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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} ; 18-52 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.

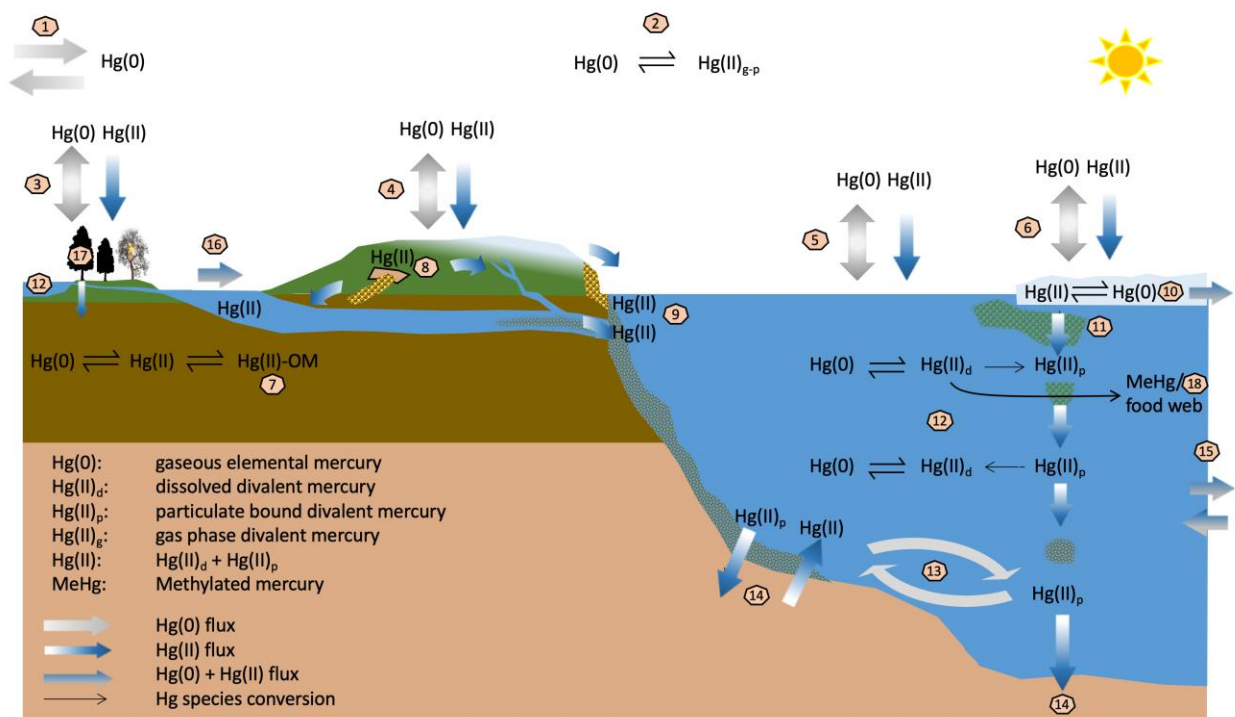
714 **Figure 3. | Spatial distribution of Hg in the Arctic Ocean.** a| Synoptic pan-Arctic transect of total Hg
 715 concentrations during the 2015 GEOTRACES cruises (ng L^{-1})^{22,191,194}. The insert map shows the transect
 716 (red band) and available Hg observations (blue dots). b| Surface water 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 **Competing interests**

792 The authors declare no competing interests.

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Figures

Arctic mercury cycling

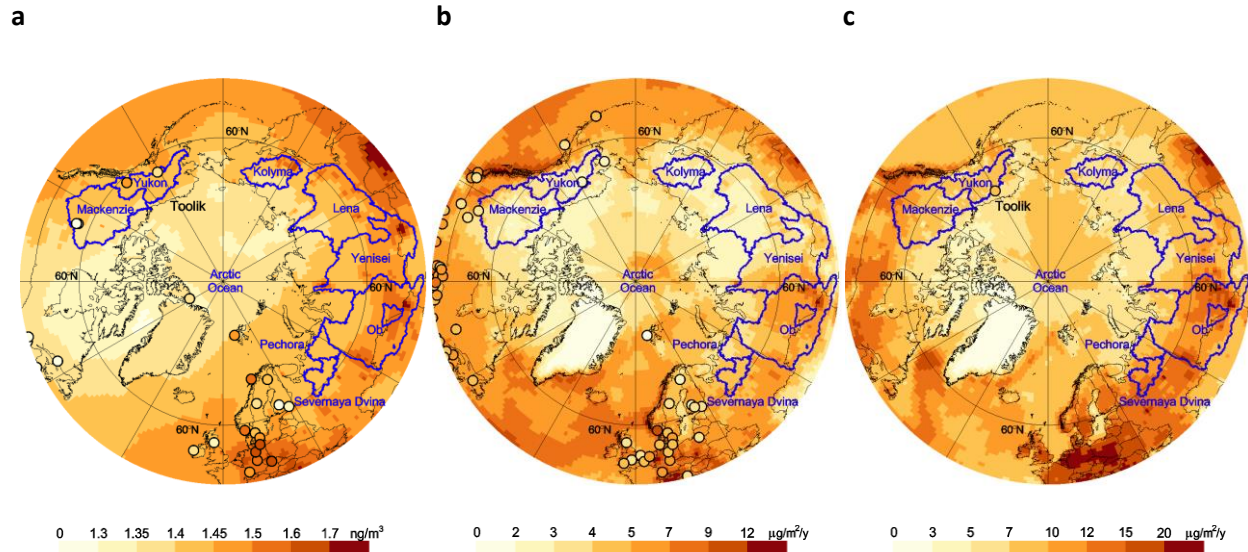


Figure 1. | Atmospheric Hg distribution in the Arctic. **a**) Model ensemble (DEHM, GEM-MACH-Hg, GLEMOS, GOES-Chem) simulated (this Review) annual average surface air gaseous elemental mercury (Hg(0)) concentrations in 2015. **b**) Annual mercury (Hg) wet deposition flux in 2015. **c**) Annual Hg total 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.

