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6

7 Unequal anthropogenic enrichment of mercury in Earth's northern and 8 southern hemispheres

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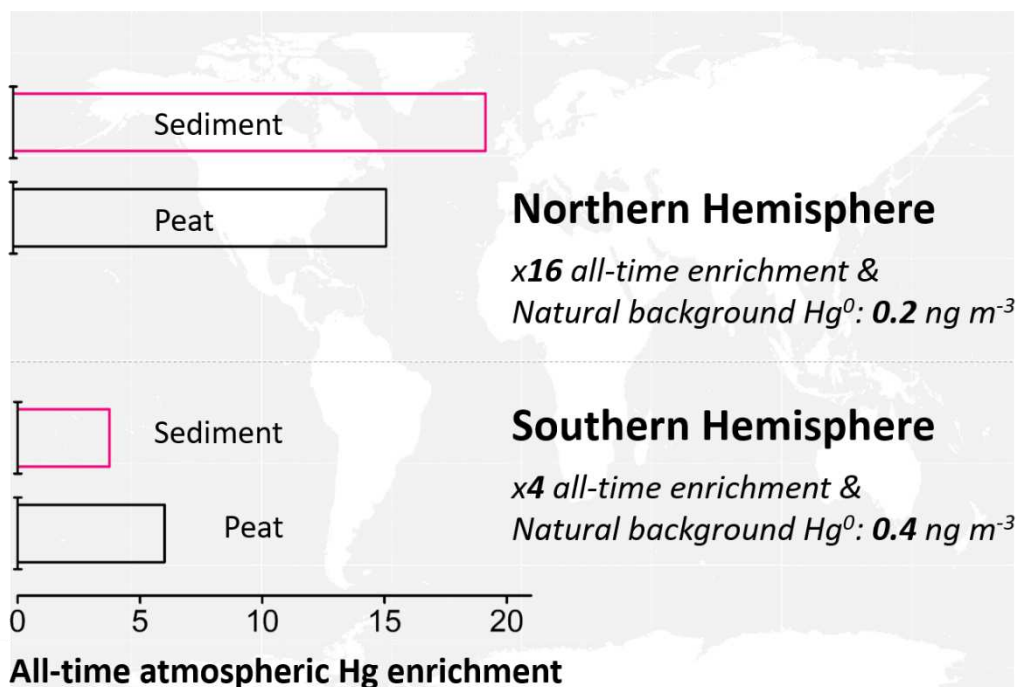
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30
 31 **Abstract:** Remote northern (NH) and southern hemisphere (SH) lake sediment and peat records of mercury
 32 (Hg) deposition show a $\times 3$ to $\times 5$ Hg enrichment since pre-industrial times (<1880AD), leading to the
 33 perception that global atmospheric Hg enrichment is moderate and uniform across the hemispheres.
 34 Anthropogenic Hg emissions in the NH are, however, approximately four times higher than in the SH. Here
 35 we reconstruct atmospheric Hg deposition to four remote SH peatlands and review sediment and peat Hg
 36 records from both hemispheres. We observe a $\times 4$ all-time enrichment in SH Hg deposition from pre-
 37 anthropogenic (<1450AD) to late 20th century periods, which is lower than the large $\times 16$ all-time
 38 enrichment in NH Hg deposition. We attribute this difference to lower anthropogenic Hg emissions in the
 39 SH, and higher natural atmospheric SH Hg concentrations, supported by $\times 2$ higher natural background Hg
 40 accumulation in SH peat records. We suggest that the higher SH natural atmospheric Hg concentration
 41 reflects the SH land-ocean distribution, and is driven by important SH marine Hg emissions. Our findings
 42 suggest that atmospheric Hg background levels and anthropogenic enrichment in both hemispheres are
 43 different and should be taken into account in international Hg assessments and environmental policy.

