 20 \text{ Mg y}^{-1}$ to the Arctic terrestrial system is balanced by legacy Hg emissions from soils and
610 vegetation (including wildfires) and riverine and erosional exports totaling $113 \pm 32 \text{ Mg y}^{-1}$ (Table S9).
611 Northward-flowing rivers import part of the $\sim 62 \text{ Mg y}^{-1}$ Hg deposited over watershed areas south of 60°N
612 to the Arctic, which is not accounted in the above terrestrial Hg inputs. Riverine Hg inflow to the Arctic is
613 presently unquantifiable, as riverine fluxes and export rates are mostly measured at river mouths only.
614 Large pools of Hg ($212 (184\text{-}240) \text{ Gg}$, 0-1m, > three orders of magnitude larger than any other reservoirs)
615 have accumulated in active layer and permafrost soils¹²⁸⁻¹³⁰, derived from thousands of years of
616 atmospheric deposition. It is likely that Arctic warming led to increased releases of terrestrial Hg in recent
617 decades by intensifying permafrost thaw^{14,218}, glacier melt^{16,17}, coastal erosion¹⁹⁷, wildfires^{13,219}, and river
618 discharge¹⁶⁸. However, observational data are currently insufficient to quantify these changes. Uptake of
619 Hg(0) by vegetation/soils/snowpack is now thought to be the dominant Hg deposition pathway in the
620 Arctic terrestrial ecosystems^{68,74,93}, which is currently underestimated in models³⁷. Legacy Hg emissions
621 from undisturbed tundra soils seem negligible⁶⁸, and are likely overestimated in models ($6.5\text{-}59 \text{ Mg y}^{-1}$;
622 Table S9). Finally, wildfire Hg emissions and its speciation from boreal forests are highly temporally
623 variable and uncertain, and reported to be rising^{56,63-66}.

624 Previously, Hg inputs to the AO from atmospheric deposition ($45\text{-}108 \text{ Mg y}^{-1}$)^{6,34,78,80}, pan-Arctic river
625 runoff ($46\text{-}80 \text{ Mg y}^{-1}$)^{6,34,36,78}, coastal erosion ($15\text{-}32 \text{ Mg y}^{-1}$)^{6,34}, and ocean current inflow ($46\text{-}55 \text{ Mg y}^{-1}$)
626 ^{6,20,197} were estimated to be offset by evasion ($33\text{-}99 \text{ Mg y}^{-1}$)^{6,34,80}, ocean outflow ($68\text{-}79 \text{ Mg y}^{-1}$)^{6,20,197}
627 and sediment burial (28 Mg y^{-1})^{6,191} fluxes. In contrast, the revised AO Hg inputs ($204.5 \pm 27.3 \text{ Mg y}^{-1}$)
628 exceed outputs ($152.3 \pm 33.9 \text{ Mg y}^{-1}$) by $52.2 \pm 43.5 \text{ Mg y}^{-1}$ (Table S9), and indicate that Hg removal from
629 the AO ocean waters is currently underestimated. Shelf Hg burial flux is likely underestimated due to a
630 lack of measurements in the inner shelf, where burial is expected to be higher²¹⁷ (arrow q, Fig. 4).
631 Mercury settling in shelf regions could be subsequently transferred into the deep basin via hydrodynamic
632 exchanges^{220,221} (arrow r, Fig. 4). Recent observations on the Siberian shelf suggest that Hg burial rates of
633 up to 75 Mg y^{-1} are possible²¹⁷, and that earlier estimates are biased low because of underestimated
634 sediment density. Large observed shelf Hg settling of $122 \pm 55 \text{ Mg y}^{-1}$ from surface waters¹⁹¹ also indicates
635 that the revised shelf burial flux ($42 \pm 31 \text{ Mg y}^{-1}$) using all available data ($n=114$)^{191,216,217} might be
636 underestimated by up to $80 \pm 63.1 \text{ Mg y}^{-1}$, which is in the range of excess AO Hg input ($52.2 \pm 43.5 \text{ Mg y}^{-1}$)
637 found here. Riverine and erosional Hg exports to the AO are highly seasonal, primarily occurring during

638 spring freshet and summer months^{7,165}. The fate of terrestrial Hg input in the AO is currently uncertain,
639 and likely varies between watersheds due to differences in the reactivity of particulate organic carbon-
640 bound mercury^{36,194,222,223}.

641 **Summary and future perspectives**

642 A quantitative understanding of the Arctic total Hg levels, movement pathways and mass balance is
643 developed by integrating observations and modeling, in support of future effectiveness evaluation of the
644 Minamata Convention. About 330 Mg Hg presently circulates in the Arctic atmosphere north of 60° N,
645 primarily transported from distant global emissions sources (6000-9000 Mg y⁻¹)³⁷, with small
646 contributions from regional anthropogenic emissions (14 Mg y⁻¹) and re-emission of legacy Hg by
647 wildfires (8.8±6.4 Mg y⁻¹) and by vegetation/soil evasion (24 (6.5-59) Mg y⁻¹). Annually, 118±20 Mg y⁻¹
648 and 64.5±19.8 Mg y⁻¹ of atmospheric Hg is deposited to Arctic terrestrial and marine environments,
649 respectively.. Mercury deposited on land is stored in permafrost soils (597,000 Mg, 0-3 m), glaciers
650 (2,415 Mg) and seasonal snowpacks (~39 (35–42) Mg). Rivers and coastal erosion respectively transfer
651 41±4 and 39 (18-52) Mg y⁻¹ of terrestrial Hg to the AO reservoir (1,870 Mg). Oceanic inflows supply
652 55±7 Mg y⁻¹ of Hg to the AO, while the outflow exports 73±8 Mg y⁻¹ to the Atlantic Ocean. About 32
653 (23-45) Mg y⁻¹ of Hg is lost from the AO by evasion to air, while sedimentation buries 42±31 Mg y⁻¹
654 (likely significantly underestimated) and 3.9±0.7 Mg y⁻¹ of oceanic Hg in continental shelves and the deep
655 basin, respectively.

656
657 Research advances in following key areas is needed to improve the understanding of the processes
658 controlling the present and future Arctic Hg cycling. To better quantify anthropogenic Hg emissions in
659 the Arctic, improved information on Hg regional sources such as activity data on quantities of fuels and
660 raw materials used at major point sources, and their associated Hg content, as well as that of their
661 products is necessary. Information on the locations, quantities and practices involved in waste disposal in
662 the Arctic, and waste characteristics would allow better estimates, not only of Hg emissions but also of
663 the releases of a range of environmental contaminants. Mercury measurements in biome-specific wildfires
664 including peat fires, and improved methods for wildfire Hg emission inventories including global
665 warming impacts are required. Mechanistic modeling schemes for vegetation Hg uptake from the
666 atmosphere incorporating plant physiology, redox chemistry and environmental variables are needed. The
667 role of sea ice/snow dynamics and photo-chemistry in marine Hg redox processes needs to be better
668 understood. More air-surface Hg flux observations from inland, coastal and marine sites would help to
669 develop mechanistic knowledge of atmosphere-surface and sediment-ocean Hg exchange processes, and
670 their climate warming effects.

671 Mercury releases from thawing Arctic permafrost into rivers and from eroding coastlines into the ocean
672 depend on factors such as soil organic carbon, geomorphology, hydrology, and ground ice, which vary
673 widely across the Arctic. Thus, spatially-resolved permafrost soil Hg inventories are needed to understand
674 and quantify Hg releases to downstream and coastal ecosystems across different regions and thaw
675 regimes. Improved estimates of glacial Hg reservoir and its outflow across a range of catchment sizes and
676 geological settings, particularly where local ecosystem impacts could be large (such as productive fjords),
677 and projections of glacial meltwater discharges into lakes and the ocean are required to forecast impacts
678 to aquatic ecosystems.

679 Further application of Hg stable isotopes might better constrain the processes influencing cycling of
680 Hg through the air-soil-river pathway into the AO and marine food webs under present and future
681 conditions. Applying extensive knowledge of the radiocarbon ages and soil profile sources of dissolved
682 and particulate organic carbon in Arctic rivers might help elucidate the natural:anthropogenic source
683 attribution of dissolved and particulate Hg fluxes into the AO. Seasonal observations of suspended
684 sediment and dissolved Hg fluxes onto the shelf and in the central AO are needed to understand and
685 quantify the fate of pulsed terrestrial Hg inputs. Finally, integrated atmosphere-land-ocean
686 biogeochemical Hg models are needed to simulate the link between terrestrial Hg deposition and its
687 export, and the impacts of concurrent changes in global climate and Hg emissions on Arctic Hg cycling.

688 **Figure 1. | Atmospheric Hg distribution in the Arctic.** a) Model ensemble (DEHM, GEM-MACH-Hg,
689 GLEMOS, GOES-Chem) simulated (this Review) annual average surface air gaseous elemental mercury
690 (Hg(0)) concentrations in 2015. b) Annual mercury (Hg) wet deposition flux in 2015. c) Annual Hg total
691 deposition in the Arctic in 2015. Circles show observations in the same color scale. The blue lines
692 delineate major pan-Arctic river watersheds. Air concentrations and wet deposition observations are from
693 ECCC-AMM⁸³, AMNet⁸⁴, EMEP⁸⁵ and REF²²⁴, and total deposition observation is from REF²⁹.

694 **Figure 2. | Distribution of total Hg in Arctic soils, wintertime deposition and rivers.** a) Mean mercury
695 (Hg) concentrations of active layer soils within 0-1 m depth. b) Mean Hg concentration of permafrost
696 layer soils within 0-1 m depth. Symbols in Figs. a-b represent the soil type with squares for organic soils,
697 circles for mineral soils and triangles for combined organic/mineral soils c) Wintertime (September-May)
698 model ensemble simulated Hg deposition to snowpack. Observations are shown in circles d) Annual river
699 total mercury (THg) yields (annual riverine THg mass flux to the Arctic Ocean divided by watershed
700 area) for pan-Arctic watersheds. Larger watersheds (> 10,000 km²) are delineated where possible, while
701 smaller watersheds are represented by symbols. Solid colored watersheds and squares represent measured
702 THg yields (n=32) reported in REF^{7,16-18,107,162,165,169,173,225-228}. Hatched watersheds and circles are THg

703 yields (n=100) modelled in REF⁷. Numbered watersheds are: 1) Amguema, 2) Palyavaam, 3) Bolshoy
 704 Anyuy, 4) Indigirka, 5) Yana, 6) Omoloy, 7) Olenek, 8) Anabar, 9) Khatanga, 10) Katun, 11) Taz, 12)
 705 Pur, 13) Nadym, 14) Pechora, 15) Mezen, 16) Onega, 17) Ponoy, 18) Varzuga, 19) Nyzhny Vig (Sor), 20)
 706 Kem, 21) Tana, 22) George, 23) À la Baleine, 24) Povungnituk, 25) Arnaud, 26) Aux Feuilles, 27)
 707 Koksoak, 28) Grande Rivière de la Baleine, 29) La Grande Rivière, 30) Eastmain, 31) Rupert, 32)
 708 Nottaway, 33) Moose, 34) Albany, 35) Attawapiskat, 36) Winisk, 37) Severn, 38) Hayes, 39) Churchill,
 709 40) Seal, 41) Thlewiaza, 42) Ferguson, 43) Thelon, 44) Back, 45) Ellice, 46) Coppermine, 47) Hornaday,
 710 48) Anderson, 49) Peel, 50) Noatak, 51) Kobuk. For rivers with multiple yield estimates, only the
 711 estimate for the most recent time period is shown. e| Repeat images during spring melt in Utqiagvik
 712 (formerly Barrow), Alaska during a typical spring melt. In a matter of a few weeks the tundra surface
 713 goes from completely snow covered to a wet vegetation and soil surface exposed to continuous sunlight.

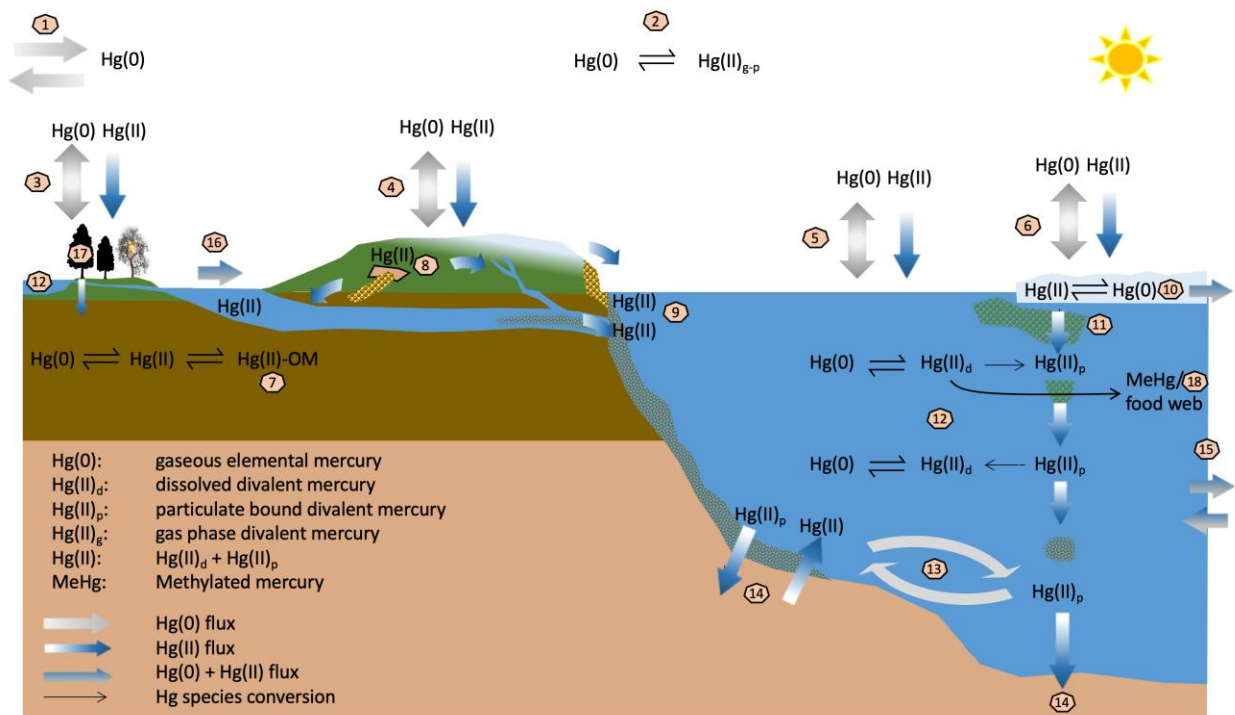
714 **Figure 3. | Spatial distribution of Hg in the Arctic Ocean.** a| Synoptic pan-Arctic transect of total Hg
 715 concentrations during the 2015 GEOTRACES cruises (ng L^{-1})^{22,191,194}. The insert map shows the transect
 716 (red band) and available Hg observations (blue dots). b| Surface water Hg(0) concentrations (ng L^{-1}) in
 717 the Arctic Ocean during a summer (left¹⁰⁹) and fall (right¹⁹) cruise. Grey areas indicate ice-covered water.
 718 The area indicated with a punctuated line represents the outflow region from the Mackenzie River Basin.
 719 Ocean currents transport Hg via waters entering the AO from the Atlantic Ocean ($235 \pm 31.7 \times 10^3 \text{ km}^3 \text{ y}^{-1}$,
 720 Fram Strait; $72.9 \pm 38.0 \times 10^3 \text{ km}^3 \text{ y}^{-1}$, Barents Sea Opening), and the Pacific Ocean ($22.2 \pm 22.2 \times 10^3 \text{ km}^3 \text{ y}^{-1}$,
 721 Bering Strait), and exiting the AO through the Davis Strait ($66.6 \pm 22.2 \times 10^3 \text{ km}^3 \text{ y}^{-1}$) and Fram Strait
 722 ($273 \pm 114 \times 10^3 \text{ km}^3 \text{ y}^{-1}$) into the Atlantic Ocean^{210,229}.