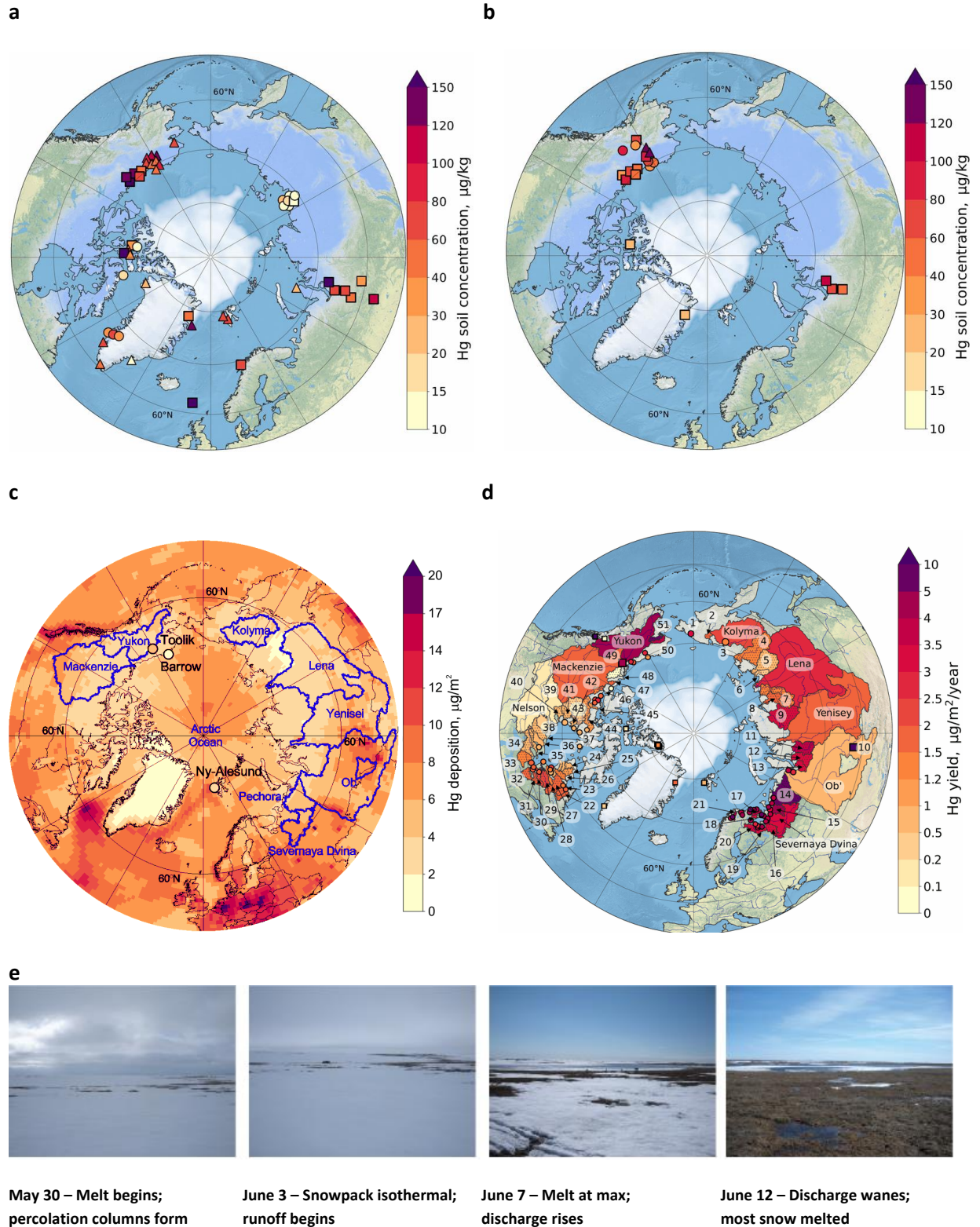


Figure 2. | Distribution of total Hg in Arctic soils, wintertime deposition and rivers. a| Mean mercury (Hg) concentrations of active layer soils within 0-1 m depth. b| Mean Hg concentration of permafrost

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.