45 **Introduction:**

46 Mercury (Hg) is a toxic trace metal that affects wildlife and human health ¹⁻⁴. Hg is discharged into the
47 environment by natural processes, such as volcanism, chemical and physical weathering, and by human
48 activities, including mining, coal burning and intentional use ⁵⁻⁷. Elemental Hg⁰, the dominant form of
49 emissions, has a long atmospheric residence time of 6 to 12 months, which allows for its intra-hemispheric
50 dispersion before being deposited to the Earth's surface, including remote environments ⁸. Assessments
51 of the extent of global Hg pollution have relied upon natural archives of Hg accumulation (e.g. sediment
52 ^{9,10}, peat ¹¹, ice cores ¹²), and on estimates of natural and anthropogenic Hg emissions ⁷.

53 Since early work on lake sediment cores in the 1970s ¹³, hundreds of remote ²¹⁰Pb dated sediment
54 cores have documented an approximate three- to five-fold increase in Hg accumulation rates (HgAR) from
55 pre-industrial (1760-1880 AD) times to the late 20th century ¹⁴⁻¹⁹. A comprehensive review in 2007
56 concluded that sediment records were more reliable than peat records in recording atmospheric HgAR ¹⁷.
57 Inferred, higher Hg accumulation in peat records was thought to be related to ²¹⁰Pb mobility, and peat
58 mass loss during remineralization. A recent review study ¹⁸ indicated that earlier peat vs sediment
59 comparisons ¹⁷ used different reference periods to calculate Hg enrichment. Using coherent reference
60 periods, dozens of peat archives and a small number glacier ice cores of atmospheric deposition also
61 document 3 to 5-fold enrichment factors, similar to sediment records, since pre-industrial times (EF_{preind})
62 ^{14,18}. Both sediment and peat records have strengths and weaknesses, with ²¹⁰Pb and Hg mobility during
63 sediment diagenesis and peat decomposition being potential factors of bias ²⁰⁻²². Yet, both archives at
64 remote locations record broadly similar Hg accumulation profiles across the past millennium, despite
65 differences in archive functioning, and therefore warrant further comparison across Earth's two
66 hemispheres. Regarding archive functioning, lake sediments integrate Hg deposition to a larger watershed,
67 Hg storage in soils, followed by Hg run-off and in-lake cycling leading to a longer Hg residence-time before
68 deposition to sediments. Peatlands integrate Hg deposition directly from the atmosphere ^{18,23,24}, leading
69 to a more direct response of peat archives to atmospheric Hg⁰ concentrations. This can generally be
70 recognized by the 2-fold drop in HgAR from the 1970s to the 1990s in peat ¹⁸, which is absent in sediment
71 records, and which mirrors the well-documented decrease in Hg emissions and observed atmospheric Hg⁰
72 concentrations ^{7,25,26}. A comparison of Hg stable isotope composition of peat and lake sediments indicates
73 that in both media, 75% of Hg derives from uptake of atmospheric Hg⁰ ²³, which further justifies comparing
74 both archives.

75 Longer radiocarbon-dated NH sediment and peat cores probe changes in the natural background Hg
76 accumulation during pre-colonial times (pre-1450AD), before large-scale mining practices, and indicate a