724 **Figure 4 | Arctic Hg inter-compartmental annual fluxes and reservoir budgets.** Best estimates (black
 725 numbers) and uncertainties in terrestrial and oceanic Hg annual fluxes (identified by arrows with letters)
 726 are: anthropogenic (a, 14 Mg y^{-1}), natural and legacy soils/vegetation volatilization (b, $24_{-17.5}^{+35.1} \text{ Mg y}^{-1}$),
 727 wildfires (c, $8.8 \pm 6.4 \text{ Mg y}^{-1}$), and ocean (d, $32_{-9}^{+13} \text{ Mg y}^{-1}$) emissions; terrestrial (e, $118 \pm 20 \text{ Mg y}^{-1}$), and
 728 ocean (f, $64.5 \pm 19.8 \text{ Mg}$) deposition; coastal erosion (g, $39_{-11}^{+13} \text{ Mg y}^{-1}$), river (h, $41 \pm 4 \text{ Mg y}^{-1}$), ocean (i,
 729 $55 \pm 7 \text{ Mg y}^{-1}$), and benthic shelf (j, 5 Mg y^{-1}) imports; sea ice (k, $1.4 \pm 0.4 \text{ Mg y}^{-1}$), ocean (l, $73 \pm 8 \text{ Mg y}^{-1}$),
 730 shelf burial (m, $42 \pm 31 \text{ Mg y}^{-1}$) and deep ocean burial (n, $3.9 \pm 0.7 \text{ Mg y}^{-1}$) exports; and in-ocean shelf (o,
 731 $122 \pm 55 \text{ Mg y}^{-1}$) and deep ocean (p, $7.2 \pm 17 \text{ Mg y}^{-1}$) downward fluxes from surface waters. Best estimates
 732 (black numbers) and uncertainties for Hg budgets in reservoirs are: atmosphere ($330_{-40}^{+30} \text{ Mg}$), glaciers
 733 ($2,415 \pm 22 \text{ Mg}$), snowpacks (39_{-4}^{+3} Mg), soils (surface, 0-0.3 m: $49 \pm 13 \text{ Gg}$; active layer, 0-1 m: 212 ± 28
 734 Gg ; permafrost, 0-3 m: 597 Gg), and Arctic Ocean ($44 \pm 22 \text{ Mg}$, 0-20 m; $228 \pm 112 \text{ Mg}$, 20-200 m;

735 224±106 Mg, 200-500 m; 1,375±616 Mg, 500 m and deeper). Red arrows indicate unknown Hg fluxes of
 736 burial in estuaries and inner shelf (q), transport from shelf to deep Arctic Ocean basin (r), and benthic
 737 deep basin (s) fluxes (identified in this Review). Arctic Ocean sediments Hg concentrations are 28.9±22.0
 738 ng g⁻¹ in shelf and 60.4±44.5 ng g⁻¹ in deep basin. Arctic land is defined as region north of 60° N and the
 739 Arctic Ocean is defined as the central basin, and Barents, Kara, Laptev East Siberian, Chukchi and
 740 Beaufort Seas, (94.5 x 10⁵ km², total ocean surface area; 53% shelf). The values of Hg fluxes and budgets
 741 are based on multi-model ensemble simulations and peer-reviewed literature (this Review).

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Box 1 | The role of speciation in Arctic mercury cycling



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749 Mercury (Hg) in the Arctic occurs in different chemical forms that are subject to (photo-)chemical and
 750 biologically-mediated transformations^{67,230,231}. Mercury is transported to and from the Arctic through air
 751 (1)²³², rivers (16)⁷ and ocean currents (15)²⁰. The atmospheric Hg pool (2) consist of gaseous elemental

752 Hg (Hg(0); >95%) and oxidized divalent Hg (Hg(II))⁶⁷, and it exchanges with vegetation, soil, snow, ice
753 and seawater (3-6)⁶ primarily through dry deposition of Hg(0) and Hg(II), wet deposition of Hg(II), and
754 evasion of Hg(0)^{19,37,68,93}. Transfers of Hg between terrestrial, aquatic and ice/snow reservoirs are
755 controlled by light, organic matter complexation, and microbial activity¹. Mercury stored in terrestrial
756 reservoirs, primarily as particulate Hg(II), is mobilized by snow/ice melt^{16,17,106}, permafrost thaw¹⁴,
757 bedrock weathering, soil erosion and surface runoff (7, 8, 10, 12); and can be transported into wetlands,
758 lakes and riverbeds or onto marine shelves (14)^{165,167,168,183}. Once it reaches the Arctic Ocean, Hg is
759 distributed by currents (13)^{20,36}, settling (11, 14), biological uptake and release (11, 18), and
760 remineralization (12)^{6,191,233}. A fraction of Hg(0) and Hg(II) can be converted to methylmercury (MeHg)
761 (18)^{231,234,235}. Unlike in other reservoirs, MeHg can in ocean waters be the dominant form of Hg, most
762 prominently observed in the deeper parts of the ocean¹⁹². MeHg is a neurotoxic that bioaccumulates and
763 biomagnifies in food webs often reaching high levels in top predators particularly in aquatic environments
764 where food webs are longer than in terrestrial systems (18)¹. Phytoplankton uptake is the main pathway of
765 MeHg into aquatic food webs²³⁶. A distinctive feature of the Arctic Ocean is the presence of a large water
766 MeHg peak overlapping with maximum phytoplankton activity^{187,192}, resulting in favorable conditions for
767 uptake by food webs. MeHg concentration in phytoplankton is 10⁴ or more times higher than in water
768 although uptake is species and conditions dependent^{237,238}. Hg concentration continues to increase at each
769 subsequent trophic level in the food web culminating in long-lived predatory fish and marine mammals
770 (including amphibious animals such as polar bears) exceeding Hg concentrations 10⁷ times that of
771 seawater¹.

772

773 **Author contributions**

774 A.D. designed, coordinated and led the study and manuscript writing, editing and revising. All authors
775 (listed in alphabetical order) contributed to analyzing data, writing and/or conducting model simulations
776 of specific sections, developing Arctic mercury mass balance, key points and future perspectives, and
777 revising manuscript. K.A.S.P. and C.Z. also contributed to overall editing and formatting.

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790
791 **Competing interests**

792 The authors declare no competing interests.

793 **References**

- 794 1. AMAP. AMAP Assessment 2011: Mercury in the Arctic. xiv + 193 pp. (Oslo, Norway, 2011).
795 2. Dietz, R. *et al.* Current state of knowledge on biological effects from contaminants on arctic
796 wildlife and fish. *Sci. Total Environ.* **696**, 133792 (2019).
797 3. Environment, U. Global Mercury Assessment 2018. (Geneva Switzerland, 2019).
798 4. Basu, N. *et al.* A State-of-the-Science Review of Mercury Biomarkers in Human Populations
799 Worldwide between 2000 and 2018. *Environ. Health Perspect.* **126**, 106001 (2018).
800 5. Chen, L. *et al.* A decline in Arctic Ocean mercury suggested by differences in decadal trends of
801 atmospheric mercury between the Arctic and northern midlatitudes. *Geophys. Res. Lett.* **42**, 6076-
802 6083 (2015).
803 6. Soerensen, A. L. *et al.* A mass budget for mercury and methylmercury in the Arctic Ocean.
804 *Global Biogeochem. Cycles* **30**, 560-575 (2016).
805 7. Sonke, J. E. *et al.* Eurasian river spring flood observations support net Arctic Ocean mercury
806 export to the atmosphere and Atlantic Ocean. *PNAS* **115**, E11586 (2018).
807 8. Qureshi, A., O'Driscoll, N. J., MacLeod, M., Neuhold, Y.-M. & Hungerbühler, K. Photoreactions
808 of Mercury in Surface Ocean Water: Gross Reaction Kinetics and Possible Pathways. *Environ.*
809 *Sci. Technol.* **44**, 644-649 (2010).
810 9. O'Driscoll, N. J. *et al.* Dissolved Gaseous Mercury Production at a Marine Aquaculture Site in
811 the Mercury-Contaminated Marano and Grado Lagoon, Italy. *Bull. Environ. Contam. Toxicol.*
812 **103**, 218-224 (2019).
813 10. Mason, R. P., Reinfelder, J. R. & Morel, F. M. M. Bioaccumulation of mercury and
814 methylmercury. *Water Air Soil Pollut.* **80**, 915-921 (1995).
815 11. AMAP. AMAP Assessment 2021: Mercury in the Arctic. Summary for Policy-Makers. 16 pp.
816 (Tromsø, Norway, 2021).
817 12. Clem, K. R. *et al.* Record warming at the South Pole during the past three decades. *Nat. Clim.*
818 *Change* **10**, 762-770 (2020).
819 13. Kumar, A. & Wu, S. Mercury Pollution in the Arctic from Wildfires: Source Attribution for the
820 2000s. *Environ. Sci. Technol.* **53**, 11269-11275 (2019).
821 14. St. Pierre, K. A. *et al.* Unprecedented Increases in Total and Methyl Mercury Concentrations
822 Downstream of Retrogressive Thaw Slumps in the Western Canadian Arctic. *Environ. Sci.*
823 *Technol.* **52**, 14099-14109 (2018).
824 15. Schaefer, K. *et al.* Potential impacts of mercury released from thawing permafrost. *Nat. Commun.*
825 **11**, 4650 (2020).

- 826 16. St. Pierre, K. A. *et al.* Drivers of Mercury Cycling in the Rapidly Changing Watershed of the
827 High Arctic's Largest Lake by Volume (Lake Hazen, Nunavut, Canada). *Environ. Sci. Technol.*
828 **53**, 1175-1185 (2019).
- 829 17. Søndergaard, J. *et al.* Mercury exports from a High-Arctic river basin in Northeast Greenland (74
830 degrees N) largely controlled by glacial lake outburst floods. *Sci. Total Environ.* **514**, 83-91
831 (2015).
- 832 18. Zdanowicz, C. *et al.* Snowmelt, glacial and atmospheric sources of mercury to a subarctic
833 mountain lake catchment, Yukon, Canada. *Geochim. Cosmochim. Acta* **238**, 374-393 (2018).
- 834 19. DiMento, B. P., Mason, R. P., Brooks, S. & Moore, C. The impact of sea ice on the air-sea
835 exchange of mercury in the Arctic Ocean. *Deep Sea Res. Part I* **144**, 28-38 (2019).
- 836 20. Petrova, M. V. *et al.* Mercury species export from the Arctic to the Atlantic Ocean. *Mar. Chem.*
837 **225**, 103855 (2020).
- 838 21. Wang, K. *et al.* Subsurface seawater methylmercury maximum explains biotic mercury
839 concentrations in the Canadian Arctic. *Sci. Rep.* **8**, 14465 (2018).
- 840 22. Agather, A. M., Bowman, K. L., Lamborg, C. H. & Hammerschmidt, C. R. Distribution of
841 mercury species in the Western Arctic Ocean (U.S. GEOTRACES GN01). *Mar. Chem.* **216**,
842 103686 (2019).
- 843 23. Schartup, A. T., Soerensen, A. L. & Heimbürger-Boavida, L.-E. Influence of the Arctic Sea-Ice
844 Regime Shift on Sea-Ice Methylated Mercury Trends. *Environ. Sci. Technol. Lett.* **7**, 708-713
845 (2020).
- 846 24. Kim, J. *et al.* Mass Budget of Methylmercury in the East Siberian Sea: The Importance of
847 Sediment Sources. *Environ. Sci. Technol.* **54**, 9949-9957 (2020).
- 848 25. Jiskra, M., Sonke, J. E., Agnan, Y., Helmig, D. & Obrist, D. Insights from mercury stable
849 isotopes on terrestrial-atmosphere exchange of Hg(0) in the Arctic tundra. *Biogeosciences* **16**,
850 4051-4064 (2019).
- 851 26. Blum, J. D., Sherman, L. S. & Johnson, M. W. Mercury Isotopes in Earth and Environmental
852 Sciences. *Annu. Rev. Earth Planet. Sci.* **42**, 249-269 (2014).
- 853 27. Štok, M., Baya, P. A. & Hintelmann, H. The mercury isotope composition of Arctic coastal
854 seawater. *C.R. Geosci.* **347**, 368-376 (2015).
- 855 28. Zdanowicz, C. M. *et al.* Historical variations of mercury stable isotope ratios in Arctic glacier firn
856 and ice cores. *Global Biogeochem. Cycles* **30**, 1324-1347 (2016).
- 857 29. Obrist, D. *et al.* A review of global environmental mercury processes in response to human and
858 natural perturbations: Changes of emissions, climate, and land use. *Ambio* **47**, 116-140 (2018).
- 859 30. Dibble, T. S., Tetu, H. L., Jiao, Y., Thackray, C. P. & Jacob, D. J. Modeling the OH-Initiated
860 Oxidation of Mercury in the Global Atmosphere without Violating Physical Laws. *J. Phys. Chem.*
861 *A* **124**, 444-453 (2020).
- 862 31. Saiz-Lopez, A. *et al.* Photochemistry of oxidized Hg(I) and Hg(II) species suggests missing
863 mercury oxidation in the troposphere. *PNAS* **117**, 30949 (2020).
- 864 32. Zhang, Y. *et al.* Observed decrease in atmospheric mercury explained by global decline in
865 anthropogenic emissions. *PNAS* **113**, 526-531 (2016).
- 866 33. Angot, H. *et al.* Chemical cycling and deposition of atmospheric mercury in polar regions: review
867 of recent measurements and comparison with models. *Atmos. Chem. Phys.* **16**, 10735-10763
868 (2016).
- 869 34. Fisher, J. A. *et al.* Riverine source of Arctic Ocean mercury inferred from atmospheric
870 observations. *Nat. Geosci.* **5**, 499-504 (2012).
- 871 35. Fisher, J. A. *et al.* Factors driving mercury variability in the Arctic atmosphere and ocean over the
872 past 30 years. *Global Biogeochem. Cycles* **27**, 1226-1235 (2013).
- 873 36. Zhang, Y. *et al.* Biogeochemical drivers of the fate of riverine mercury discharged to the global
874 and Arctic oceans. *Global Biogeochem. Cycles* **29**, 854-864 (2015).
- 875 37. Zhou, J., Obrist, D., Dastoor, A., Jiskra, M. & Ryjkov, A. Vegetation uptake of mercury and
876 impacts on global cycling. *Nat. Rev. Earth Environ.* **2**, 269-284 (2021).