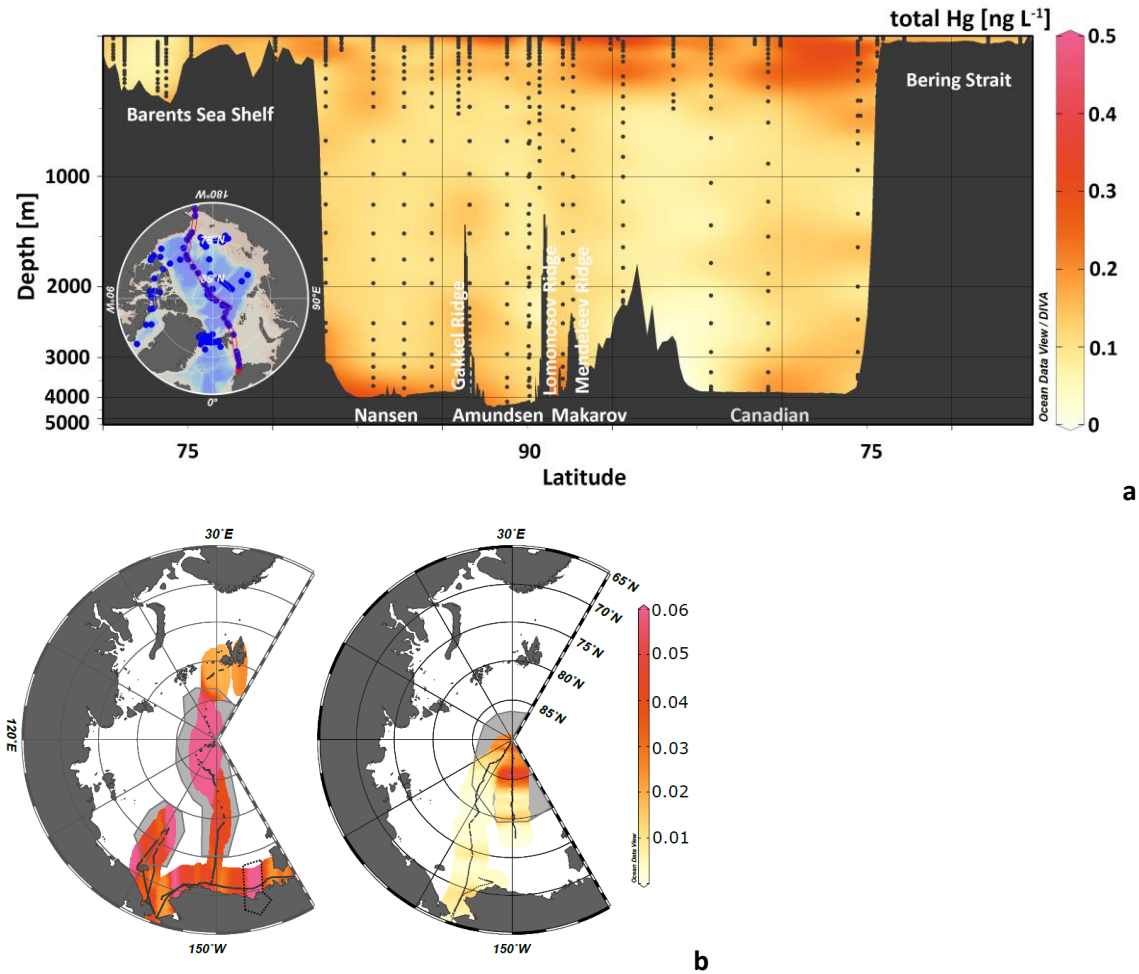


Figure 3. | Spatial distribution of Hg in the Arctic Ocean. **a**) Synoptic pan-Arctic transect of total Hg concentrations during the 2015 GEOTRACES cruises (ng L^{-1})^{22,191,194}. The insert map shows the transect (red band) and available Hg observations (blue dots). **b**) Surface water Hg(0) concentrations (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}.