77 more dramatic difference in Hg deposition. Millennial sediment and peat records show that HgAR already
78 increased five-fold during the earlier transition from pre-large-scale mining to pre-colonial times around
79 approximately 1450 AD ¹⁸. All-time anthropogenic Hg enrichment factors ($EF_{alltime}$, the ratio of 20th century
80 to pre-1450AD HgAR), determined in sediment and peat records therefore ranges from 16 to 26 ¹⁸. The
81 cause for the increase in NH Hg enrichment around 1450AD is debated. Some Hg inventory and modeling
82 studies have argued for enhanced Hg emissions from Spanish colonial silver and gold mining ²⁷⁻²⁹. Other
83 studies argue that Hg associated with mining has been immobilized in mining waste, rather than volatilized
84 ^{1,9,30}. A study on Hg stable isotopes in peat has recently shown evidence how enhanced deforestation
85 during the Middle Ages may have impacted regional atmospheric Hg dynamics in Europe with lower
86 vegetation uptake of Hg, and wood burning emissions leading to enhanced atmospheric Hg concentrations
87 and deposition ²⁵. What nearly all the above cited studies have in common, is that they are situated in the
88 northern hemisphere (NH) where the majority of historical anthropogenic Hg emissions have taken place
89 and have been abundantly investigated. Relative to the NH, anthropogenic Hg emissions in the SH have
90 continuously been four times lower ³¹. Reviews of anthropogenic Hg enrichment in the environment
91 generally provide a global picture without discerning the hemispheres ^{20,32-34}. Lake sediment records of Hg
92 accumulation have been studied in the SH and will be reviewed here. Three southern hemisphere (SH)
93 peat records have been studied for HgAR ^{35,36}, but are all incomplete (see Methods, and Extended Data 2)
94 and preclude a rigorous assessment of SH atmospheric Hg enrichment based on both sediment and peat
95 archives.

96 The aim of this study was therefore to investigate differences in anthropogenic Hg enrichment, if any,
97 in Earth's SH and NH. We hypothesize that, in regard of the lower historical SH anthropogenic Hg emissions,
98 enrichment will also be lower. We extend the limited number of peat archives studied in the SH, by
99 investigating Hg accumulation rates in four new radiocarbon and ²¹⁰Pb and ¹⁴C bomb-pulse dated SH peat
100 records. We then review all the existing SH sediment and peat HgAR (Extended Data 2), compare Hg
101 enrichments factors to the NH, and discuss findings in the context of revised volcanic Hg emissions,
102 published historical anthropogenic Hg emissions, and Hg cycling in both hemispheres. We do not include
103 glacier ice cores in our review due to the limited number of studies available, and we do not consider
104 marine sediment records. Four reference time periods, operationally defined for NH natural archives
105 elsewhere ^{18,19}, will be used throughout: natural background (pre-1450AD), pre-industrial period (1450-
106 1880 AD), 20th century extended HgAR maximum (20Cmax, approximately from 1940-1990; see also
107 Methods), and the recent post-1990 modern period.

108 **Materials & Methods**

109 **The study sites.** We investigate four new cores from remote ombrotrophic peat bogs in the SH mid-
110 latitudes: Amsterdam Island (AMS, S-Indian Ocean), Falkland Islands (SCB, San Carlos bog, Islas Malvinas,
111 S-Atlantic Ocean), Andorra and Harberton (AND, HBT, Tierra del Fuego, Argentina) (SI Appendix Table S1;
112 Figure S1; Text S1; Extended Data 1). These four sites are situated in the Southern Westerly wind belt, far
113 away from anthropogenic Hg sources, which makes them ideal recorders of SH remote atmospheric Hg
114 deposition trends. Details about the field campaigns and sampling sites are given in SI Appendix Table S1
115 and Text S1. After collection, all the cores were photographed, described and packed in plastic film and
116 PVC tubes and shipped to EcoLab, Toulouse, France. There, the cores were cut and processed following
117 published trace metal clean protocols, freeze-dried and stored dry until analysis^{37,38}.