- 877 38. Bishop, K. *et al.* Recent advances in understanding and measurement of mercury in the
878 environment: Terrestrial Hg cycling. *Sci. Total Environ.* **721**, 137647 (2020).
- 879 39. AMAP & Environment, U. Technical Background Report for the Global Mercury Assessment
880 2018. Arctic Monitoring and Assessment Programme. viii + 426 pp including E-Annexes (Oslo,
881 Norway & Geneva, Switzerland, 2019).
- 882 40. Outridge, P. M., Mason, R. P., Wang, F., Guerrero, S. & Heimbürger-Boavida, L. E. Updated
883 Global and Oceanic Mercury Budgets for the United Nations Global Mercury Assessment 2018.
884 *Environ. Sci. Technol.* **52**, 11466-11477 (2018).
- 885 41. De Simone, F. *et al.* Particulate-phase mercury emissions from biomass burning and impact on
886 resulting deposition: a modelling assessment. *Atmos. Chem. Phys.* **17**, 1881-1899 (2017).
- 887 42. Kumar, A., Wu, S., Huang, Y., Liao, H. & Kaplan, J. O. Mercury from wildfires: Global emission
888 inventories and sensitivity to 2000–2050 global change. *Atmos. Environ.* **173**, 6-15 (2018).
- 889 43. Friedli, H. R., Arellano, A. F., Cinnirella, S. & Pirrone, N. Initial Estimates of Mercury Emissions
890 to the Atmosphere from Global Biomass Burning. *Environ. Sci. Technol.* **43**, 3507-3513 (2009).
- 891 44. Bozem, H. *et al.* Characterization of transport regimes and the polar dome during Arctic spring
892 and summer using in situ aircraft measurements. *Atmos. Chem. Phys.* **19**, 15049-15071 (2019).
- 893 45. Law, K. S. *et al.* Arctic Air Pollution: New Insights from POLARCAT-IPY. *Bull. Ameri.*
894 *Meteorol. Soc.* **95**, 1873-1895 (2014).
- 895 46. Monks, P. S. *et al.* Tropospheric ozone and its precursors from the urban to the global scale from
896 air quality to short-lived climate forcer. *Atmos. Chem. Phys.* **15**, 8889-8973 (2015).
- 897 47. Weiss-Penzias, P. *et al.* Quantifying Asian and biomass burning sources of mercury using the
898 Hg/CO ratio in pollution plumes observed at the Mount Bachelor observatory. *Atmos. Environ.*
899 **41**, 4366-4379 (2007).
- 900 48. Durnford, D., Dastoor, A., Figueras-Nieto, D. & Ryjkov, A. Long range transport of mercury to
901 the Arctic and across Canada. *Atmos. Chem. Phys.* **10**, 6063-6086 (2010).
- 902 49. Pithan, F. *et al.* Role of air-mass transformations in exchange between the Arctic and mid-
903 latitudes. *Nat. Geosci.* **11**, 805-812 (2018).
- 904 50. Lee, M.-Y., Hong, C.-C. & Hsu, H.-H. Compounding effects of warm sea surface temperature
905 and reduced sea ice on the extreme circulation over the extratropical North Pacific and North
906 America during the 2013–2014 boreal winter. *Geophys. Res. Lett.* **42**, 1612-1618 (2015).
- 907 51. Pozzoli, L., Dobricic, S., Russo, S. & Vignati, E. Impacts of large-scale atmospheric circulation
908 changes in winter on black carbon transport and deposition to the Arctic. *Atmos. Chem. Phys.* **17**,
909 11803-11818 (2017).
- 910 52. Dastoor, A. *et al.* Atmospheric mercury in the Canadian Arctic. Part II: Insight from modeling.
911 *Sci. Total Environ.* **509-510**, 16-27 (2015).
- 912 53. Steenhuisen, F. & Wilson, S. J. Development and application of an updated geospatial
913 distribution model for gridding 2015 global mercury emissions. *Atmos. Environ.* **211**, 138-150
914 (2019).
- 915 54. Friedli, H. R. *et al.* Mercury emissions from burning of biomass from temperate North American
916 forests: laboratory and airborne measurements. *Atmos. Environ.* **37**, 253-267 (2003).
- 917 55. Webster, J. P., Kane, T. J., Obrist, D., Ryan, J. N. & Aiken, G. R. Estimating mercury emissions
918 resulting from wildfire in forests of the Western United States. *Sci. Total Environ.* **568**, 578-586
919 (2016).
- 920 56. McLagan, D. S., Stupple, G. W., Darlington, A., Hayden, K. & Steffen, A. Where there is smoke
921 there is mercury: Assessing boreal forest fire mercury emissions using aircraft and highlighting
922 uncertainties associated with upscaling emissions estimates. *Atmos. Chem. Phys.* **21**, 5635-5653
923 (2021).
- 924 57. Giglio, L., Randerson, J. T. & van der Werf, G. R. Analysis of daily, monthly, and annual burned
925 area using the fourth-generation global fire emissions database (GFED4). *J. Geophys. Res.:*
926 *Biogeosci.* **118**, 317-328 (2013).

- 927 58. Giglio, L., Schroeder, W. & Justice, C. O. The collection 6 MODIS active fire detection
928 algorithm and fire products. *Remote Sens. Environ.* **178**, 31-41 (2016).
- 929 59. Lizundia-Loiola, J., Otón, G., Ramo, R. & Chuvieco, E. A spatio-temporal active-fire clustering
930 approach for global burned area mapping at 250 m from MODIS data. *Remote Sens. Environ.*
931 **236**, 111493 (2020).
- 932 60. Amiro, B. D. *et al.* Direct carbon emissions from Canadian forest fires, 1959-1999. *Can. J. Forest*
933 *Res.* **31**, 512-525 (2001).
- 934 61. Turetsky, M. R. *et al.* Wildfires threaten mercury stocks in northern soils. *Geophys. Res. Lett.* **33**,
935 L16403 (2006).
- 936 62. Kohlenberg, A. J., Turetsky, M. R., Thompson, D. K., Branfireun, B. A. & Mitchell, C. P. J.
937 Controls on boreal peat combustion and resulting emissions of carbon and mercury. *Environ. Res.*
938 *Lett.* **13**, 035005 (2018).
- 939 63. AMAP. Impacts of short-lived climate forcers on Arctic climate, air quality, and human health.
940 Summary for policy-makers. (Oslo, Norway, 2021).
- 941 64. Comeau, A. M., Li, W. K. W., Tremblay, J.-É., Carmack, E. C. & Lovejoy, C. Arctic Ocean
942 Microbial Community Structure before and after the 2007 Record Sea Ice Minimum. *PLoS One*
943 **6**, e27492 (2011).
- 944 65. Veira, A., Lasslop, G. & Kloster, S. Wildfires in a warmer climate: Emission fluxes, emission
945 heights, and black carbon concentrations in 2090–2099. *J. Geophys. Res.: Atmos.* **121**, 3195-3223
946 (2016).
- 947 66. Walker, X. J. *et al.* Increasing wildfires threaten historic carbon sink of boreal forest soils. *Nature*
948 **572**, 520-523 (2019).
- 949 67. Steffen, A. *et al.* Atmospheric mercury in the Canadian Arctic. Part I: A review of recent field
950 measurements. *Sci. Total Environ.* **509-510**, 3-15 (2015).
- 951 68. Obrist, D. *et al.* Tundra uptake of atmospheric elemental mercury drives Arctic mercury
952 pollution. *Nature* **547**, 201-204 (2017).
- 953 69. Wang, S. *et al.* Direct detection of atmospheric atomic bromine leading to mercury and ozone
954 depletion. *PNAS* **116**, 14479 (2019).
- 955 70. Abbatt, J. P. D. *et al.* Halogen activation via interactions with environmental ice and snow in the
956 polar lower troposphere and other regions. *Atmos. Chem. Phys.* **12**, 6237-6271 (2012).
- 957 71. Pratt, K. A. *et al.* Photochemical production of molecular bromine in Arctic surface snowpacks.
958 *Nat. Geosci.* **6**, 351-356 (2013).
- 959 72. Toyota, K., McConnell, J. C., Staebler, R. M. & Dastoor, A. P. Air–snowpack exchange of
960 bromine, ozone and mercury in the springtime Arctic simulated by the 1-D model PHANTAS
961 – Part 1: In-snow bromine activation and its impact on ozone. *Atmos. Chem. Phys.* **14**,
962 4101-4133 (2014).
- 963 73. Marelle, L. *et al.* Implementation and Impacts of Surface and Blowing Snow Sources of Arctic
964 Bromine Activation Within WRF-Chem 4.1.1. *J. Adv. Model. Earth Syst.* **13**, e2020MS002391
965 (2021).
- 966 74. Agnan, Y., Douglas, T. A., Helmig, D., Hueber, J. & Obrist, D. Mercury in the Arctic tundra
967 snowpack: temporal and spatial concentration patterns and trace gas exchanges. *Cryosphere* **12**,
968 1939-1956 (2018).
- 969 75. Travnikov, O. *et al.* Multi-model study of mercury dispersion in the atmosphere: atmospheric
970 processes and model evaluation. *Atmos. Chem. Phys.* **17**, 5271-5295 (2017).
- 971 76. Travnikov, O. & Ilyin, I. in *Mercury Fate and Transport in the Global Atmosphere: Emissions,*
972 *Measurements and Models* (eds Robert Mason & Nicola Pirrone) 571-587 (Springer US, 2009).
- 973 77. Holmes, C. D. *et al.* Global atmospheric model for mercury including oxidation by bromine
974 atoms. *Atmos. Chem. Phys.* **10**, 12037-12057 (2010).
- 975 78. Dastoor, A. P. & Durnford, D. A. Arctic Ocean: Is It a Sink or a Source of Atmospheric Mercury?
976 *Environ. Sci. Technol.* **48**, 1707-1717 (2014).

- 977 79. Fraser, A., Dastoor, A. & Ryjkov, A. How important is biomass burning in Canada to mercury
978 contamination? *Atmos. Chem. Phys.* **18**, 7263-7286 (2018).
- 979 80. Durnford, D. *et al.* How relevant is the deposition of mercury onto snowpacks? – Part 2: A
980 modeling study. *Atmos. Chem. Phys.* **12**, 9251-9274 (2012).
- 981 81. Christensen, J. H., Brandt, J., Frohn, L. M. & Skov, H. Modelling of Mercury in the Arctic with
982 the Danish Eulerian Hemispheric Model. *Atmos. Chem. Phys.* **4**, 2251-2257 (2004).
- 983 82. Skov, H. *et al.* Variability in gaseous elemental mercury at Villum Research Station, Station
984 Nord, in North Greenland from 1999 to 2017. *Atmos. Chem. Phys.* **20**, 13253-13265 (2020).
- 985 83. Cole, A. S. *et al.* Ten-year trends of atmospheric mercury in the high Arctic compared to
986 Canadian sub-Arctic and mid-latitude sites. *Atmos. Chem. Phys.* **13**, 1535-1545 (2013).
- 987 84. Gay, D. A. *et al.* The Atmospheric Mercury Network: measurement and initial examination of an
988 ongoing atmospheric mercury record across North America. *Atmos. Chem. Phys.* **13**, 11339-
989 11349 (2013).
- 990 85. Tørseth, K. *et al.* Introduction to the European Monitoring and Evaluation Programme (EMEP)
991 and observed atmospheric composition change during 1972–2009. *Atmos. Chem. Phys.* **12**,
992 5447-5481 (2012).
- 993 86. Steffen, A. *et al.* A synthesis of atmospheric mercury depletion event chemistry in the atmosphere
994 and snow. *Atmos. Chem. Phys.* **8**, 1445-1482 (2008).
- 995 87. Steffen, A. *et al.* Atmospheric mercury speciation and mercury in snow over time at Alert,
996 Canada. *Atmos. Chem. Phys.* **14**, 2219-2231 (2014).
- 997 88. Toyota, K., Dastoor, A. P. & Ryzhkov, A. Air–snowpack exchange of bromine, ozone and
998 mercury in the springtime Arctic simulated by the 1-D model PHANTAS – Part 2:
999 Mercury and its speciation. *Atmos. Chem. Phys.* **14**, 4135-4167 (2014).
- 1000 89. Sanei, H. *et al.* Wet deposition mercury fluxes in the Canadian sub-Arctic and southern Alberta,
1001 measured using an automated precipitation collector adapted to cold regions. *Atmos. Environ.* **44**,
1002 1672-1681 (2010).
- 1003 90. Sprovieri, F. *et al.* Five-year records of mercury wet deposition flux at GMOS sites in the
1004 Northern and Southern hemispheres. *Atmos. Chem. Phys.* **17**, 2689-2708 (2017).
- 1005 91. Zhou, H., Zhou, C., Hopke, P. K. & Holsen, T. M. Mercury wet deposition and speciated mercury
1006 air concentrations at rural and urban sites across New York state: Temporal patterns, sources and
1007 scavenging coefficients. *Sci. Total Environ.* **637-638**, 943-953 (2018).
- 1008 92. Qin, C., Wang, Y., Peng, Y. & Wang, D. Four-year record of mercury wet deposition in one
1009 typical industrial city in southwest China. *Atmos. Environ.* **142**, 442-451 (2016).
- 1010 93. Douglas, T. A. & Blum, J. D. Mercury Isotopes Reveal Atmospheric Gaseous Mercury
1011 Deposition Directly to the Arctic Coastal Snowpack. *Environ. Sci. Technol. Lett.* **6**, 235-242
1012 (2019).
- 1013 94. Yang, Y., Liu, J. & Wang, Z. Reaction mechanisms and chemical kinetics of mercury
1014 transformation during coal combustion. *Prog. Energy Combust. Sci.* **79**, 100844 (2020).
- 1015 95. Galloway, J. N. & Likens, G. E. The collection of precipitation for chemical analysis. *Tellus* **30**,
1016 71-82 (1978).
- 1017 96. Pearson, C., Howard, D., Moore, C. & Obrist, D. Mercury and trace metal wet deposition across
1018 five stations in Alaska: controlling factors, spatial patterns, and source regions. *Atmos. Chem.*
1019 *Phys.* **19**, 6913-6929 (2019).
- 1020 97. Kochendorfer, J. *et al.* Testing and development of transfer functions for weighing precipitation
1021 gauges in WMO-SPICE. *Hydrol. Earth Syst. Sci.* **22**, 1437-1452 (2018).
- 1022 98. Rasmussen, R. *et al.* How Well Are We Measuring Snow: The NOAA/FAA/NCAR Winter
1023 Precipitation Test Bed. *Bull. Ameri. Meteorol. Soc.* **93**, 811-829 (2012).
- 1024 99. Yang, D., Goodison, B. E., Ishida, S. & Benson, C. S. Adjustment of daily precipitation data at 10
1025 climate stations in Alaska: Application of World Meteorological Organization intercomparison
1026 results. *Water Resour. Res.* **34**, 241-256 (1998).

- 1027 100. Yang, D. An improved precipitation climatology for the Arctic Ocean. *Geophys. Res. Lett.* **26**,
1028 1625-1628 (1999).
- 1029 101. Wang, X., Bao, Z., Lin, C.-J., Yuan, W. & Feng, X. Assessment of Global Mercury Deposition
1030 through Litterfall. *Environ. Sci. Technol.* **50**, 8548-8557 (2016).
- 1031 102. Kirk, J. L. *et al.* Climate Change and Mercury Accumulation in Canadian High and Subarctic
1032 Lakes. *Environ. Sci. Technol.* **45**, 964-970 (2011).
- 1033 103. Lehnerr, I. *et al.* The world's largest High Arctic lake responds rapidly to climate warming.
1034 *Nature Commun.* **9**, 1290 (2018).
- 1035 104. Muir, D. C. G. *et al.* Spatial Trends and Historical Deposition of Mercury in Eastern and
1036 Northern Canada Inferred from Lake Sediment Cores. *Environ. Sci. Technol.* **43**, 4802-4809
1037 (2009).
- 1038 105. Korosi, J. B. *et al.* Long-term changes in organic matter and mercury transport to lakes in the
1039 sporadic discontinuous permafrost zone related to peat subsidence. *Limnol. Oceanogr.* **60**, 1550-
1040 1561 (2015).
- 1041 106. Douglas, T. A. *et al.* A Pulse of Mercury and Major Ions in Snowmelt Runoff from a Small
1042 Arctic Alaska Watershed. *Environ. Sci. Technol.* **15**, 11145-11155 (2017).
- 1043 107. Dommergue, A. *et al.* Deposition of Mercury Species in the Ny-Ålesund Area (79°N) and Their
1044 Transfer during Snowmelt. *Environ. Sci. Technol.* **44**, 901-907 (2010).
- 1045 108. Steffen, A. *et al.* Atmospheric mercury over sea ice during the OASIS-2009 campaign. *Atmos.*
1046 *Chem. Phys.* **13**, 7007-7021 (2013).
- 1047 109. Andersson, M. E., Sommar, J., Gårdfeldt, K. & Lindqvist, O. Enhanced concentrations of
1048 dissolved gaseous mercury in the surface waters of the Arctic Ocean. *Mar. Chem.* **110**, 190-194
1049 (2008).
- 1050 110. Kalinichuk, V. V., Lopatnikov, E. A., Astakhov, A. S., Ivanov, M. V. & Hu, L. Distribution of
1051 atmospheric gaseous elemental mercury (Hg(0)) from the Sea of Japan to the Arctic, and Hg(0)
1052 evasion fluxes in the Eastern Arctic Seas: Results from a joint Russian-Chinese cruise in fall
1053 2018. *Sci. Total Environ.* **753**, 142003 (2021).
- 1054 111. Berg, T., Pfaffhuber, K. A., Cole, A. S., Engelsen, O. & Steffen, A. Ten-year trends in
1055 atmospheric mercury concentrations, meteorological effects and climate variables at Zeppelin,
1056 Ny-Ålesund. *Atmos. Chem. Phys.* **13**, 6575-6586 (2013).
- 1057 112. Wang, X. *et al.* Underestimated Sink of Atmospheric Mercury in a Deglaciated Forest
1058 Chronosequence. *Environ. Sci. Technol.* **54**, 8083-8093 (2020).
- 1059 113. Overland, J. E. Less climatic resilience in the Arctic. *Weather Clim. Extremes* **30**, 100275 (2020).
- 1060 114. Bougoudis, I. *et al.* Long-term time series of Arctic tropospheric BrO derived from UV-VIS
1061 satellite remote sensing and its relation to first-year sea ice. *Atmos. Chem. Phys.* **20**, 11869-11892
1062 (2020).
- 1063 115. Goodsite, M. E., Plane, J. M. C. & Skov, H. A Theoretical Study of the Oxidation of Hg⁰ to
1064 HgBr₂ in the Troposphere. *Environ. Sci. Technol.* **38**, 1772-1776 (2004).
- 1065 116. Goodsite, M. E., Plane, J. M. C. & Skov, H. Correction to A Theoretical Study of the Oxidation
1066 of Hg⁰ to HgBr₂ in the Troposphere. *Environ. Sci. Technol.* **46**, 5262-5262 (2012).
- 1067 117. Shah, V. *et al.* Improved Mechanistic Model of the Atmospheric Redox Chemistry of Mercury.
1068 *Environ. Sci. Technol.* (2021).
- 1069 118. Moore, C. W. *et al.* Convective forcing of mercury and ozone in the Arctic boundary layer
1070 induced by leads in sea ice. *Nature* **506**, 81-84 (2014).
- 1071 119. Douglas, T. A. *et al.* Elevated mercury measured in snow and frost flowers near Arctic sea ice
1072 leads. *Geophys. Res. Lett.* **32** (2005).
- 1073 120. Gleason, J. D. *et al.* Sources and cycling of mercury in the paleo Arctic Ocean from Hg stable
1074 isotope variations in Eocene and Quaternary sediments. *Geochim. Cosmochim. Acta* **197**, 245-262
1075 (2017).
- 1076 121. Masbou, J. *et al.* Hg-Stable Isotope Variations in Marine Top Predators of the Western Arctic
1077 Ocean. *ACS Earth Space Chem.* **2**, 479-490 (2018).