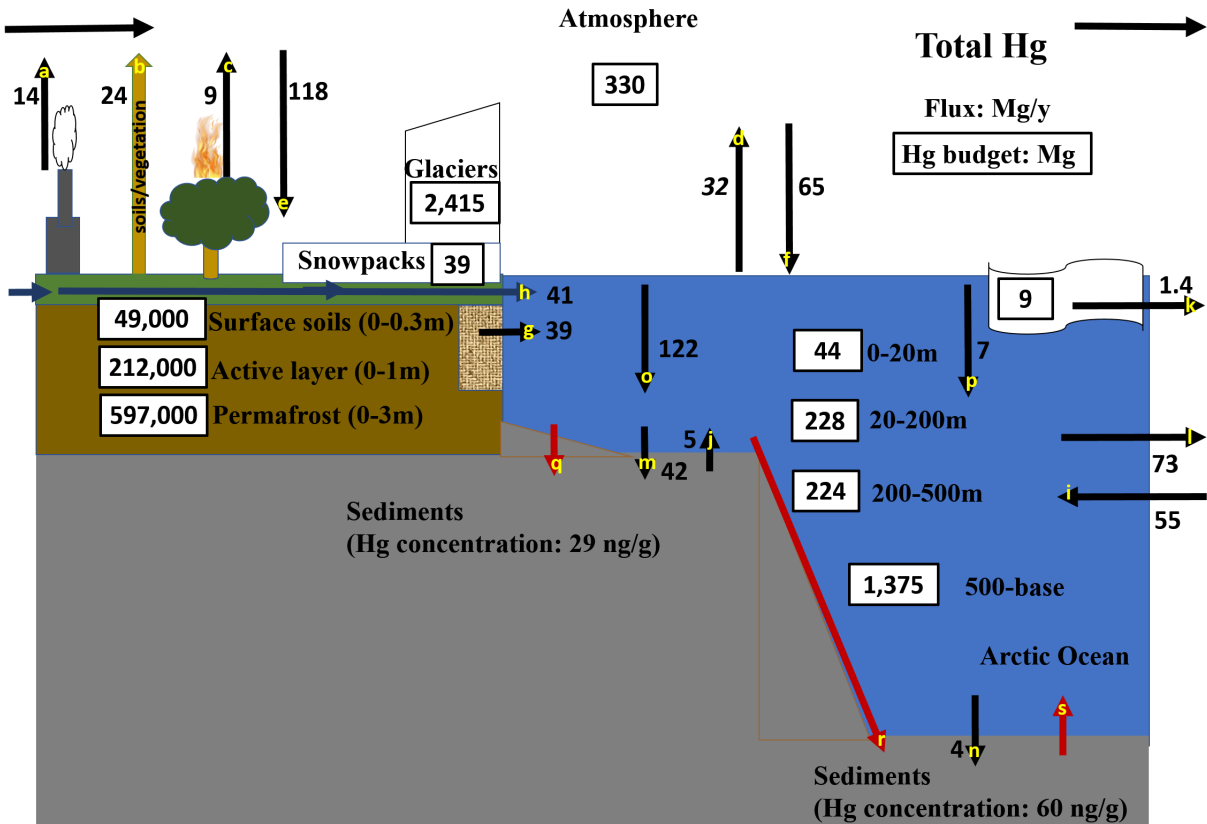
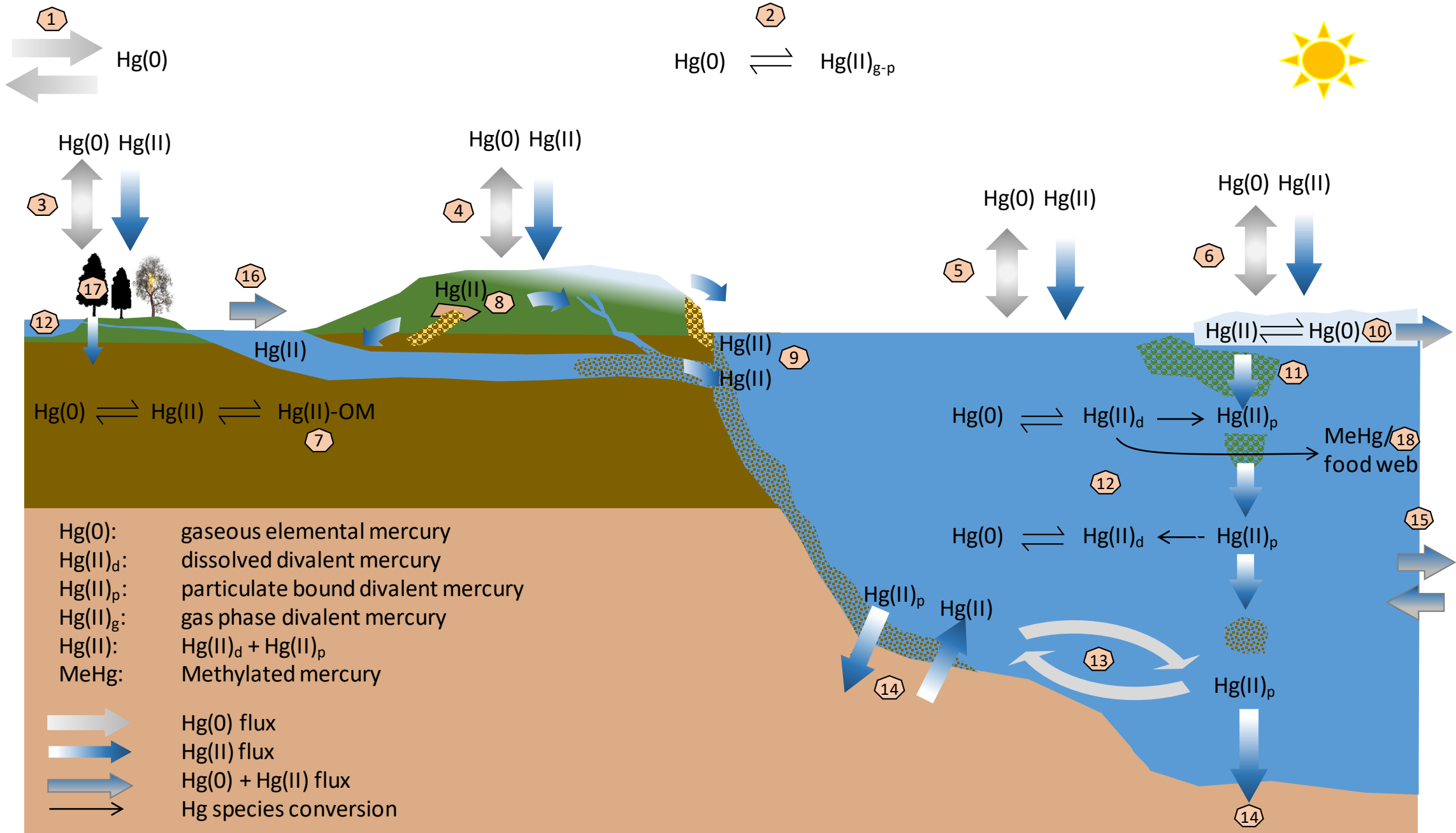