118 **Chronology.** Age model output of the AMS peat core is adopted from ref³⁹. In brief, a total of 20 samples
119 were picked for plant macrofossils and subsequently radiocarbon-dated at the LMC14 Artemis Laboratory
120 (Saclay, France, SacA code) or GADAM center (Gliwice, Poland, GdA code). Recent age control in the AMS
121 peat core is based upon 4 post-bomb radiocarbon dates⁴⁰ together with ²¹⁰Pb dating using the constant
122 rate of supply model, and ¹³⁷Cs, ²⁴¹Am⁴¹. A total of 9 samples of plant macrofossils/charcoal from SCB 10
123 and 13 samples of *Sphagnum* macrofossils from AND and HBT respectively, were radiocarbon dated. These
124 radiocarbon samples were pre-treated and graphitized at the GADAM center (Gliwice, Poland, GdA code)
125 ⁴². Subsequently, their ¹⁴C concentration in graphite was measured at the DirectAMS Laboratory (Bothell,
126 WA, USA; ⁴³). The NIST Oxalic Acid II standard was used for normalization, and black coal used as a blank.
127 A total of 22 samples from the top 62 cm of the SCB peat core were selected for ²¹⁰Pb measurement by
128 alpha counting to constrain the recent age (see Extended Data 1). The recent age control of the AND and
129 HBT peat cores derive from 5 and 10 post-bomb radiocarbon dates, respectively^{40,44}.

130 Details of radiocarbon dates are summarized in SI Appendix Table S2. Age-depth models were
131 generated from a combination of radiocarbon dating, post-bomb and ²¹⁰Pb dating with the Bacon package
132 within R software⁴⁵, using the SHCal13 calibration curve for positive ¹⁴C ages⁴⁶, while the post-bomb
133 radiocarbon dates were calibrated with SH zone 1-2 curve⁴⁷. The prior settings and model outputs are
134 presented in SI Appendix Figure S2. The modelled median age was used for calculating and plotting HgAR
135 against time (Figure 1). The average age uncertainties (1-sigma) derived from the age-depth models range
136 from 1-5 years for the topmost part of the cores, up to ca. 100 years around 1000 AD. The investigated
137 peat profiles of AMS, SCB, AND, and HBT cover periods of 6600, 2000, 200 and 800 years, respectively.
138 Corresponding mean peat accumulation rates are 0.76, 0.85, 3.6 and 0.91 mm yr⁻¹ respectively.

139 **Peat Hg accumulation rates (HgAR).** HgAR is calculated as the product of Hg concentration (ng g^{-1}), peat
140 density (g cm^{-3}) and peat mass accumulation rate ($\text{g m}^{-2} \text{yr}^{-1}$). Peat density was determined for each 1 cm
141 slice by measuring its volume using a Vernier caliper and dry peat mass after freeze-drying. Peat samples
142 were analyzed for total Hg (THg) concentration on a combustion cold vapor atomic absorption
143 spectrometer (CV-AAS, Milestone DMA-80) at the University of Toulouse, France. The IPE 176 CRM (Reed
144 / *Phragmites communis*), NIST 1632d (Coal), and BCR 482 (Lichen) were analyzed with mean recoveries
145 ranging from 93-100% (SI Appendix Table S3). Replicate/triplicate analyses of THg in peat samples were
146 found to vary by less than 6% (1σ). Profiles of peat Hg concentration in AMS, SCB, AND, and HBT are shown
147 in SI Appendix Figure S5. Peat mass accumulation rate was determined from the age models and dry peat
148 mass. All raw data is summarized in Extended Data 1.

149 **Literature review, reference time periods and statistics.** We expand on a previous literature review of
150 sediment and peat Hg archives. We examined the remote HgAR records from SH lake sediments and peat
151 records in Southern South America, lake sediments in New Zealand, lake sediments in East Africa, and lake
152 sediments in Antarctica (see Extended data 2 for details). We did not retain: a lake sediment core 6 km
153 downstream from the Potosi mine (Bolivia) with pronounced local mining influences on HgAR⁴⁹; a lake
154 sediment core in the Patagonian volcanic zone with multiple tephra layers associated with high HgAR⁵⁰.
155 Two remote Bolivian cores and one Peruvian core also showed evidence for the release of Hg due to
156 regional Spanish colonial mining activities^{36,51}, but were retained in Extended Data 2. Extended Data 2
157 indicates which records were only partially used, often due to lack of recent ²¹⁰Pb or ¹⁴C bomb pulse dates.
158 This applies in particular to three SH peat records, where one lacks a recent ²¹⁰Pb chronology and therefore
159 20Cmax and pre-industrial HgAR³⁶, one lacks pre-1988 layers⁴⁸, and one is nearly complete³⁵, except for
160 the 1826-1935 period, which we extrapolate (see Extended Data 2).