- 1078 122. Olson, C. L., Jiskra, M., Sonke, J. E. & Obrist, D. Mercury in tundra vegetation of Alaska: Spatial
1079 and temporal dynamics and stable isotope patterns. *Sci. Total Environ.* **660**, 1502-1512 (2019).
- 1080 123. Zhou, J. & Obrist, D. Global Mercury Assimilation by Vegetation. *Environ. Sci. Technol.*, In
1081 Press (2021).
- 1082 124. St. Pierre, K. A. *et al.* Importance of Open Marine Waters to the Enrichment of Total Mercury
1083 and Methylmercury in Lichens in the Canadian High Arctic. *Environ. Sci. Technol.* **49**,
1084 5930-5938 (2015).
- 1085 125. Landers, D. H. *et al.* Mercury in vegetation and lake sediments from the US Arctic. *Water Air
1086 and Soil Pollution* **80**, 591-601 (1995).
- 1087 126. Drbal, K., Elster, J. & Komarek, J. Heavy metals in water, ice and biological material from
1088 Spitsbergen, Svalbard. *Polar Res.* **11**, 99-101 (1992).
- 1089 127. Wohlgeuth, L. *et al.* A bottom-up quantification of foliar mercury uptake fluxes across Europe.
1090 *Biogeosciences* **17**, 6441-6456 (2020).
- 1091 128. Schuster, P. F. *et al.* Permafrost Stores a Globally Significant Amount of Mercury. *Geophys. Res.
1092 Lett.* **45**, 1463-1471 (2018).
- 1093 129. Lim, A. G. *et al.* A revised northern soil Hg pool, based on western Siberia permafrost peat Hg
1094 and carbon observations. *Biogeosciences* **17**, 3083-3097 (2020).
- 1095 130. Olson, C., Jiskra, M., Biester, H., Chow, J. & Obrist, D. Mercury in Active-Layer Tundra Soils of
1096 Alaska: Concentrations, Pools, Origins, and Spatial Distribution. *Global Biogeochem. Cycles* **32**,
1097 1058-1073 (2018).
- 1098 131. Halbach, K., Mikkelsen, Ø., Berg, T. & Steinnes, E. The presence of mercury and other trace
1099 metals in surface soils in the Norwegian Arctic. *Chemosphere* **188**, 567-574 (2017).
- 1100 132. Hugelius, G. *et al.* Estimated stocks of circumpolar permafrost carbon with quantified uncertainty
1101 ranges and identified data gaps. *Biogeosciences* **11**, 6573-6593 (2014).
- 1102 133. Durnford, D. & Dastoor, A. The behavior of mercury in the cryosphere: A review of what we
1103 know from observations. *J. Geophys. Res.: Atmos.* **116** (2011).
- 1104 134. Hoyer, M., Burke, J. & Keeler, G. Atmospheric sources, transport and deposition of mercury in
1105 Michigan: Two years of event precipitation. *Water Air Soil Pollut.* **80**, 199-208 (1995).
- 1106 135. Keeler, G. J., Gratz, L. E. & Al-wali, K. Long-term Atmospheric Mercury Wet Deposition at
1107 Underhill, Vermont. *Ecotoxicology* **14**, 71-83 (2005).
- 1108 136. Nelson, S. J. *et al.* A comparison of winter mercury accumulation at forested and no-canopy sites
1109 measured with different snow sampling techniques. *Appl. Geochem.* **23**, 384-398 (2008).
- 1110 137. Kurz, A. Y., Blum, J. D., Johnson, M. W., Nadelhoffer, K. & Zak, D. R. Isotopic composition of
1111 mercury deposited via snow into mid-latitude ecosystems. *Sci. Total Environ.* **784**, 147252
1112 (2021).
- 1113 138. Bargagli, R., Agnorelli, C., Borghini, F. & Monaci, F. Enhanced Deposition and Bioaccumulation
1114 of Mercury in Antarctic Terrestrial Ecosystems Facing a Coastal Polynya. *Environ. Sci. Technol.*
1115 **39**, 8150-8155 (2005).
- 1116 139. Sherman, L. S., Blum, J. D., Douglas, T. A. & Steffen, A. Frost flowers growing in the Arctic
1117 ocean-atmosphere–sea ice–snow interface: 2. Mercury exchange between the atmosphere, snow,
1118 and frost flowers. *J. Geophys. Res.: Atmos.* **117**, D00R10 (2012).
- 1119 140. Douglas, T. A. *et al.* Influence of Snow and Ice Crystal Formation and Accumulation on Mercury
1120 Deposition to the Arctic. *Environ. Sci. Technol.* **42**, 1542-1551 (2008).
- 1121 141. Domine, F. *et al.* The specific surface area and chemical composition of diamond dust near
1122 Barrow, Alaska. *J. Geophys. Res.: Atmos.* **116**, D00R06 (2011).
- 1123 142. Xu, W., Tenuta, M. & Wang, F. Bromide and chloride distribution across the snow-sea ice-ocean
1124 interface: A comparative study between an Arctic coastal marine site and an experimental sea ice
1125 mesocosm. *J. Geophys. Res.: Oceans* **121**, 5535-5548 (2016).
- 1126 143. Lalonde, J. D., Poulain, A. J. & Amyot, M. The Role of Mercury Redox Reactions in Snow on
1127 Snow-to-Air Mercury Transfer. *Environ. Sci. Technol.* **36**, 174-178 (2002).

- 1128 144. Poulain, A. J. *et al.* Redox transformations of mercury in an Arctic snowpack at springtime.
1129 *Atmos. Environ.* **38**, 6763-6774 (2004).
- 1130 145. Dommergue, A. *et al.* The fate of mercury species in a sub-arctic snowpack during snowmelt.
1131 *Geophys. Res. Lett.* **30**, 1621 (2003).
- 1132 146. St. Louis, V. L. *et al.* Some sources and sinks of monomethyl and inorganic mercury on
1133 Ellesmere island in the Canadian high arctic. *Environ. Sci. Technol.* **39**, 2686-2701 (2005).
- 1134 147. Faïn, X. *et al.* Mercury in the snow and firn at Summit Station, Central Greenland, and
1135 implications for the study of past atmospheric mercury levels. *Atmos. Chem. Phys.* **8**, 3441-3457
1136 (2008).
- 1137 148. Ferrari, C. P. *et al.* Snow-to-air exchanges of mercury in an Arctic seasonal snow pack in Ny-
1138 Ålesund, Svalbard. *Atmos. Environ.* **39**, 7633-7645 (2005).
- 1139 149. Johnson, K. P., Blum, J. D., Keeler, G. J. & Douglas, T. A. Investigation of the deposition and
1140 emission of mercury in arctic snow during an atmospheric mercury depletion event. *J. Geophys.*
1141 *Res.: Atmos.* **113** (2008).
- 1142 150. Kamp, J., Skov, H., Jensen, B. & Sørensen, L. L. Fluxes of gaseous elemental mercury (GEM) in
1143 the High Arctic during atmospheric mercury depletion events (AMDEs). *Atmos. Chem. Phys.* **18**,
1144 6923-6938 (2018).
- 1145 151. Mann, E. A. *et al.* Photoreducible Mercury Loss from Arctic Snow Is Influenced by Temperature
1146 and Snow Age. *Environ. Sci. Technol.* **49**, 12120-12126 (2015).
- 1147 152. Boutron, C. F., Vandal, G. M., Fitzgerald, W. F. & Ferrari, C. P. A forty year record of Mercury
1148 in central Greenland snow. *Geophys. Res. Lett.* **25**, 3315-3318 (1998).
- 1149 153. Brooks, S. *et al.* Temperature and sunlight controls of mercury oxidation and deposition atop the
1150 Greenland ice sheet. *Atmos. Chem. Phys.* **11**, 8295-8306 (2011).
- 1151 154. Zheng, J. Archives of total mercury reconstructed with ice and snow from Greenland and the
1152 Canadian High Arctic. *Sci. Total Environ.* **509-510**, 133-144 (2015).
- 1153 155. Farinotti, D. *et al.* A consensus estimate for the ice thickness distribution of all glaciers on Earth.
1154 *Nat. Geosci.* **12**, 168-173 (2019).
- 1155 156. Forsberg, R., Sørensen, L. & Simonsen, S. in *Integrative Study of the Mean Sea Level and Its*
1156 *Components* (eds Anny Cazenave, Nicolas Champollion, Frank Paul, & Jérôme Benveniste) 91-
1157 106 (Springer International Publishing, 2017).
- 1158 157. Ciraci, E., Velicogna, I. & Swenson, S. Continuity of the Mass Loss of the World's Glaciers and
1159 Ice Caps From the GRACE and GRACE Follow-On Missions. *Geophys. Res. Lett.* **47**,
1160 e2019GL086926 (2020).
- 1161 158. Hawkings, J. R. *et al.* Large subglacial source of mercury from the southwestern margin of the
1162 Greenland Ice Sheet. *Nat. Geosci.* **14**, 496-502 (2021).
- 1163 159. Friske, P. W. B. *et al.* (ed Geological Survey of Canada) (1994).
- 1164 160. Halm, D. R. & Dornblaser, M. M. Water and Sediment Quality in the Yukon River and its
1165 Tributaries Between Atlin, British Columbia, Canada, and Eagle, Alaska, USA, 2004. Report No.
1166 2007-1197, (2007).
- 1167 161. Nagorski, S. A., Vermilyea, A. W. & Lamborg, C. H. Mercury export from glacierized Alaskan
1168 watersheds as influenced by bedrock geology, watershed processes, and atmospheric deposition.
1169 *Geochim. Cosmochim. Acta* **304**, 32-49 (2021).
- 1170 162. Søndergaard, J., Riget, F., Tamstorf, M. P. & Larsen, M. M. Mercury Transport in a Low-Arctic
1171 River in Kobbefjord, West Greenland (64A degrees N). *Water Air Soil Pollut.* **223**, 4333-4342
1172 (2012).
- 1173 163. Overeem, I. *et al.* Substantial export of suspended sediment to the global oceans from glacial
1174 erosion in Greenland. *Nat. Geosci.* **10**, 859-863 (2017).
- 1175 164. Pfeffer, W. T. *et al.* The Randolph Glacier Inventory: a globally complete inventory of glaciers. *J.*
1176 *Glaciol.* **60**, 537-552 (2014).
- 1177 165. Zolkos, S. *et al.* Mercury Export from Arctic Great Rivers. *Environ. Sci. Technol.* **54**, 4140-4148
1178 (2020).

- 1179 166. Leitch, D. R. *et al.* The delivery of mercury to the Beaufort Sea of the Arctic Ocean by the
1180 Mackenzie River. *Sci. Total Environ.* **373**, 178-195 (2007).
- 1181 167. Lim, A. G. *et al.* Enhanced particulate Hg export at the permafrost boundary, western Siberia.
1182 *Environ. Pollut.* **254**, 113083 (2019).
- 1183 168. Schuster, P. F. *et al.* Mercury Export from the Yukon River Basin and Potential Response to a
1184 Changing Climate. *Environ. Sci. Technol.* **45**, 9262-9267 (2011).
- 1185 169. Emmerton, C. A. *et al.* Mercury Export to the Arctic Ocean from the Mackenzie River, Canada.
1186 *Environ. Sci. Technol.* **47**, 7644-7654 (2013).
- 1187 170. Walvoord, M. A. & Striegl, R. G. Increased groundwater to stream discharge from permafrost
1188 thawing in the Yukon River basin: Potential impacts on lateral export of carbon and nitrogen.
1189 *Geophys. Res. Lett.* **34**, L12402 (2007).
- 1190 171. Vonk, J. E. *et al.* Reviews and syntheses: Effects of permafrost thaw on Arctic aquatic
1191 ecosystems. *Biogeosciences* **12**, 7129-7167 (2015).
- 1192 172. Tank, S. E. *et al.* Landscape matters: Predicting the biogeochemical effects of permafrost thaw on
1193 aquatic networks with a state factor approach. *Permafrost Periglacial Processes* **31**, 358-370
1194 (2020).
- 1195 173. Sukhenko, S. A., Papina, T. S. & Pozdnjakov, S. R. Transport of mercury by the Katun river,
1196 West Siberia. *Hydrobiologia* **228**, 23-28 (1992).
- 1197 174. Fedorov, Y. A. *et al.* Patterns of mercury distribution in bottom sediments along the Severnaya
1198 Dvina-White Sea section. *Dokl. Earth Sci.* **436**, 51-54 (2011).
- 1199 175. Bliss, A., Hock, R. & Radić, V. Global response of glacier runoff to twenty-first century climate
1200 change. *J. Geophys. Res.: Earth Surf.* **119**, 717-730 (2014).
- 1201 176. Delaney, I. & Adhikari, S. Increased Subglacial Sediment Discharge in a Warming Climate:
1202 Consideration of Ice Dynamics, Glacial Erosion, and Fluvial Sediment Transport. *Geophys. Res.*
1203 *Lett.* **47**, e2019GL085672 (2020).
- 1204 177. van Pelt, W. J. J., Schuler, T. V., Pohjola, V. A. & Pettersson, R. Accelerating future mass loss of
1205 Svalbard glaciers from a multi-model ensemble. *J. Glaciol.* **67**, 485-499 (2021).
- 1206 178. Muntjewerf, L. *et al.* Accelerated Greenland Ice Sheet Mass Loss Under High Greenhouse Gas
1207 Forcing as Simulated by the Coupled CESM2.1-CISM2.1. *J. Adv. Model. Earth Syst.* **12**,
1208 e2019MS002031 (2020).
- 1209 179. Mu, C. *et al.* Carbon and mercury export from the Arctic rivers and response to permafrost
1210 degradation. *Water Res.* **161**, 54-60 (2019).
- 1211 180. Overduin, P. P. *et al.* Coastal changes in the Arctic. *Geological Society, London, Special*
1212 *Publications* **388**, 103 (2014).
- 1213 181. Gibbs, A. E., Ohman, K. A. & Richmond, B. M. (ed United States Geological Survey) (2015).
- 1214 182. Couture, N. J., Irrgang, A., Pollard, W., Lantuit, H. & Fritz, M. Coastal Erosion of Permafrost
1215 Soils Along the Yukon Coastal Plain and Fluxes of Organic Carbon to the Canadian Beaufort
1216 Sea. *J. Geophys. Res.: Biogeosci.* **123**, 406-422 (2018).
- 1217 183. Leitch, D. R. *Mercury Distribution in Water and Permafrost of the Lower Mackenzie Basin,*
1218 *Their Contribution to the Mercury Contamination in the Beaufort Sea Marine Ecosystem, and*
1219 *Potential Effects of Climate Variation* Master of Science thesis, University of Manitoba, (2006).
- 1220 184. Outridge, P. M. & Sanei, H. Does organic matter degradation affect the reconstruction of pre-
1221 industrial atmospheric mercury deposition rates from peat cores? — A test of the hypothesis
1222 using a permafrost peat deposit in northern Canada. *Int. J. Coal Geol.* **83**, 73-81 (2010).
- 1223 185. Lantuit, H. *et al.* The Arctic Coastal Dynamics Database: A New Classification Scheme and
1224 Statistics on Arctic Permafrost Coastlines. *Estuaries Coasts* **35**, 383-400 (2012).
- 1225 186. Irrgang, A. M. *et al.* Variability in Rates of Coastal Change Along the Yukon Coast, 1951 to
1226 2015. *J. Geophys. Res.: Earth Surf.* **123**, 779-800 (2018).
- 1227 187. Wang, F., Macdonald, R. W., Armstrong, D. A. & Stern, G. A. Total and Methylated Mercury in
1228 the Beaufort Sea: The Role of Local and Recent Organic Remineralization. *Environ. Sci. Technol.*
1229 **46**, 11821-11828 (2012).