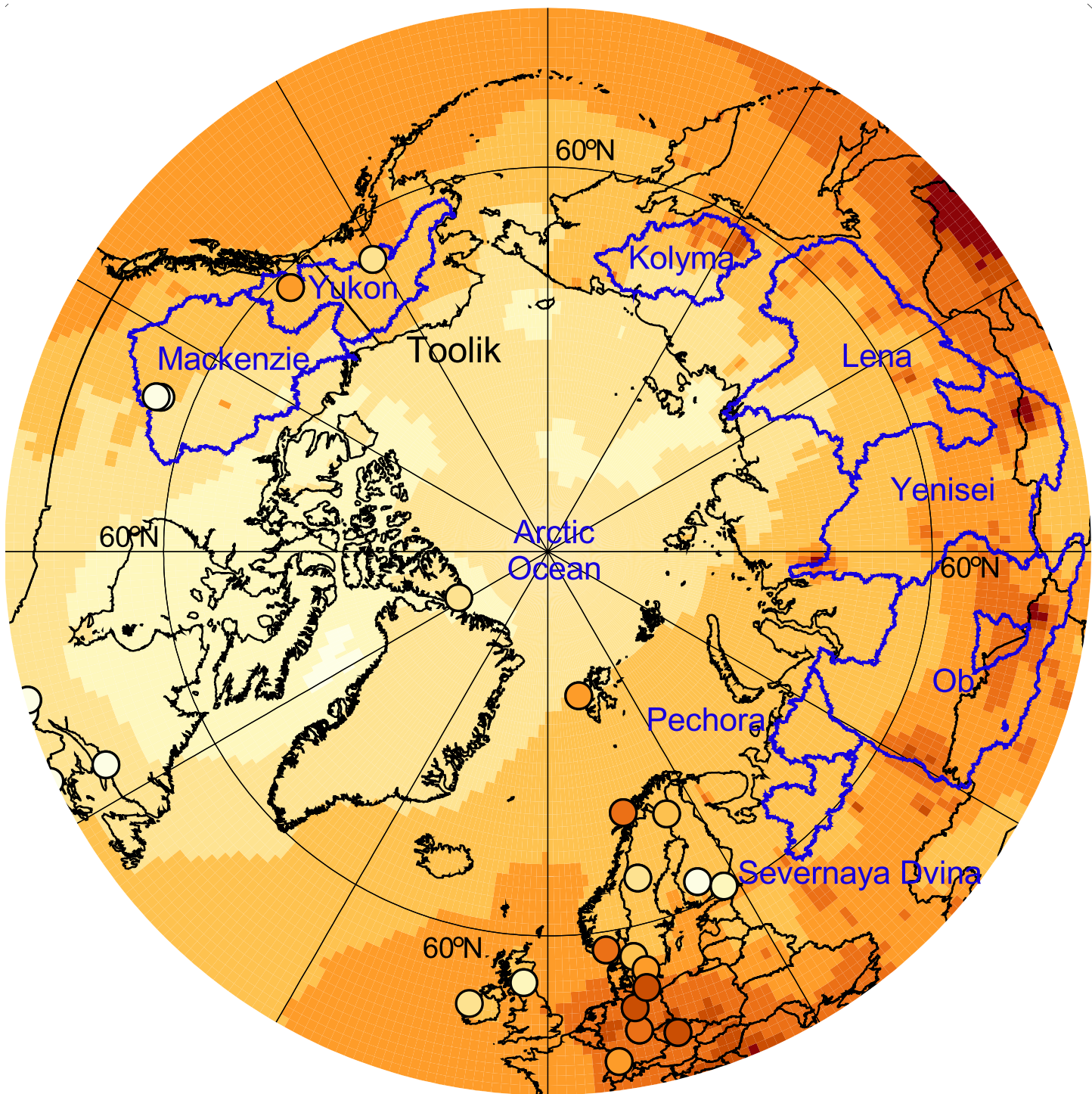