161 We use four reference time periods, based on previous studies and which were originally
162 operationally derived for NH natural archives¹⁸: natural background (pre-1450AD), pre-industrial period
163 (1450-1880AD), 20th century extended HgAR maximum (20Cmax, approx. 1940-1990), and the recent,
164 modern period (post-1990AD). For each published study, we calculate mean HgAR during the four
165 reference intervals. The operational cut-off years, e.g. 1450, 1880, 1990, are mean values based on the
166 remote NH sediment (n=49) and peat cores (n=19) reviewed here. In other words, each archive and each
167 regional context shows variation in the exact timing of gradual or abrupt increases (~1450, ~1880) or
168 decreases (~1990) in HgAR (Extended Data 2). Several long SH sediment records probe the effect of climate
169 change on variations in HgAR during the Holocene and since the last glacial maximum. Depending on

170 watershed type and location these studies document substantial natural variability in HgAR that is beyond
171 the scope of this study, but no less important. Therefore, in order to assess to the best of our ability the
172 impact of humans on recent, millennial atmospheric Hg enrichment, we integrated natural background
173 HgAR between on average -1700BC to 1450AD, but on occasion as far back as 10,000BC (Extended Data
174 2). We define enrichment factors (EF) based on the evolution of mean HgAR during the four reference
175 periods as follows:

176 $EF_{\text{preind}} = \text{HgAR (20Cmax)} / \text{HgAR (pre-industrial)}$

177 $EF_{\text{alltime}} = \text{HgAR (20Cmax)} / \text{HgAR (natural background)}$

178 $EF_{\text{p/b}} = \text{HgAR (pre-industrial)} / \text{HgAR (natural background)}$

179 $EF_{\text{mod/bck}} = \text{HgAR (modern)} / \text{HgAR (natural background)}$. Statistical descriptions are parametric
180 (mean, standard deviation (SD)) for normally distributed HgAR and EF, and non-parametric (median, Q25%
181 and Q75% quartiles, interquartile range (IQR)) for non-normally distributed HgAR and EF. Outlier tests
182 were performed only on EFs, and observations were excluded (in *italics* in Extended Data 2) when they
183 exceeded 2*SD around the mean, or 1.5*IQR around Q25% and Q75%. All data generated or analyzed
184 during this study are included in the SI Appendix.

185 **Results & Discussion**

186 ***New southern hemisphere peat records***

187 HgAR profiles in the four SH peat records show maximum values during the 20th century (Figure 1). Natural
188 background (pre-1450 AD) HgAR in the HBT, SCB and AMS cores show a mean of $4.9 \pm 3.5 \mu\text{g m}^{-2} \text{yr}^{-1}$ (mean,
189 1σ , $n=33$ in 3 cores, Figure 1). Pre-industrial HgAR in the four cores averages $5.9 \pm 2.5 \mu\text{g m}^{-2} \text{yr}^{-1}$, 20Cmax
190 HgAR is $20 \pm 7.9 \mu\text{g m}^{-2} \text{yr}^{-1}$, and modern HgAR is $9.7 \pm 2.9 \mu\text{g m}^{-2} \text{yr}^{-1}$ (means, 1σ , $n=4$, Figure 1). AND and
191 HBT have more pronounced 20Cmax peaks than SCB and AMS, which is due to a combination of peaks in
192 Hg concentration (Figure S5) and enhanced peat mass accumulation rate occurring simultaneously
193 (Extended Data 1). Whereas absolute HgAR for the different time periods vary between cores, the relative
194 HgAR changes between cores are similar and can be expressed by enrichment factors, EF. The four SH
195 cores show evidence for 3.1-fold (mean, $1\sigma=1.6$) enhanced net Hg deposition during the 20Cmax,
196 compared to the pre-industrial period (EF_{preind} , Table 1), which at first sight appears similar to NH natural
197 archives. SH historical HgARs have thus far been studied in 20 lake sediment and 3 peat cores (see Methods
198 and Extended Data 2 for full list). Figure 2 summarizes HgAR and EF's in all published SH sediment and peat
199 records, as well as updated NH data for the reference periods of interest (Extended Data 2). The temporal