- 1230 188. Lehnherr, I., St Louis, V. L., Hintelmann, H. & Kirk, J. L. Methylation of inorganic mercury in
1231 polar marine waters. *Nat. Geosci.* **4**, 298-302 (2011).
- 1232 189. Kirk, J. L. *et al.* Methylated Mercury Species in Marine Waters of the Canadian High and Sub
1233 Arctic. *Environ. Sci. Technol.* **42**, 8367-8373 (2008).
- 1234 190. St. Louis, V. L. *et al.* Methylated Mercury Species in Canadian High Arctic Marine Surface
1235 Waters and Snowpacks. *Environ. Sci. Technol.* **41**, 6433-6441 (2007).
- 1236 191. Tesán Onrubia, J. A. *et al.* Mercury Export Flux in the Arctic Ocean Estimated from ²³⁴Th/²³⁸U
1237 Disequilibria. *ACS Earth Space Chem.* **4**, 795-801 (2020).
- 1238 192. Heimbürger, L.-E. *et al.* Shallow methylmercury production in the marginal sea ice zone of the
1239 central Arctic Ocean. *Sci. Rep.* **5**, 10318 (2015).
- 1240 193. Cossa, D. *et al.* Mercury distribution and transport in the North Atlantic Ocean along the
1241 GEOTRACES-GA01 transect. *Biogeosciences* **15**, 2309-2323 (2018).
- 1242 194. Charette, M. A. *et al.* The Transpolar Drift as a Source of Riverine and Shelf-Derived Trace
1243 Elements to the Central Arctic Ocean. *J. Geophys. Res.: Oceans* **125**, e2019JC015920 (2020).
- 1244 195. Bowman, K. L., Lamborg, C. H. & Agather, A. M. A global perspective on mercury cycling in
1245 the ocean. *Sci. Total Environ.* **710**, 136166 (2020).
- 1246 196. Kim, H. *et al.* Contrasting distributions of dissolved gaseous mercury concentration and evasion
1247 in the North Pacific Subarctic Gyre and the Subarctic Front. *Deep Sea Res. Part I* **110**, 90-98
1248 (2016).
- 1249 197. Outridge, P. M., Macdonald, R. W., Wang, F., Stern, G. A. & Dastoor, A. P. A mass balance
1250 inventory of mercury in the Arctic Ocean. *Environ. Chem.* **5**, 89-111 (2008).
- 1251 198. Parkinson, C. L. & Cavalieri, D. J. Arctic sea ice variability and trends, 1979–2006. *J. Geophys.*
1252 *Res.: Oceans* **113**, C07003 (2008).
- 1253 199. Cavalieri, D. J. & Parkinson, C. L. Arctic sea ice variability and trends, 1979-2010. *Cryosphere* **6**,
1254 881-889 (2012).
- 1255 200. Arrigo, K. R. & van Dijken, G. L. Continued increases in Arctic Ocean primary production. *Prog.*
1256 *Oceanogr.* **136**, 60-70 (2015).
- 1257 201. Hu, H. *et al.* Mercury Reduction and Cell-Surface Adsorption by *Geobacter sulfurreducens* PCA.
1258 *Environ. Sci. Technol.* **47**, 10922-10930 (2013).
- 1259 202. Møller, A. K. *et al.* Mercuric reductase genes (*merA*) and mercury resistance plasmids in High
1260 Arctic snow, freshwater and sea-ice brine. *FEMS Microbiol. Ecol.* **87**, 52-63 (2014).
- 1261 203. Whalin, L., Kim, E.-H. & Mason, R. Factors influencing the oxidation, reduction, methylation
1262 and demethylation of mercury species in coastal waters. *Mar. Chem.* **107**, 278-294 (2007).
- 1263 204. Zhang, Y., Soerensen, A. L., Schartup, A. T. & Sunderland, E. M. A Global Model for
1264 Methylmercury Formation and Uptake at the Base of Marine Food Webs. *Global Biogeochem.*
1265 *Cycles* **34**, e2019GB006348 (2020).
- 1266 205. Chaulk, A., Stern, G. A., Armstrong, D., Barber, D. G. & Wang, F. Mercury Distribution and
1267 Transport Across the Ocean–Sea-Ice–Atmosphere Interface in the Arctic Ocean. *Environ. Sci.*
1268 *Technol.* **45**, 1866-1872 (2011).
- 1269 206. Beattie, S. A. *et al.* Total and Methylated Mercury in Arctic Multiyear Sea Ice. *Environ. Sci.*
1270 *Technol.* **48**, 5575-5582 (2014).
- 1271 207. Klunder, M. B. *et al.* Dissolved iron in the Arctic shelf seas and surface waters of the central
1272 Arctic Ocean: Impact of Arctic river water and ice-melt. *J. Geophys. Res.: Oceans* **117**, C01027
1273 (2012).
- 1274 208. Cossa, D. *et al.* Mercury in the Southern Ocean. *Geochim. Cosmochim. Acta* **75**, 4037-4052
1275 (2011).
- 1276 209. Wang, F., Pućko, M. & Stern, G. Transport and transformation of contaminants in sea ice. *Sea*
1277 *Ice*, 472-491 (2017).
- 1278 210. Tsubouchi, T. *et al.* The Arctic Ocean Seasonal Cycles of Heat and Freshwater Fluxes:
1279 Observation-Based Inverse Estimates. *J. Phys. Oceanogr.* **48**, 2029-2055 (2018).

- 1280 211. Østerhus, S. *et al.* Arctic Mediterranean exchanges: a consistent volume budget and trends in
1281 transports from two decades of observations. *Ocean Sci.* **15**, 379-399 (2019).
- 1282 212. Rachold, V. *et al.* in *The Organic Carbon Cycle in the Arctic Ocean* (eds Ruediger Stein &
1283 Robie W. MacDonald) 33-55 (Springer Berlin Heidelberg, 2004).
- 1284 213. Gamboa, A., Montero-Serrano, J.-C., St-Onge, G., Rochon, A. & Desiagne, P.-A. Mineralogical,
1285 geochemical, and magnetic signatures of surface sediments from the Canadian Beaufort Shelf and
1286 Amundsen Gulf (Canadian Arctic). *Geochem. Geophys. Geosyst.* **18**, 488-512 (2017).
- 1287 214. Lamborg, C. H., Hammerschmidt, C. R. & Bowman, K. L. An examination of the role of particles
1288 in oceanic mercury cycling. *Philos Trans A Math Phys Eng Sci* **374**, 20150297 (2016).
- 1289 215. Pućko, M. *et al.* Transformation of Mercury at the Bottom of the Arctic Food Web: An
1290 Overlooked Puzzle in the Mercury Exposure Narrative. *Environ. Sci. Technol.* **48**, 7280-7288
1291 (2014).
- 1292 216. Hayes, C. T. *et al.* Global Ocean Sediment Composition and Burial Flux in the Deep Sea. *Global*
1293 *Bigeochem. Cycles* **35**, e2020GB006769 (2021).
- 1294 217. Aksentov, K. I. *et al.* Assessment of mercury levels in modern sediments of the East Siberian Sea.
1295 *Mar. Pollut. Bull.* **168**, 112426 (2021).
- 1296 218. MacMillan, G. A., Girard, C., Chételat, J., Laurion, I. & Amyot, M. High Methylmercury in
1297 Arctic and Subarctic Ponds is Related to Nutrient Levels in the Warming Eastern Canadian
1298 Arctic. *Environ. Sci. Technol.* **49**, 7743-7753 (2015).
- 1299 219. Pelletier, N., Chételat, J., Blarquez, O. & Vermaire, J. C. Paleolimnological Assessment of
1300 Wildfire-Derived Atmospheric Deposition of Trace Metal(loid)s and Major Ions to Subarctic
1301 Lakes (Northwest Territories, Canada). *J. Geophys. Res.: Biogeosci.* **125**, e2020JG005720
1302 (2020).
- 1303 220. Ivanov, V. V., Shapiro, G. I., Huthnance, J. M., Aleynik, D. L. & Golovin, P. N. Cascades of
1304 dense water around the world ocean. *Prog. Oceanogr.* **60**, 47-98 (2004).
- 1305 221. Roeske, T., Loeff, M. R. v., Middag, R. & Bakker, K. Deep water circulation and composition in
1306 the Arctic Ocean by dissolved barium, aluminium and silicate. *Mar. Chem.* **132-133**, 56-67
1307 (2012).
- 1308 222. Bianchi, T. S. The role of terrestrially derived organic carbon in the coastal ocean: A changing
1309 paradigm and the priming effect. *PNAS* **108**, 19473 (2011).
- 1310 223. Rontani, J.-F. *et al.* Degradation of sterols and terrigenous organic matter in waters of the
1311 Mackenzie Shelf, Canadian Arctic. *Org. Geochem.* **75**, 61-73 (2014).
- 1312 224. Custodio, D., Ebinghaus, R., Spain, T. G. & Bieser, J. Source apportionment of atmospheric
1313 mercury in the remote marine atmosphere: Mace Head GAW station, Irish western coast. *Atmos.*
1314 *Chem. Phys.* **20**, 7929-7939 (2020).
- 1315 225. Vermilyea, A. W. *et al.* Continuous proxy measurements reveal large mercury fluxes from glacial
1316 and forested watersheds in Alaska. *Sci. Total Environ.* **599-600**, 145-155 (2017).
- 1317 226. Hare, A., Stern, G. A., Macdonald, R. W., Kuzyk, Z. Z. & Wang, F. Contemporary and
1318 preindustrial mass budgets of mercury in the Hudson Bay Marine System: The role of sediment
1319 recycling. *Sci. Total Environ.* **406**, 190-204 (2008).
- 1320 227. Kirk, J. L. & St. Louis, V. L. Multiyear Total and Methyl Mercury Exports from Two Major Sub-
1321 Arctic Rivers Draining into Hudson Bay, Canada. *Environ. Sci. Technol.* **43**, 2254-2261 (2009).
- 1322 228. Semkin, R. G., Mierle, G. & Neureuther, R. J. Hydrochemistry and mercury cycling in a High
1323 Arctic watershed. *Sci. Total Environ.* **342**, 199-221 (2005).
- 1324 229. Haine, T. W. N. *et al.* Arctic freshwater export: Status, mechanisms, and prospects. *Global*
1325 *Planet. Change* **125**, 13-35 (2015).
- 1326 230. Mason, R. P. *et al.* Mercury biogeochemical cycling in the ocean and policy implications.
1327 *Environ. Res.* **119**, 101-117 (2012).
- 1328 231. Bravo, A. G. & Cosio, C. Biotic formation of methylmercury: A bio-physico-chemical
1329 conundrum. *Limnol. Oceanogr.* **65**, 1010-1027 (2020).

- 1330 232. Durnford, D., Dastoor, A., Figuera-Nieto, D. & Ryjkov, A. Long range transport of mercury to
1331 the Arctic and across Canada. *Atmos. Chem. Phys.* **10**, 6063-6086 (2010).
- 1332 233. Burt, A. *et al.* Mercury uptake within an ice algal community during the spring bloom in first-
1333 year Arctic sea ice. *J. Geophys. Res.: Oceans* **118**, 4746-4754 (2013).
- 1334 234. Villar, E., Cabrol, L. & Heimbürger-Boavida, L.-E. Widespread microbial mercury methylation
1335 genes in the global ocean. *Environ. Microbiol. Rep.* **12**, 277-287 (2020).
- 1336 235. Gilmour, C. C. *et al.* Mercury Methylation by Novel Microorganisms from New Environments.
1337 *Environ. Sci. Technol.* **47**, 11810-11820 (2013).
- 1338 236. Lee, C.-S. & Fisher, N. S. Methylmercury uptake by diverse marine phytoplankton. *Limnol.*
1339 *Oceanogr.* **61**, 1626-1639 (2016).
- 1340 237. Schartup, A. T. *et al.* A Model for Methylmercury Uptake and Trophic Transfer by Marine
1341 Plankton. *Environ. Sci. Technol.* **52**, 654-662 (2018).
- 1342 238. Wu, P. *et al.* The importance of bioconcentration into the pelagic food web base for
1343 methylmercury biomagnification: A meta-analysis. *Sci. Total Environ.* **646**, 357-367 (2019).

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Figures

Arctic mercury cycling

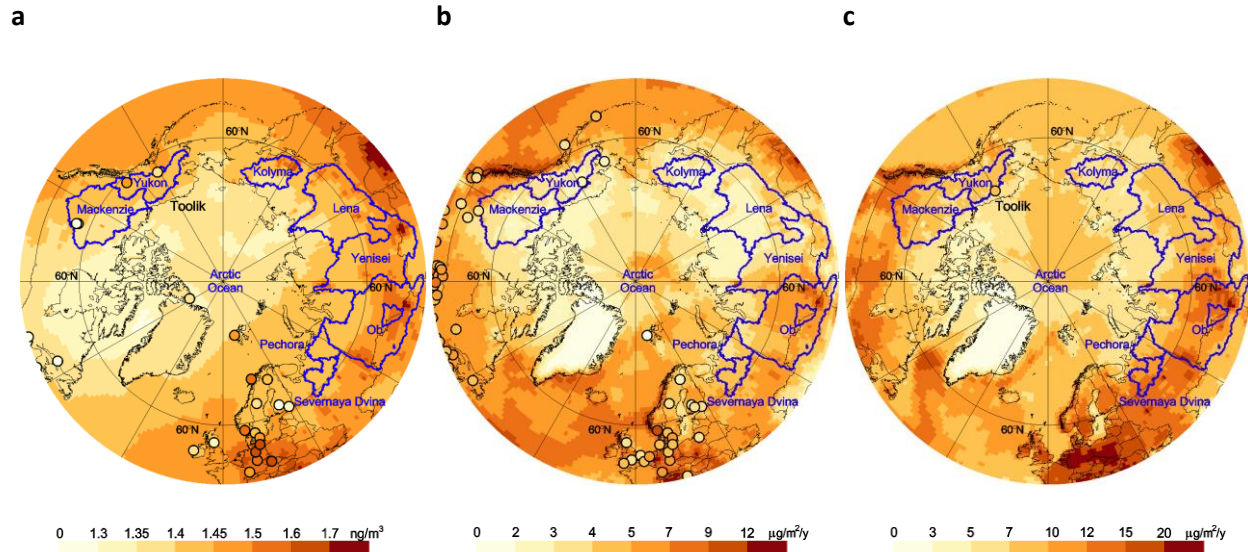


Figure 1. | Atmospheric Hg distribution in the Arctic. **a**) Model ensemble (DEHM, GEM-MACH-Hg, GLEMOS, GOES-Chem) simulated (this Review) annual average surface air gaseous elemental mercury (Hg(0)) concentrations in 2015. **b**) Annual mercury (Hg) wet deposition flux in 2015. **c**) Annual Hg total deposition in the Arctic in 2015. Circles show observations in the same color scale. The blue lines delineate major pan-Arctic river watersheds. Air concentrations and wet deposition observations are from ECCC-AMM⁸³, AMNet⁸⁴, EMEP⁸⁵ and REF²²⁴, and total deposition observation is from REF²⁹.