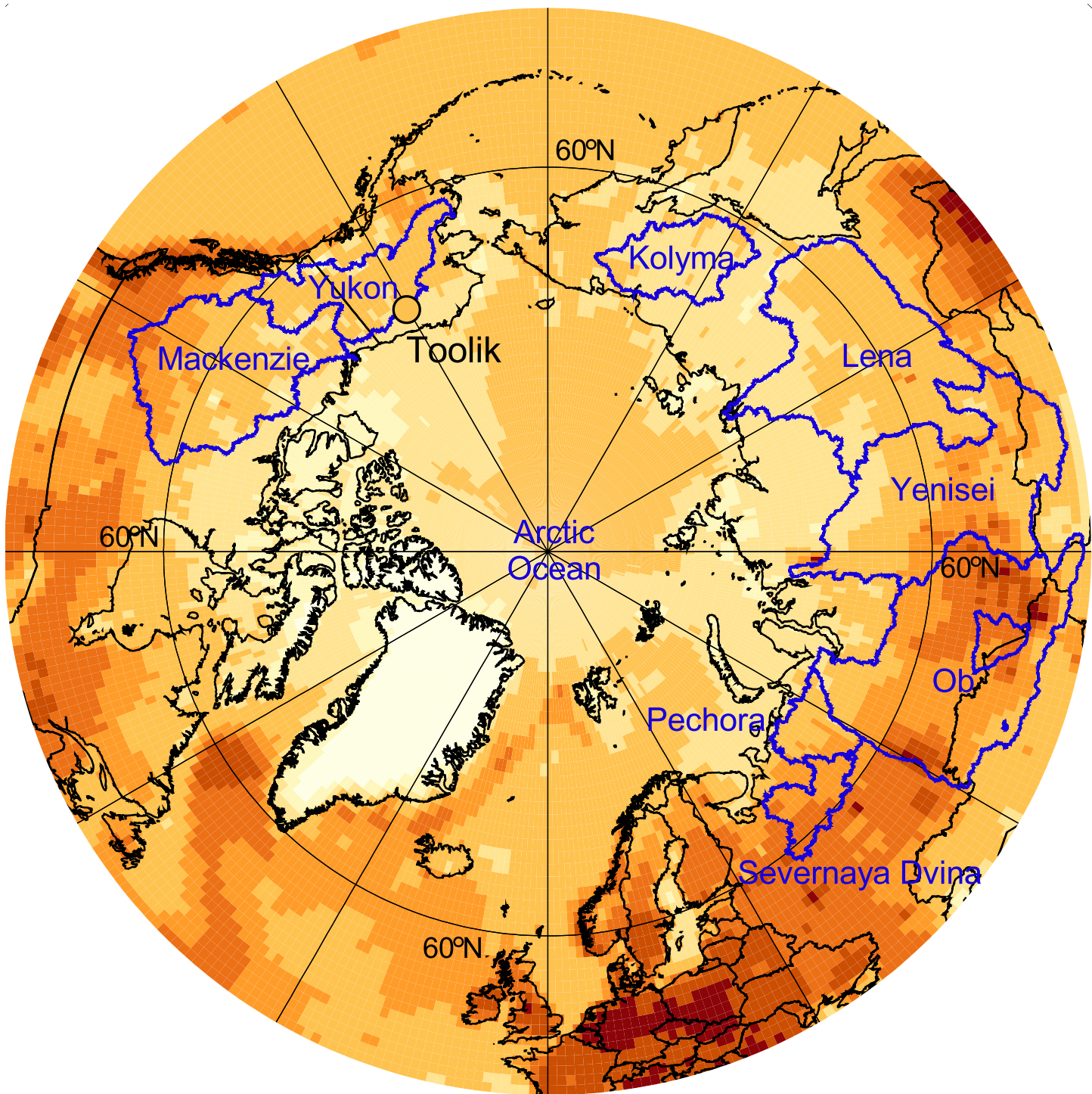
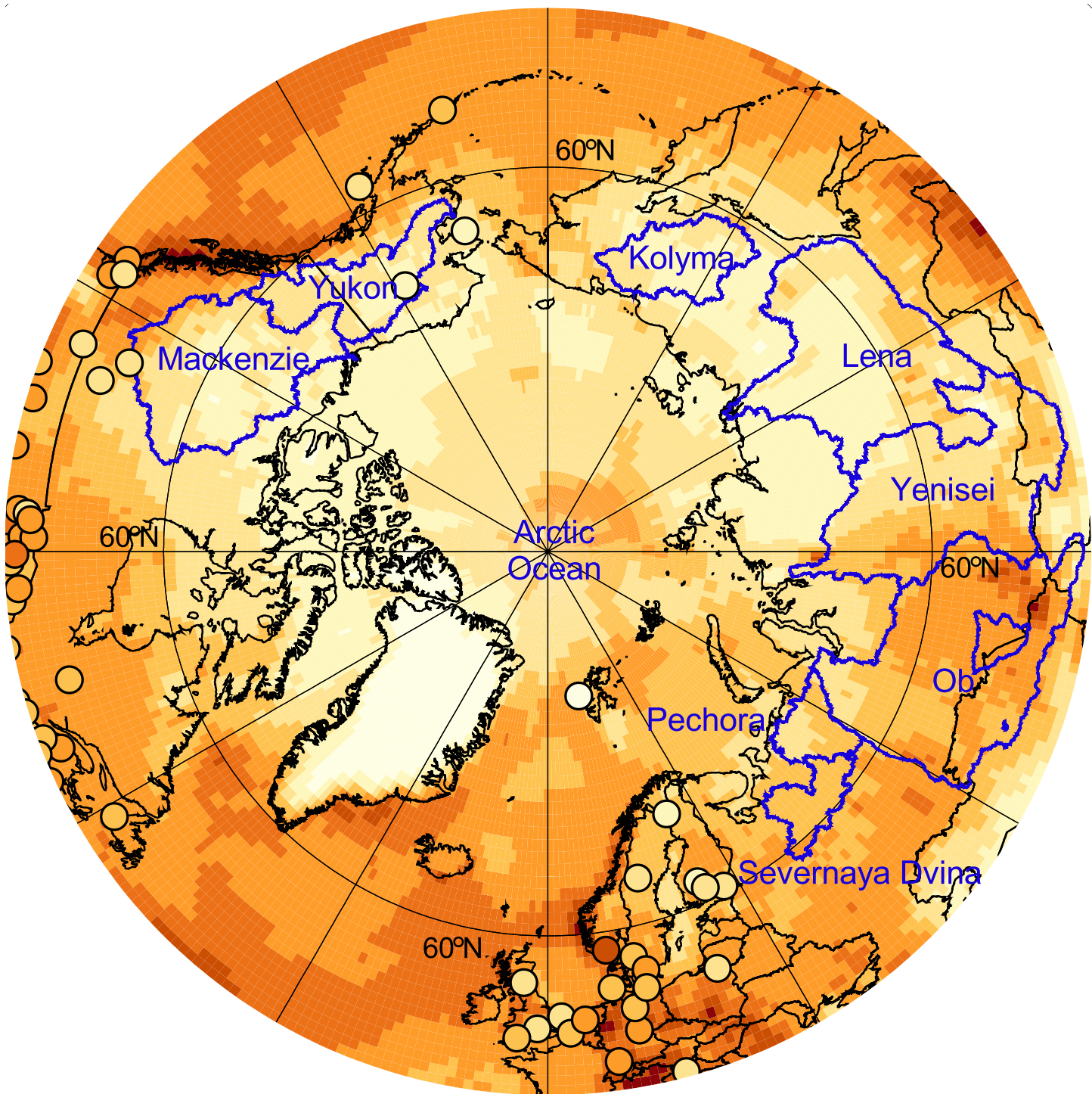