200 evolution of HgAR in peat and sediment cores is similar between the NH and SH in a broad sense (Figure
201 2a, b). HgAR increases stepwise from natural background to pre-industrial and then to 20Cmax periods in
202 both sediment and peat archives. Similar to NH peat records¹⁸, modern-day (post-1990) HgAR in SH peat
203 decreases by a factor of 2 from 20Cmax values (SI Appendix Figure S4), in line with declining global
204 anthropogenic Hg emissions and deposition from the 1970s to 2000s (Figure S6^{25,26}). Sediment records in
205 both the NH and SH do not record this decrease (Figure S4), presumably due to the longer residence of Hg
206 in lake catchment soils, leading to a slower recovery of Hg concentrations in soil run-off into lakes¹⁸.

207

208 ***Hemispheric trends in historical Hg enrichment***

209 The historical evolution of trends in hemispheric HgARs are shown in EF_{preind} and $EF_{alltime}$ diagrams (Figure
210 2c, 2d). Pre-industrial to 20Cmax enrichment in HgAR (EF_{preind}) is higher in peat compared to sediment in
211 both NH and SH (Kruskal-Wallis test, NH, $P=0.01$; SH, $P=0.10$). EF_{preind} is higher in the NH than in the SH for
212 sediment (3.1 vs 1.8), but not peat (4.6 vs 3.1; Kruskal-Wallis test, peat, $P=0.15$; sediment $P=0.001$; Figure
213 2c, 2d; Figure 3a). We find in particular that in long, millennial NH records, HgAR increased 3.9-fold in peat
214 and 3.7-fold in sediments across the natural background to pre-industrial periods around 1450AD ($EF_{p/b}$,
215 Figure 2c, d, Table 2). On the contrary, $EF_{p/b}$ in SH millennial records show negligible, mean 1.2-fold
216 enrichment in peat, to a small, median 1.4-fold enrichment in sediments across the natural background
217 (<1450AD) to pre-industrial periods. Consequently, all-time NH enrichment factors, $EF_{alltime}$, reach 16 in
218 peat and 13 in sediments and are larger than the 6.0-fold and 3.8-fold Hg all-time enrichment in SH peat
219 and sediments (Table 2; Figure 3B; Kruskal-Wallis test, $P = 0.02$ for peat, $P = 0.09$ for sediment). Historical
220 Hg emission inventory and associated box modeling studies have suggested that the 4-fold increase in NH
221 HgAR around 1450AD is related to Spanish colonial Hg and silver mining^{7,27}. This interpretation has been
222 questioned by studies arguing that the associated emissions are overestimated^{1,9,52}. SH archives show
223 little evidence of Spanish colonial mining impacts in South-America on large scale SH atmospheric Hg
224 deposition (Figure 2). Similarly, neither NH peat, nor sediment records show evidence of a pronounced
225 late 19th century peak in HgAR, in contrast to large estimated N-American gold-rush Hg emissions⁷. Across
226 the natural background and pre-industrial reference periods, the world's global population increased 5-
227 fold from 0.22 to 1.2 billion⁵³. We therefore suggest the 4-fold NH increase in HgAR around 1450AD is
228 more likely related to demography driven changes in land-use and associated Hg emissions and deposition
229 (e.g. deforestation²⁵, wood and peat combustion, urbanization), than to direct Spanish colonial mining
230 emissions of Hg to the global pool. More research is needed to explore this in detail. In summary, our