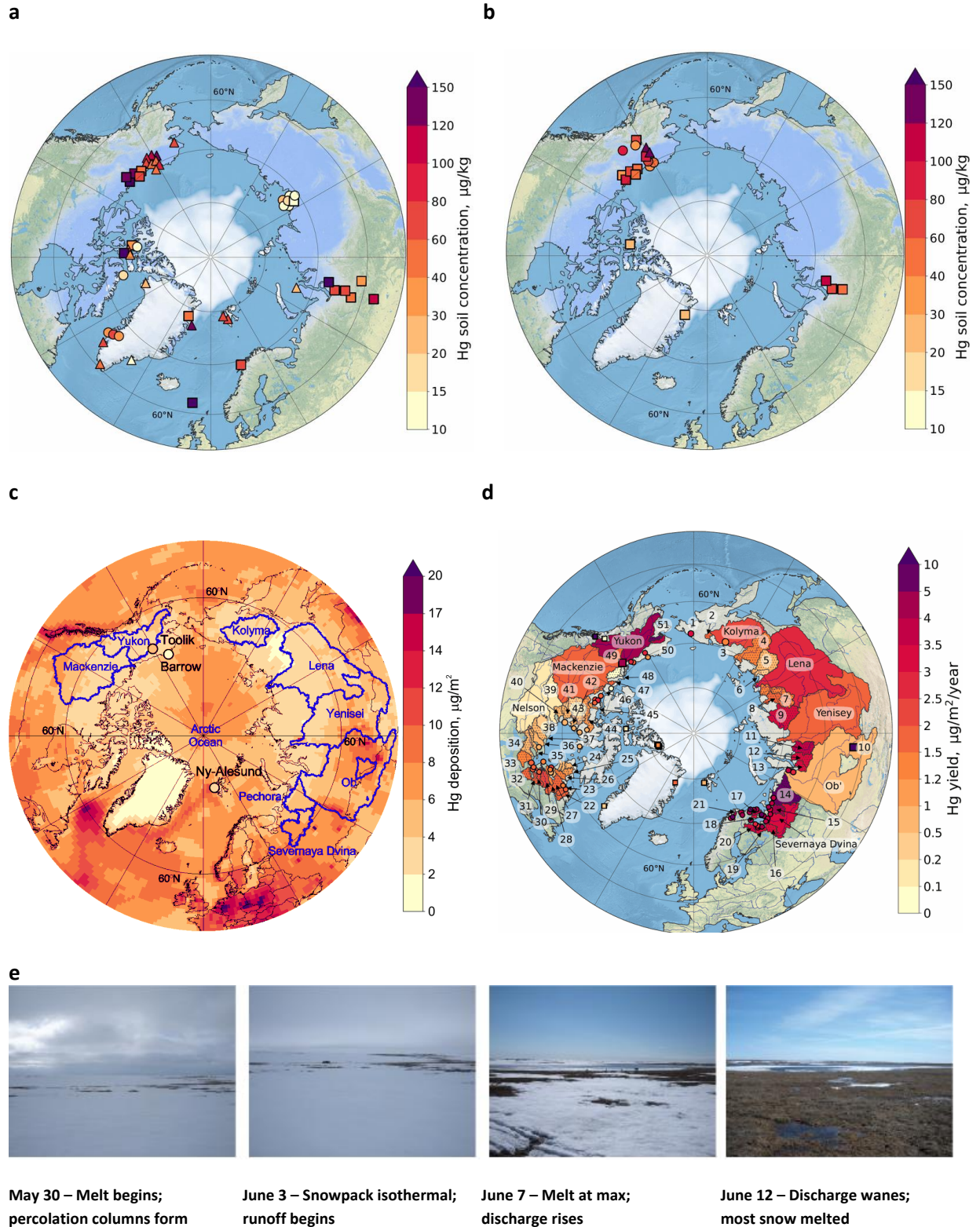


Figure 2. | Distribution of total Hg in Arctic soils, wintertime deposition and rivers. a| Mean mercury (Hg) concentrations of active layer soils within 0-1 m depth. b| Mean Hg concentration of permafrost

layer soils within 0-1 m depth. Symbols in Figs. a-b represent the soil type with squares for organic soils, circles for mineral soils and triangles for combined organic/mineral soils c| Wintertime (September-May) model ensemble simulated Hg deposition to snowpack. Observations are shown in circles d| Annual river total mercury (THg) yields (annual riverine THg mass flux to the Arctic Ocean divided by watershed area) for pan-Arctic watersheds. Larger watersheds (> 10,000 km²) are delineated where possible, while smaller watersheds are represented by symbols. Solid colored watersheds and squares represent measured THg yields (n=32) reported in REF^{7,16-18,107,162,165,169,173,225-228}. Hatched watersheds and circles are THg yields (n=100) modelled in REF⁷. Numbered watersheds are: 1) Amguema, 2) Palyavaam, 3) Bolshoy Anyuy, 4) Indigirka, 5) Yana, 6) Omoloy, 7) Olenek, 8) Anabar, 9) Khatanga, 10) Katun, 11) Taz, 12) Pur, 13) Nadym, 14) Pechora, 15) Mezen, 16) Onega, 17) Ponoy, 18) Varzuga, 19) Nyzhny Vig (Sor), 20) Kem, 21) Tana, 22) George, 23) À la Baleine, 24) Povungnituk, 25) Arnaud, 26) Aux Feuilles, 27) Koksoak, 28) Grande Rivière de la Baleine, 29) La Grande Rivière, 30) Eastmain, 31) Rupert, 32) Nottaway, 33) Moose, 34) Albany, 35) Attawapiskat, 36) Winisk, 37) Severn, 38) Hayes, 39) Churchill, 40) Seal, 41) Thlewiaza, 42) Ferguson, 43) Thelon, 44) Back, 45) Ellice, 46) Coppermine, 47) Hornaday, 48) Anderson, 49) Peel, 50) Noatak, 51) Kobuk. For rivers with multiple yield estimates, only the estimate for the most recent time period is shown. e| Repeat images during spring melt in Utqiagvik (formerly Barrow), Alaska during a typical spring melt. In a matter of a few weeks the tundra surface goes from completely snow covered to a wet vegetation and soil surface exposed to continuous sunlight.

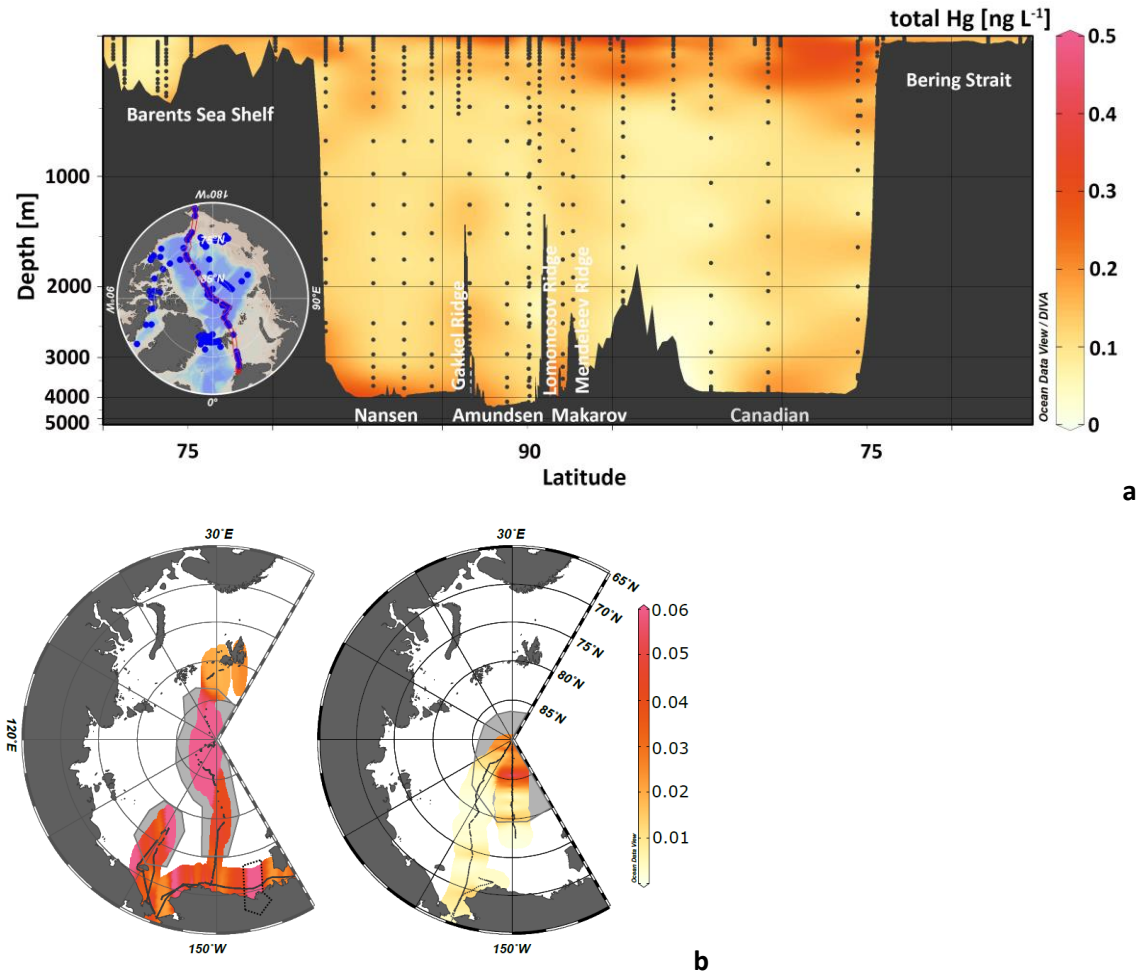


Figure 3. | Spatial distribution of Hg in the Arctic Ocean. **a**) Synoptic pan-Arctic transect of total Hg concentrations during the 2015 GEOTRACES cruises (ng L^{-1})^{22,191,194}. The insert map shows the transect (red band) and available Hg observations (blue dots). **b**) Surface water Hg(0) concentrations (ng L^{-1}) in the Arctic Ocean during a summer (left¹⁰⁹) and fall (right¹⁹) cruise. Grey areas indicate ice-covered water. The area indicated with a punctuated line represents the outflow region from the Mackenzie River Basin. Ocean currents transport Hg via waters entering the AO from the Atlantic Ocean ($235 \pm 31.7 \times 10^3 \text{ km}^3 \text{ y}^{-1}$, Fram Strait; $72.9 \pm 38.0 \times 10^3 \text{ km}^3 \text{ y}^{-1}$, Barents Sea Opening), and the Pacific Ocean ($22.2 \pm 22.2 \times 10^3 \text{ km}^3 \text{ y}^{-1}$, Bering Strait), and exiting the AO through the Davis Strait ($66.6 \pm 22.2 \times 10^3 \text{ km}^3 \text{ y}^{-1}$) and Fram Strait ($273 \pm 114 \times 10^3 \text{ km}^3 \text{ y}^{-1}$) into the Atlantic Ocean^{210,229}.