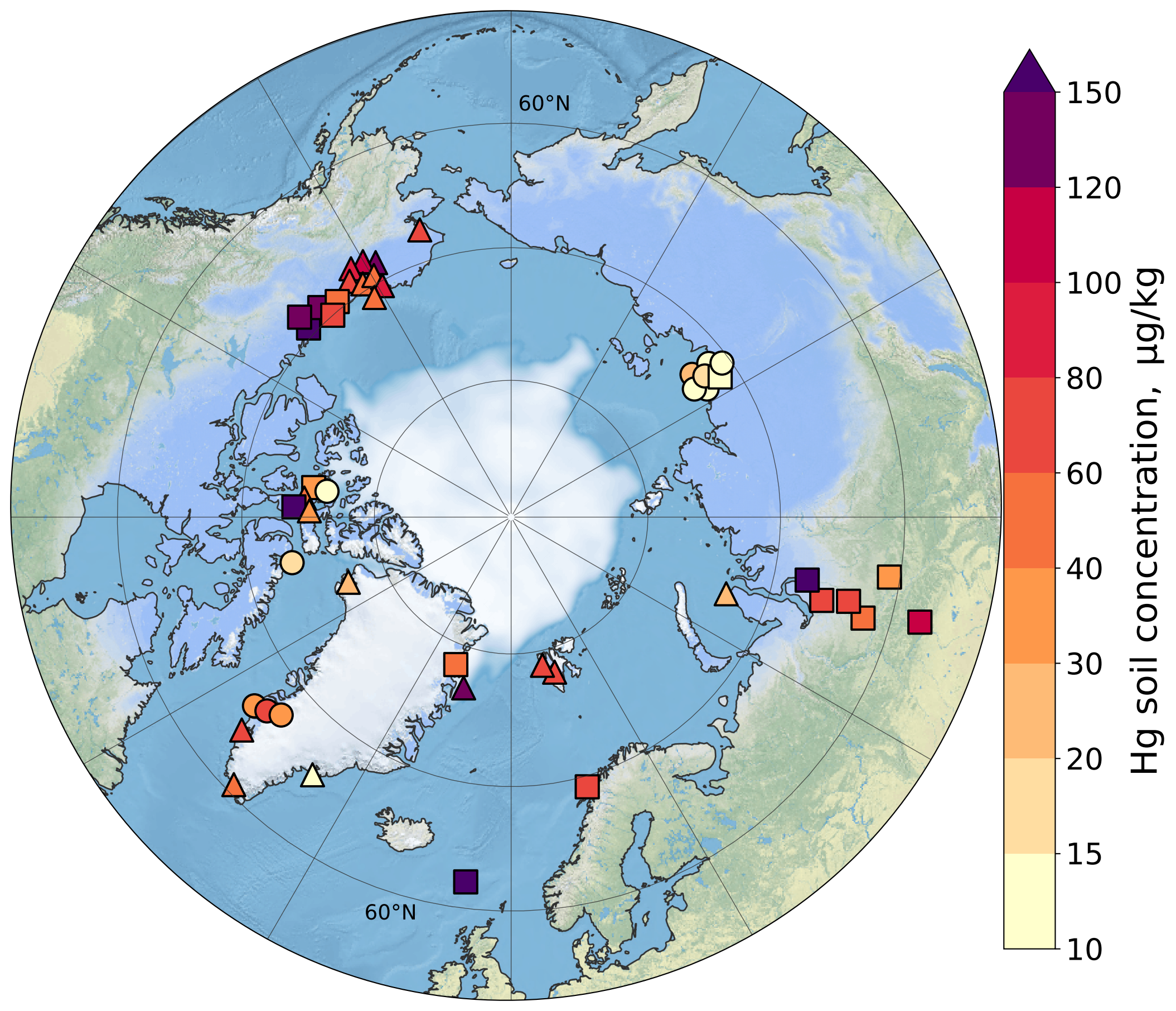
Figure 4 | Arctic Hg inter-compartmental annual fluxes and reservoir budgets. Best estimates (black numbers) and uncertainties in terrestrial and oceanic Hg annual fluxes (identified by arrows with letters) are: anthropogenic (a, 14 Mg y⁻¹), natural and legacy soils/vegetation volatilization (b, 24^{+35.1}_{-17.5} 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.