231 findings based on combined sediment and peat archive HgAR observations, suggest that all-time
232 atmospheric Hg enrichment during the 20Cmax period (1940-1990) reached 11-fold globally ($EF_{alltime} = 4-$
233 24 , 25%-75% quartiles, $n=39$), 16-fold in the NH ($EF_{alltime} = 10-30$, 25%-75% quartiles, $n=26$), and 4-fold in
234 the SH ($EF_{alltime} = 2-6$, 25%-75% quartiles, $n=13$). Atmospheric Hg concentrations decreased from the 1970's
235 to the 2000's by a factor of about 2, a trend that is recorded in the peat archive HgAR (Figure S4, S6).
236 Natural background to modern period (1990-2010) Hg enrichment, $EF_{mod/bck}$, based on peat archives, is
237 currently 10-fold globally (± 7.7 , 1σ , $n=18$), 12 in the NH (± 7.5 , 1σ , $n=14$) and 3 in the SH (± 2.5 , 1σ , $n=4$).

238

239 ***Natural and anthropogenic hemispheric Hg emissions***

240 In the following sections we will further discuss this sizeable difference in hemispheric $EF_{alltime}$ in terms
241 of NH and SH Hg emissions, and in terms of natural background HgAR. The all-time NH and SH enrichment
242 factors, based on Hg deposition to natural archives, can be directly compared to independent estimates
243 of NH and SH emission factors, i.e. $EF_{emission}$, the ratio of primary, i.e. first time, total Hg emission flux to
244 natural Hg emission flux ($EF_{emission} = F_{anthro} + F_{natural} / F_{natural}$; Table 3). Streets et al. estimated global
245 anthropogenic Hg emissions to the atmosphere of $2.4 \pm 0.5 \text{ Gg yr}^{-1}$ during the 20Cmax period (1940-1990)
246 ⁷. Natural Hg emissions are the sum of volcanic degassing and crustal degassing from naturally enriched
247 soils. Passive, non-eruptive, volcanic degassing is an important direct natural source of Hg to the
248 atmosphere, with a previously estimated total flux of $76 \pm 30 \text{ Mg yr}^{-1}$ (1σ) based on observed Hg/SO₂ ratios
249 of $7.8 \pm 1.5 \times 10^{-6}$ and a global passive degassing SO₂ flux of 9.7 Tg yr^{-1} ^{54,55}. Recent advances in remote
250 sensing of SO₂ from 2005-2015 indicate a higher SO₂ flux of $23.0 \pm 2.3 \text{ Tg yr}^{-1}$ (1σ) ⁵⁶, which we use here to
251 revise the global passive volcanic degassing Hg flux to $179 \pm 39 \text{ Mg yr}^{-1}$ (1σ). Eruptive volcanic SO₂ emissions
252 are indicated to be one order of magnitude smaller than passive degassing at $2.6 \pm 2.6 \text{ Tg yr}^{-1}$ ⁵⁶. Assuming
253 similar Hg/SO₂ ratios, we estimate eruptive volcanic Hg emissions at $20 \pm 20 \text{ Mg yr}^{-1}$, and total volcanic Hg
254 emissions as the sum of eruptive and passive emissions at $200 \pm 60 \text{ Mg yr}^{-1}$ (1σ). Global emissions from
255 naturally enriched soils can be estimated from reviews of flux chamber and soil Hg studies ^{57,58} and equal
256 $135 \pm 40 \text{ Mg yr}^{-1}$ (1σ , Table 3). These estimates indicate that global anthropogenic 20Cmax Hg emissions
257 of 2.4 Gg yr^{-1} have been 7.3 times larger than global natural Hg emissions of 0.34 Gg yr^{-1} , and result in a
258 global $EF_{emission}$ of 8.2. Volcanic SO₂ emissions are similar for the NH and SH (11.8 vs. 11.2 Tg yr^{-1}) ⁵⁶, leading
259 to NH and SH Hg emission budgets of 0.1 Gg yr^{-1} each. We scale naturally enriched soil emissions with
260 continental surface area, to estimate 91 and 44 Mg yr^{-1} in NH and SH. The 20Cmax 2.4 Gg yr^{-1} global
261 anthropogenic Hg emissions to the atmosphere were released for 80% to the NH and 20% to the SH ⁷. We