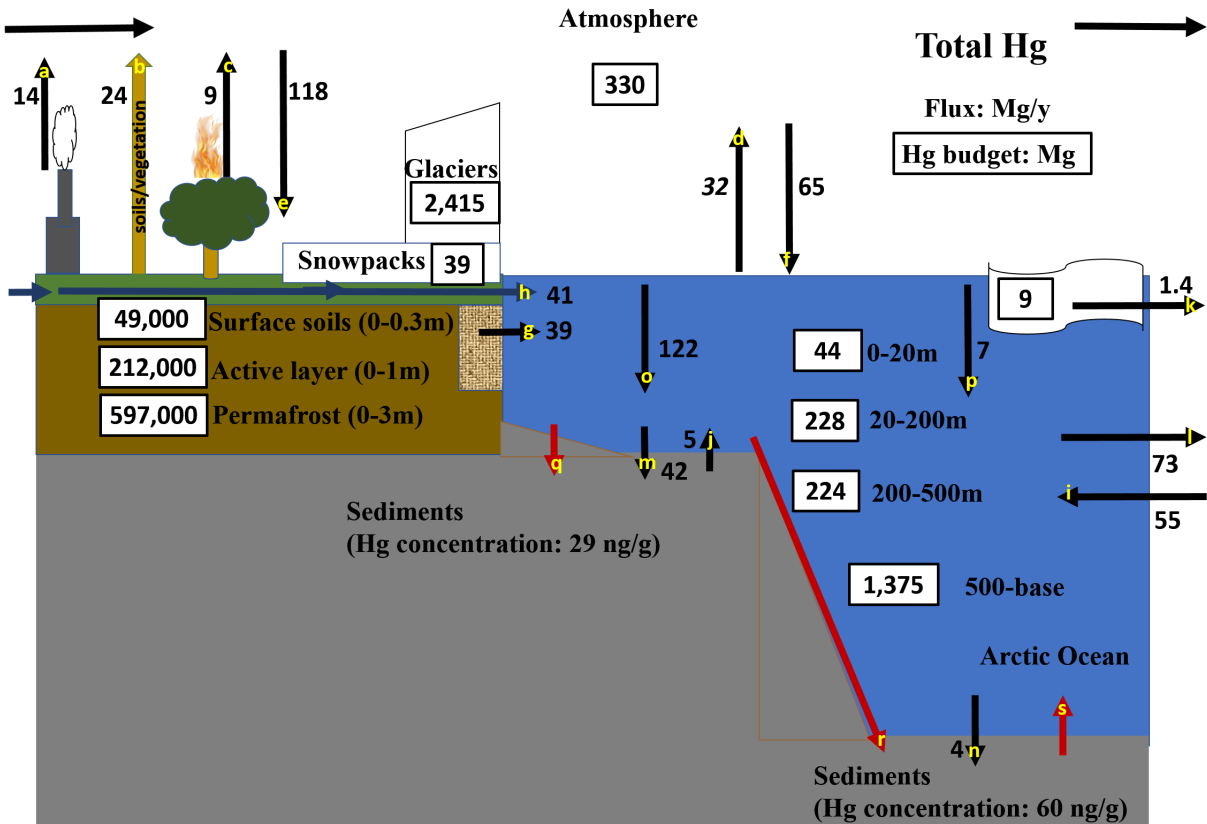
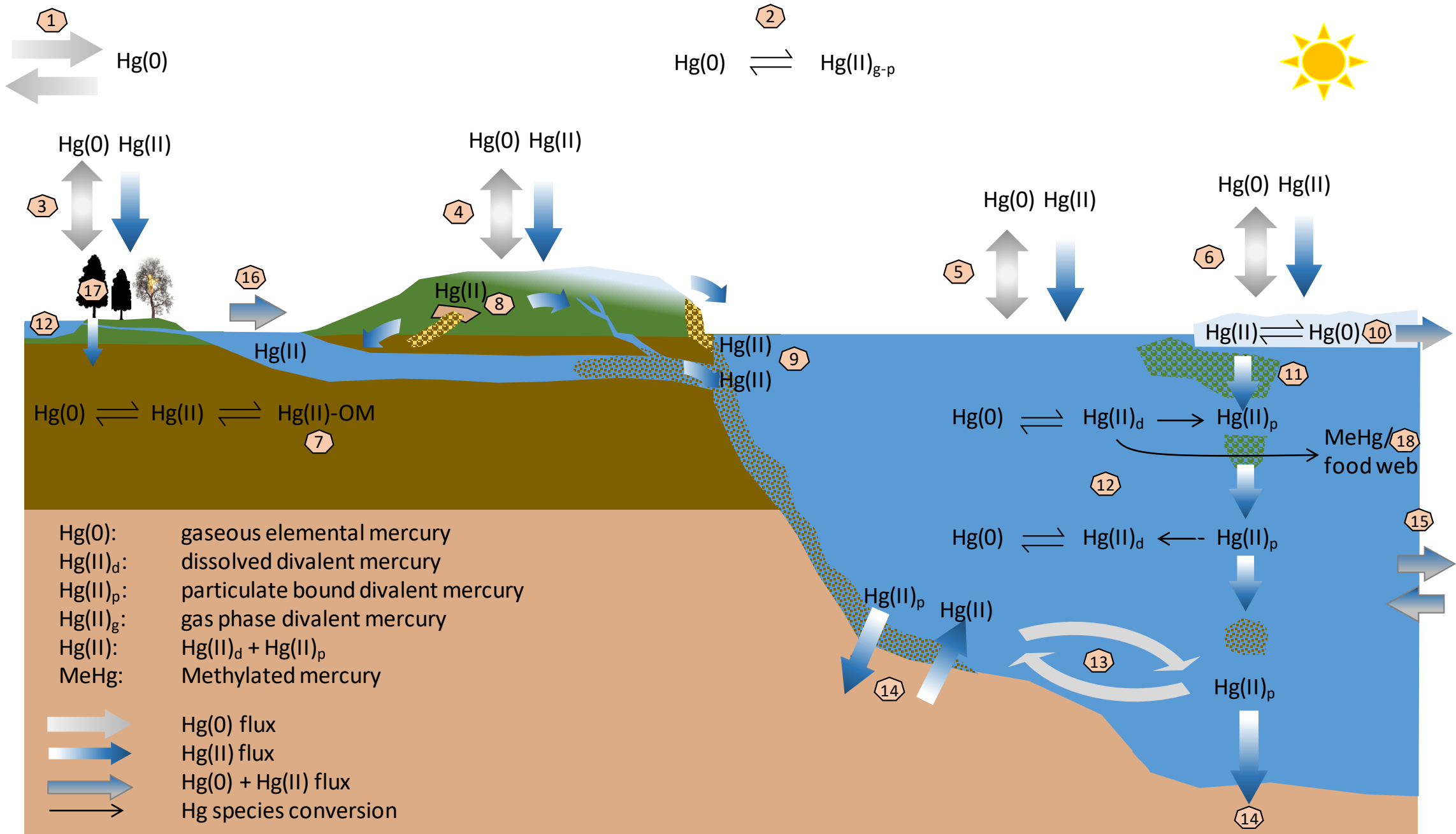
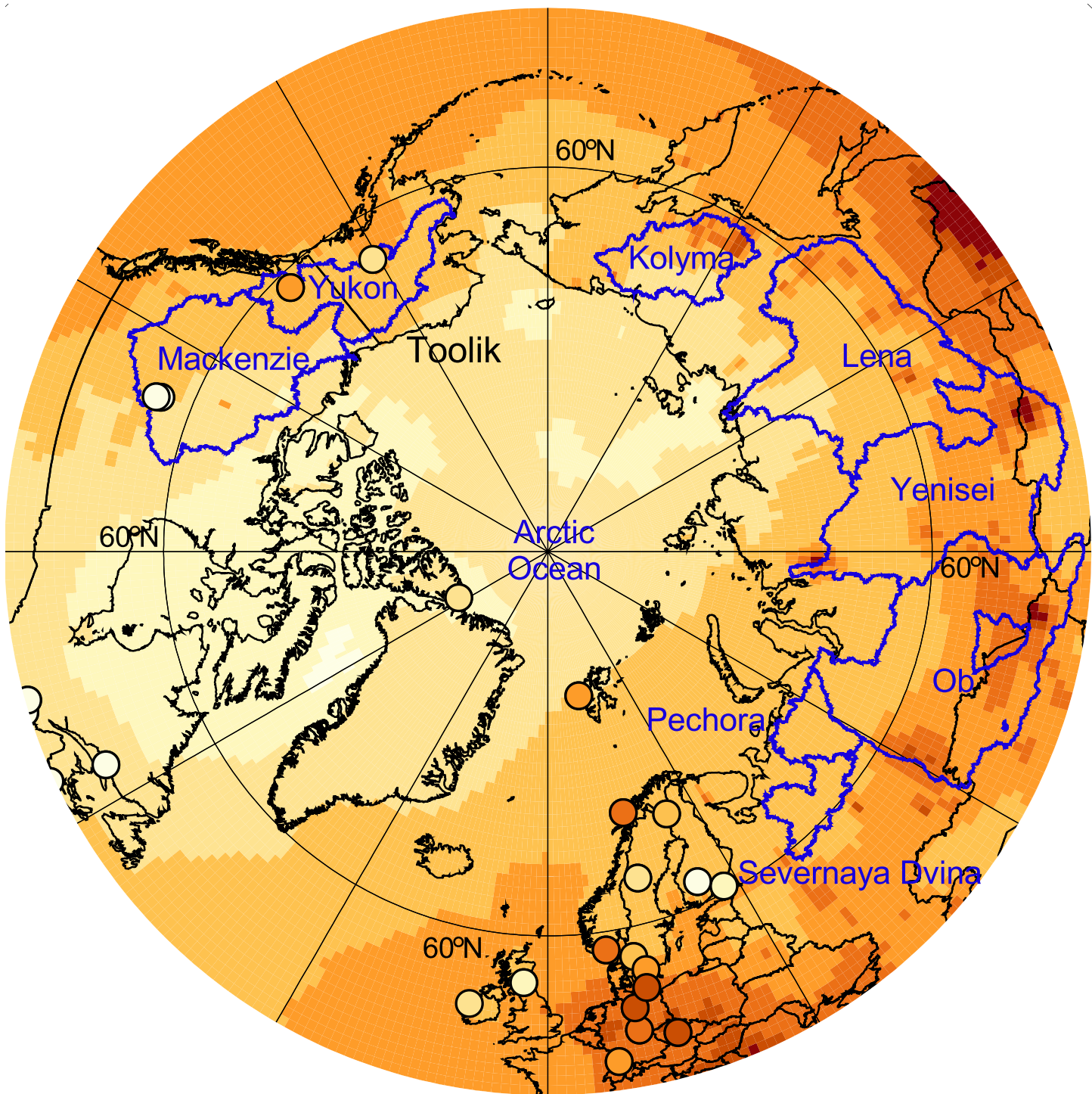
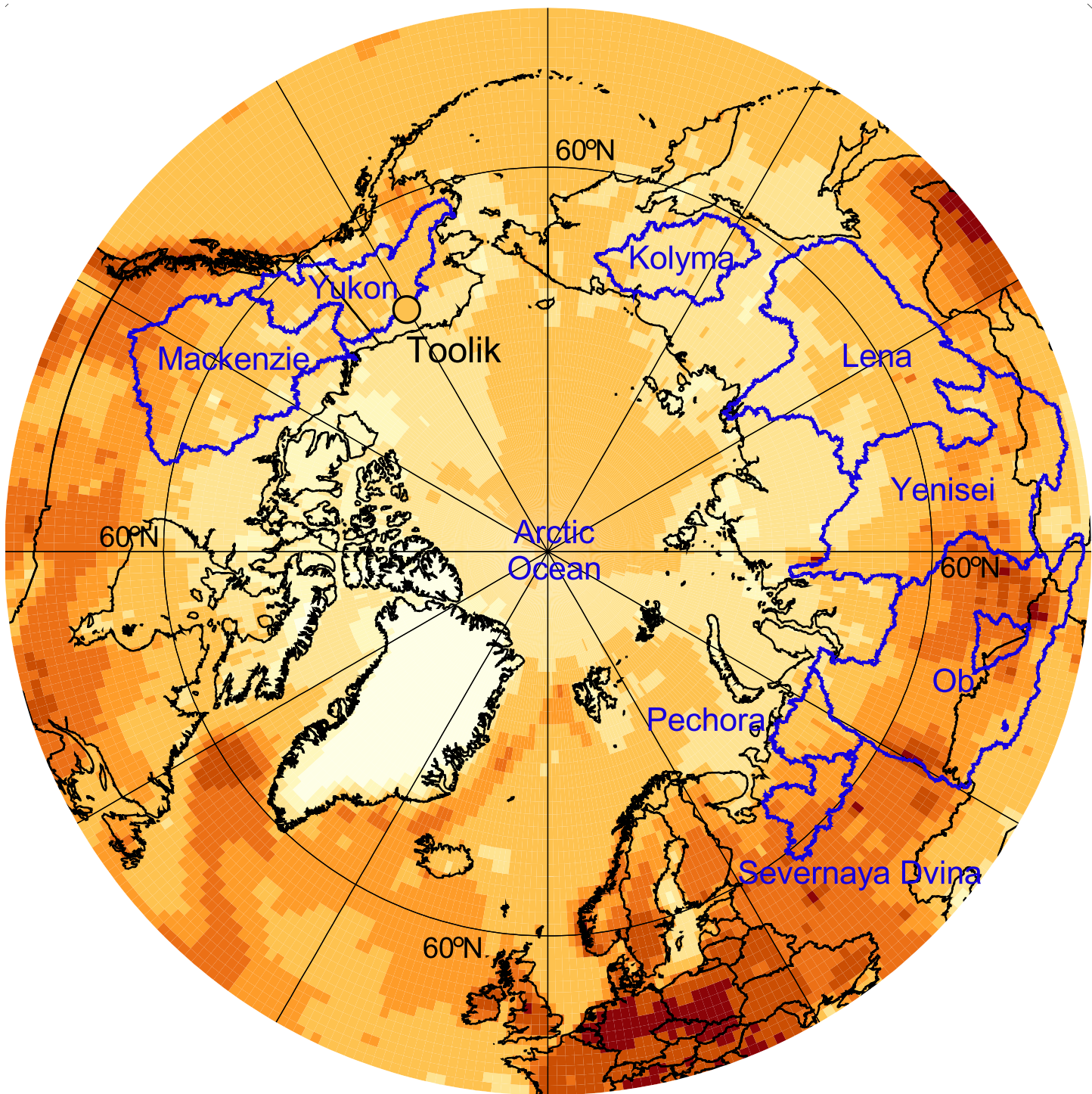
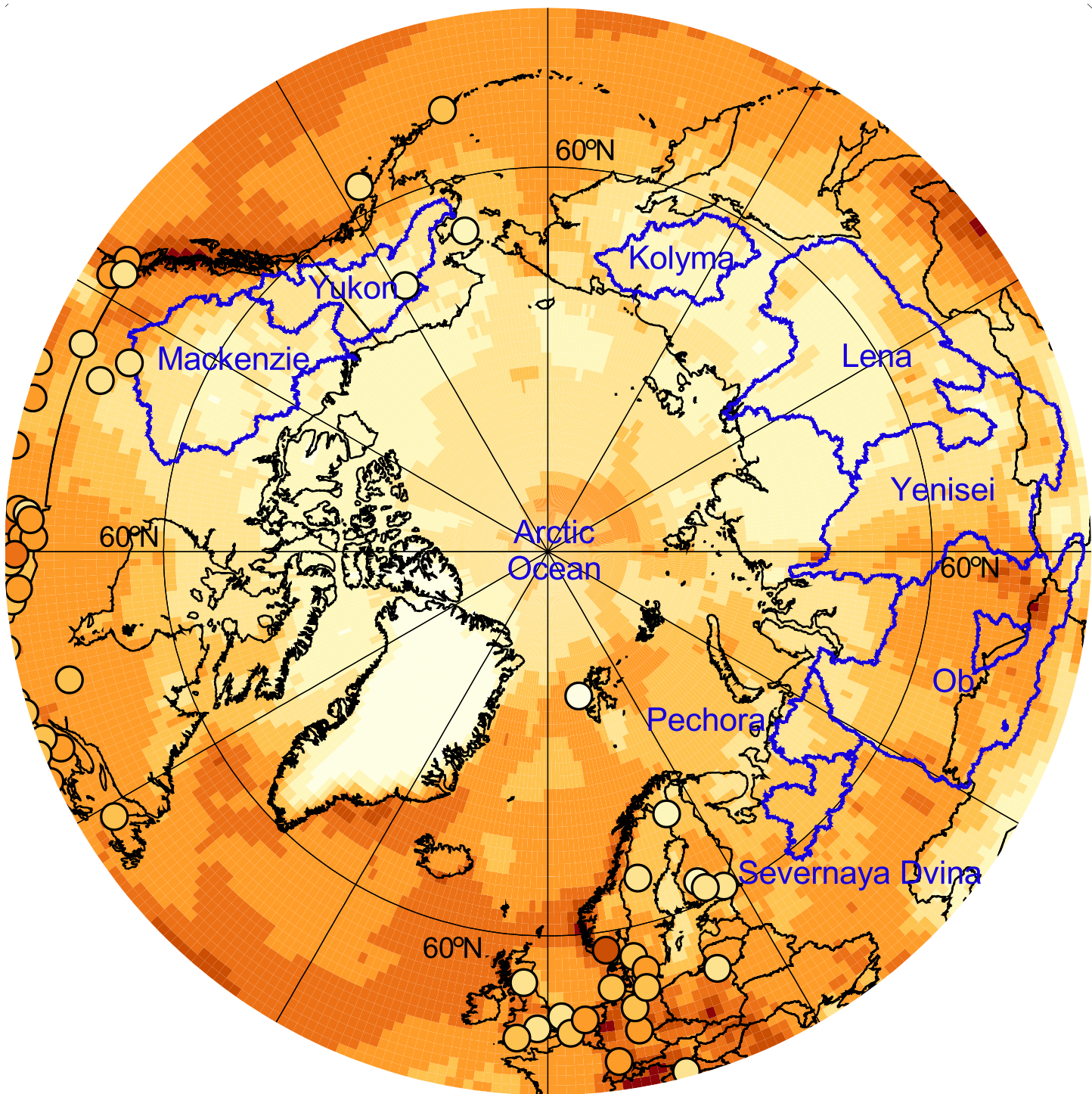