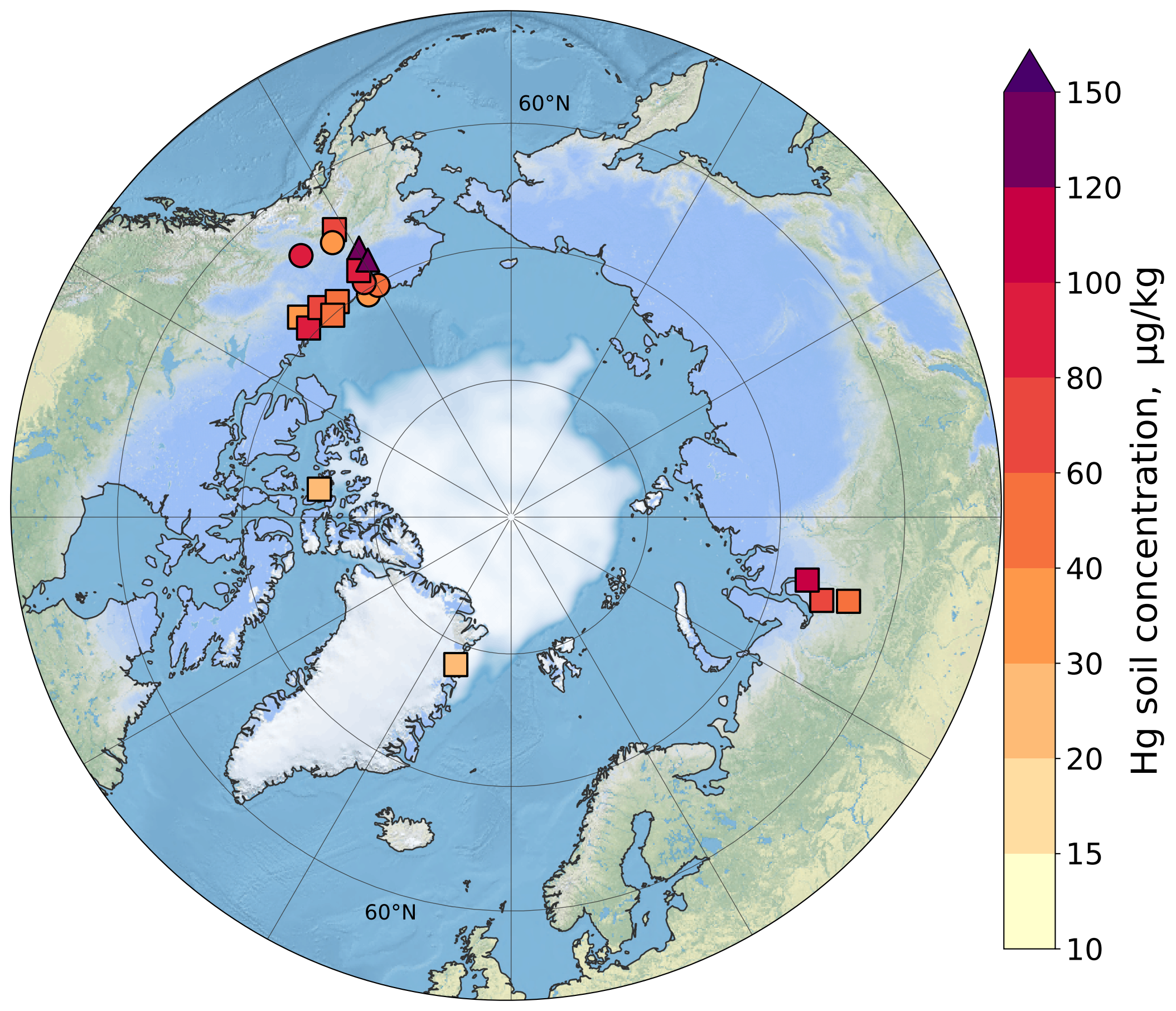


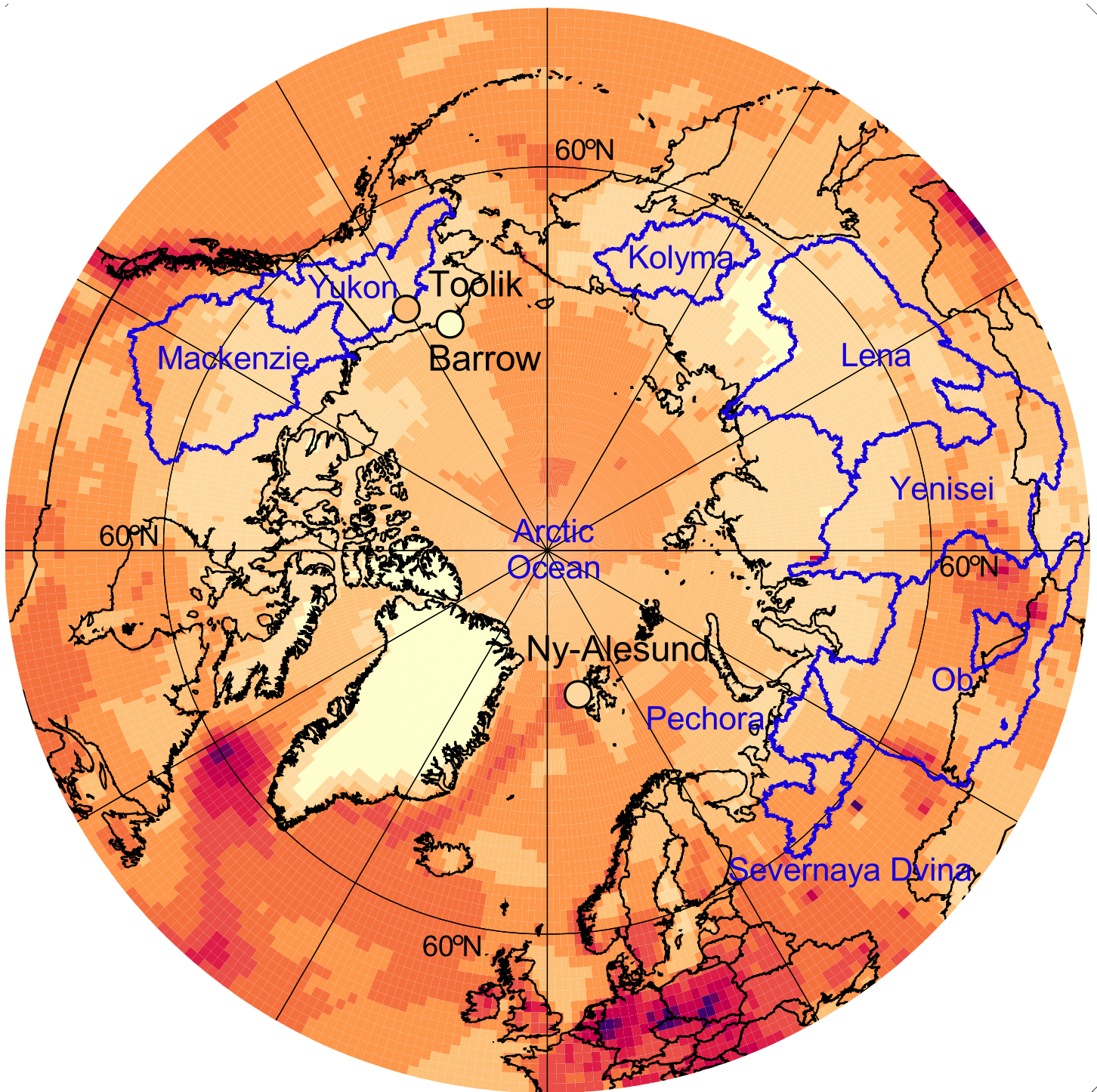


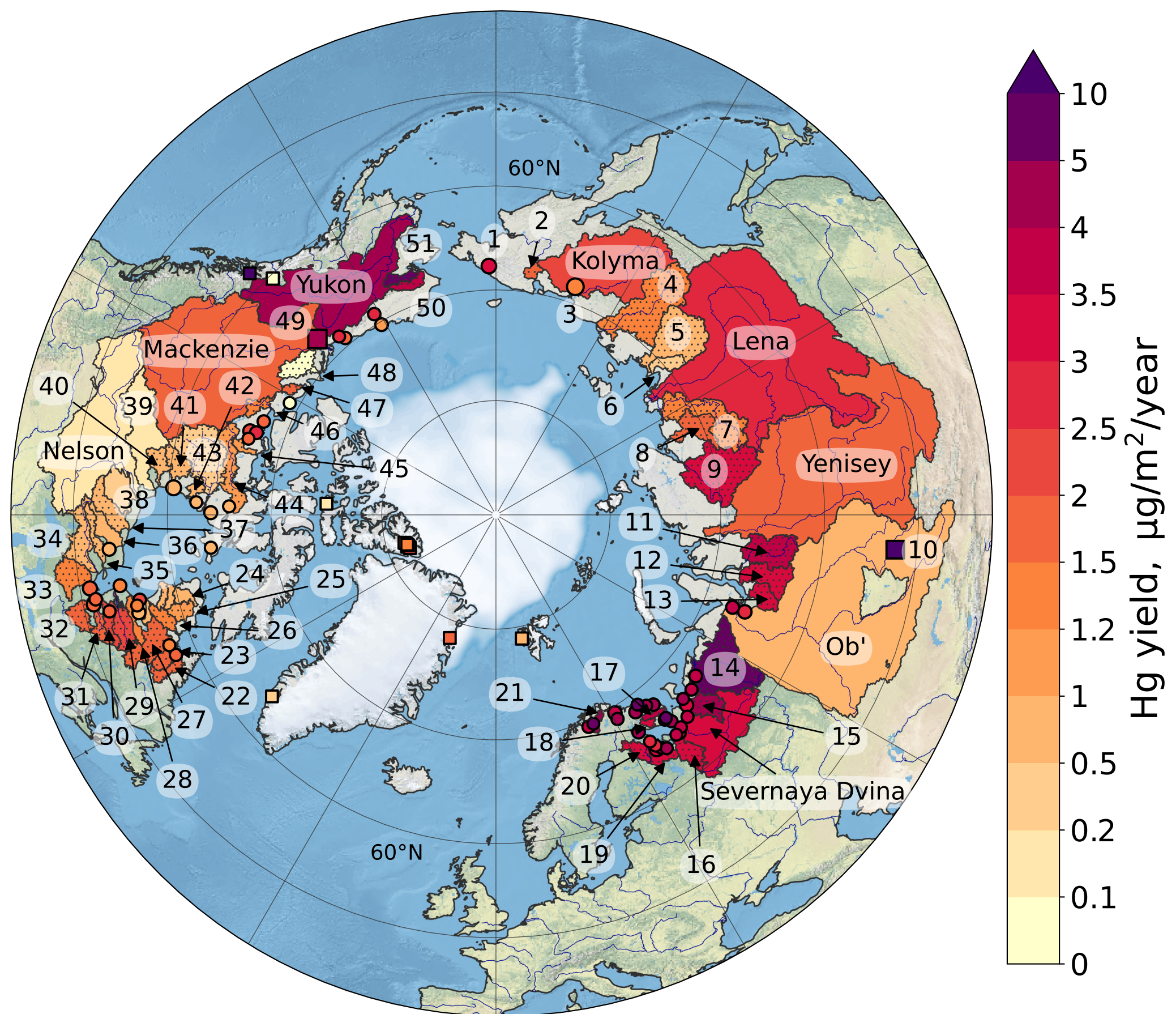




















total Hg [ng L⁻¹]