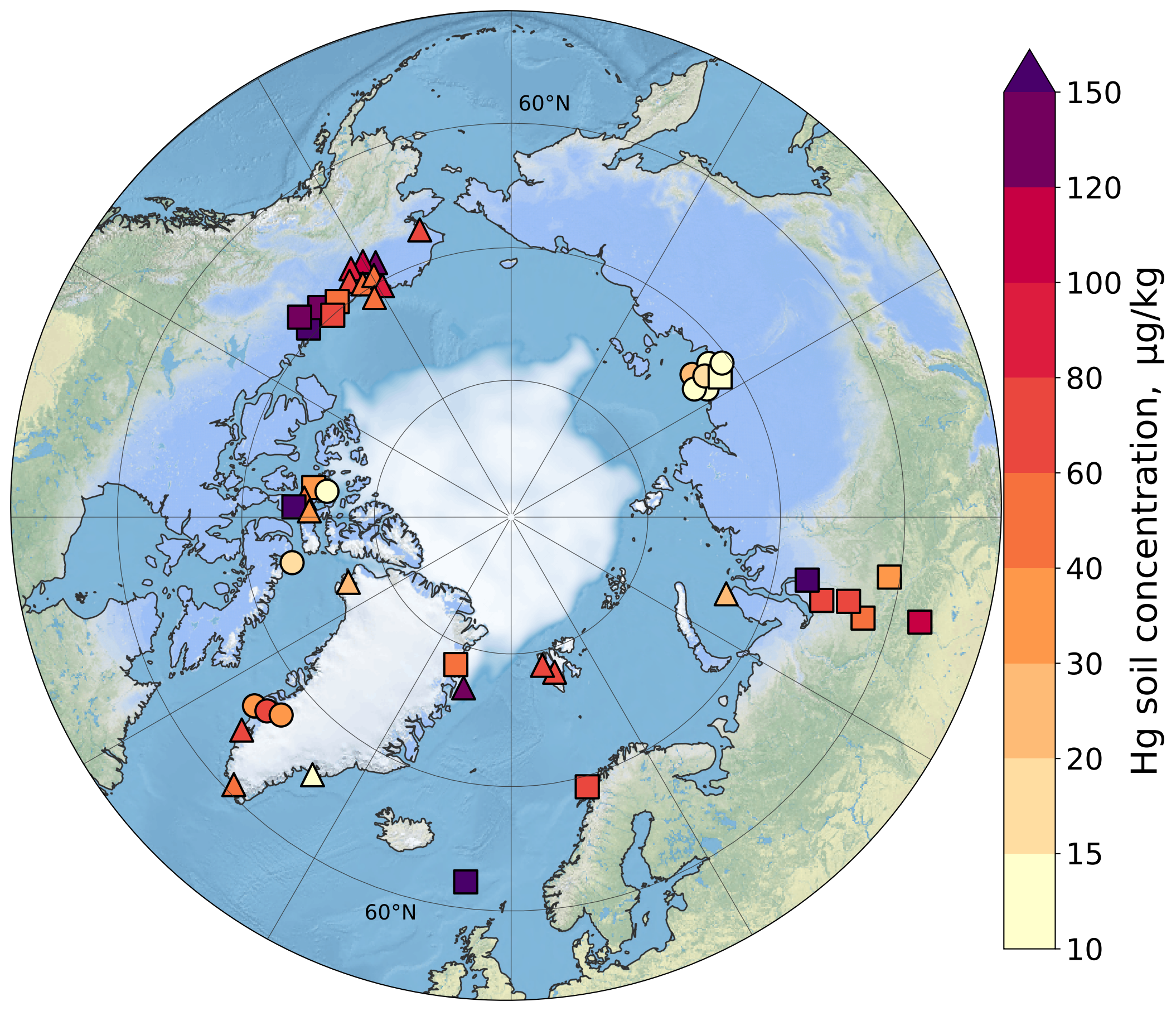
Figure 4 | Arctic Hg inter-compartmental annual fluxes and reservoir budgets. Best estimates (black numbers) and uncertainties in terrestrial and oceanic Hg annual fluxes (identified by arrows with letters) are: anthropogenic (a, 14 Mg y⁻¹), natural and legacy soils/vegetation volatilization (b, 24^{+35.1}_{-17.5} Mg y⁻¹), wildfires (c, 8.8±6.4 Mg y⁻¹), and ocean (d, 32⁺¹³₋₉ Mg y⁻¹) emissions; terrestrial (e, 118±20 Mg y⁻¹), and ocean (f, 64.5±19.8 Mg) deposition; coastal erosion (g, 39⁺¹³₋₂₁ Mg y⁻¹), river (h, 41±4 Mg y⁻¹), ocean (i, 55±7 Mg y⁻¹), and benthic shelf (j, 5 Mg y⁻¹) imports; sea ice (k, 1.4±0.4 Mg y⁻¹), ocean (l, 73±8 Mg y⁻¹), shelf burial (m, 42±31 Mg y⁻¹) and deep ocean burial (n, 3.9±0.7 Mg y⁻¹) exports; and in-ocean shelf (o, 122±55 Mg y⁻¹) and deep ocean (p, 7.2±17 Mg y⁻¹) downward fluxes from surface waters. Best estimates (black numbers) and uncertainties for Hg budgets in reservoirs are: atmosphere (330⁺³⁰₋₄₀ Mg), glaciers (2,415±22 Mg), snowpacks (39⁺³₋₄ Mg), soils (surface, 0-0.3 m: 49±23 Gg; active layer, 0-1 m: 212±28 Gg; permafrost, 0-3 m: 597 Gg), and Arctic Ocean (44±22 Mg, 0-20 m; 228±112 Mg, 20-200 m; 224±106 Mg, 200-500 m; 1,375±616 Mg, 500 m and deeper). Red arrows indicate unknown Hg fluxes of burial in estuaries and inner shelf (q), transport from shelf to deep Arctic Ocean basin (r), and benthic deep basin (s) fluxes. Arctic Ocean sediments Hg concentrations are 28.9±22.0 ng g⁻¹ in shelf and 60.4±44.5 ng g⁻¹ in deep basin. Arctic land is defined as region north of 60° N and the Arctic Ocean is defined as the central basin, and Barents, Kara, Laptev East Siberian, Chukchi and Beaufort Seas (94.5 x 10⁵ km², total ocean surface area; 53% shelf). The Hg fluxes and budgets were estimated in this Review based on multi-model ensemble simulations and peer-reviewed literature.

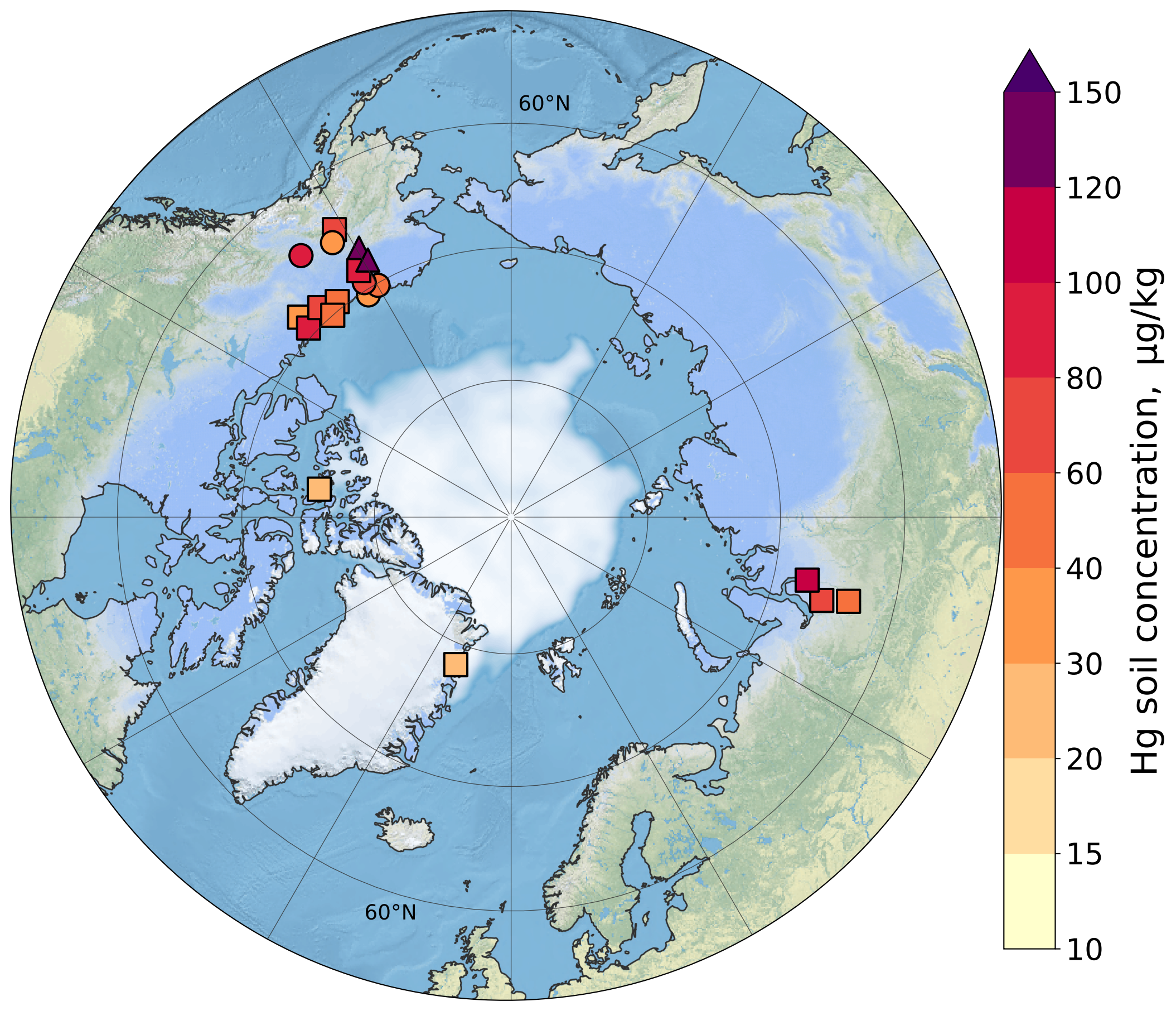


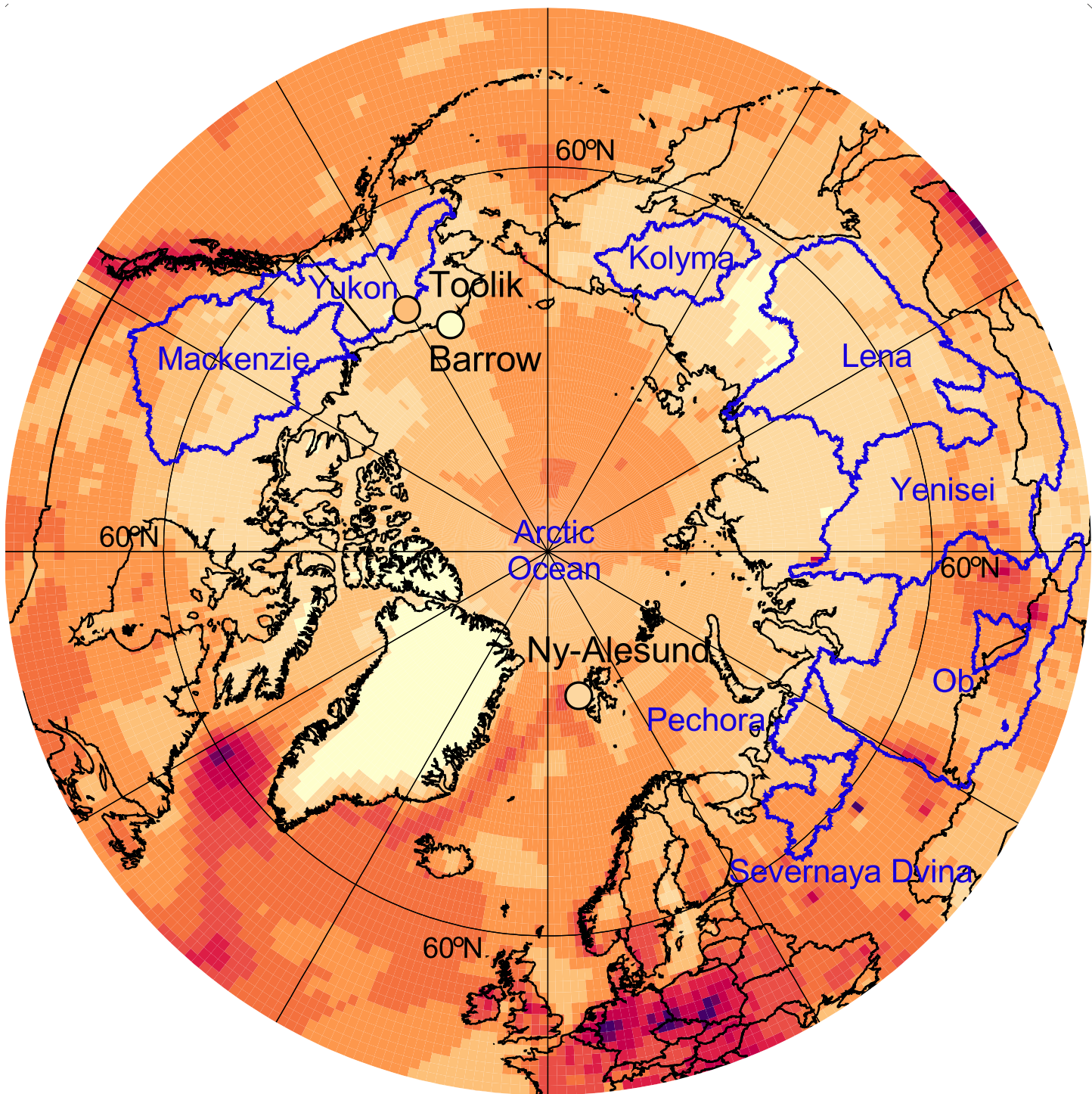


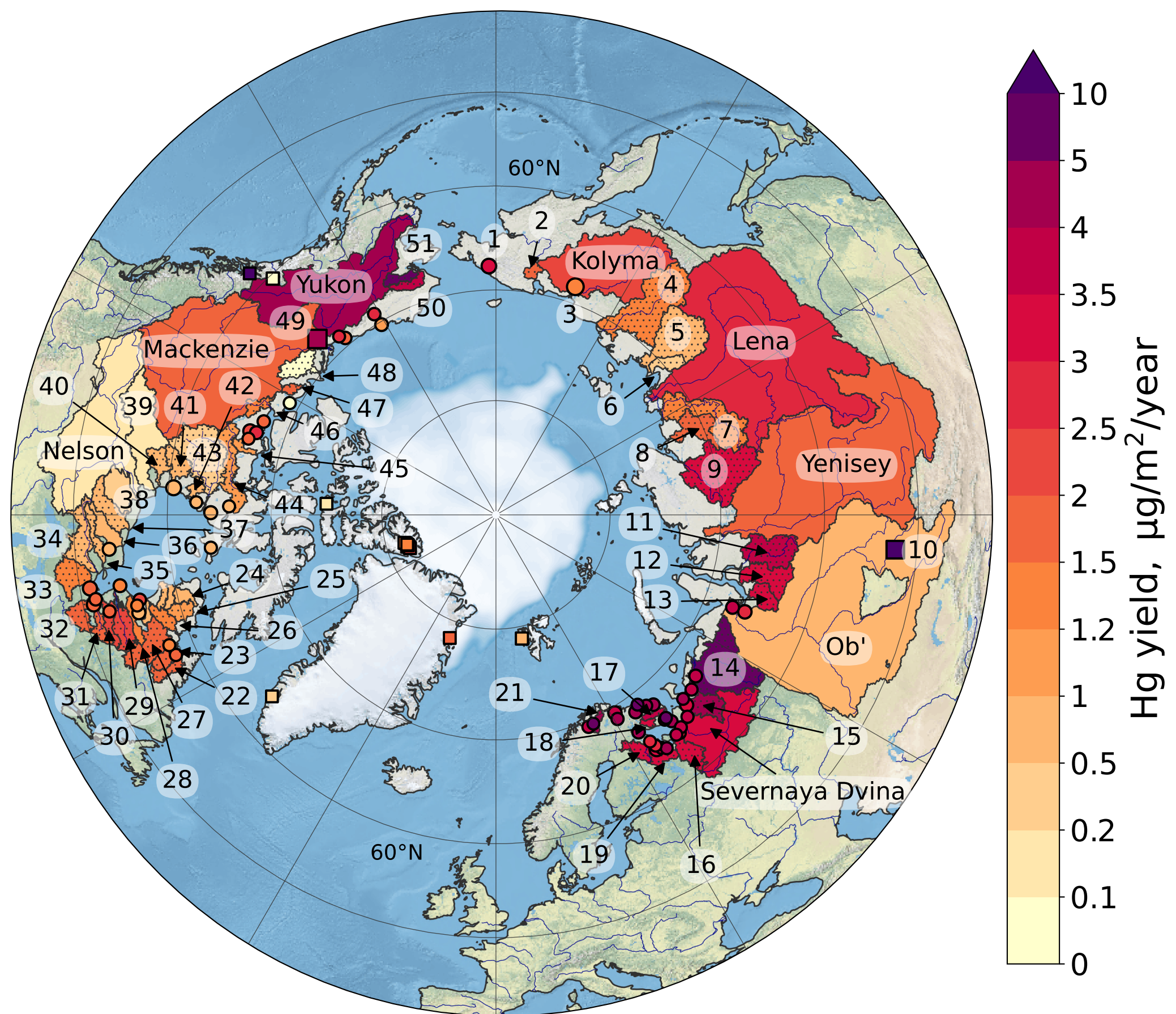




















total Hg [ng L⁻¹]