262 therefore estimate hemispheric EF_{emission} , for the NH at 11.2 ± 4.6 and for the SH at 4.4 ± 1.5 (1σ). The SH
263 EF_{emission} of 4.4 is in good agreement with the natural archive-based SH EF_{alltime} of 4. The NH EF_{emission} of 11
264 however, underestimates the NH EF_{alltime} of 16 by 43%, suggesting that either the 2.0 ± 0.5 Gg yr⁻¹ NH
265 anthropogenic Hg emissions to air⁷ are underestimated, or that the NH natural primary emissions of $91 \pm$
266 27 Mg yr⁻¹ are overestimated, or that inter-hemispheric exchange has transported NH anthropogenic Hg
267 to the SH. There is a final caveat in this analysis that deserves a mention: We assume that the ill-
268 constrained, but potentially important, submarine volcanic Hg flux⁵⁹ is locally or regionally deposited to
269 marine sediments before any of it can be emitted to the atmosphere. This assumption is based on evidence
270 for Hg scavenging in submarine hydrothermal plumes^{60,61}.

271 The most recent, 2018 UNEP global Hg assessment, which provides the state of the science basis for
272 the implementation of the UNEP Minamata Convention on Mercury, states that “Human activities have
273 increased total atmospheric Hg concentrations by about 450% (i.e. a factor 4.5) above natural levels.”¹⁴.
274 Our findings therefore suggest that modern (1990-2010) atmospheric Hg enrichment is larger, 10-fold
275 globally. In addition we find consistently lower anthropogenic Hg enrichment in emissions and in
276 deposition in the SH compared to the NH.

277

278 ***Hemispheric differences in Hg deposition and cycling***

279 The important difference in NH and SH EF_{alltime} is not only related to hemispheric differences in primary Hg
280 emissions, but also to differences in natural background atmospheric Hg concentrations and HgAR. A
281 notable outcome of the new SH peat records is that the natural SH background HgAR of $4.3 \mu\text{g m}^{-2} \text{yr}^{-1}$ in
282 the SH mid-latitudes (30-60°S) is x2.5 higher than the NH background HgAR of $1.7 \mu\text{g m}^{-2} \text{yr}^{-1}$ in the NH
283 mid-latitudes (Kruskal-Wallis test, $P=0.02$, Figures 2a, 3c, S3). Recent Hg stable isotope work on Hg
284 deposition to vegetation and soils suggests that 75% derives from direct uptake of atmospheric Hg(0), and
285 less from Hg(II) wet deposition^{23,25,62,63}. Consequently, peat vegetation Hg(0) uptake is primarily driven by
286 atmospheric Hg(0) concentration and primary productivity²⁵. Peat vegetation primary productivity
287 depends on climate, which, at the NH and SH mid-latitude sites we study and review, shows similar mean
288 annual air temperatures (NH, 6.7; SH, 6.3 °C), precipitation (NH, 1110; SH, 1120 mm yr⁻¹) and cloud cover
289 (NH, 72; SH 77%)⁶⁴. We therefore suggest that the marked NH/SH mid-latitude difference in HgAR is driven
290 by x2.5 higher natural atmospheric Hg concentrations in the SH, rather than climate factors. Climate
291 factors, such as temperature and length of growth season only become visible in NH high latitude (>60°N),
292 where HgAR becomes limited by peat bog primary productivity, via the vegetation Hg⁰ pump²⁴. The

293 observation that the SH natural background HgAR is x2.5 higher than the NH background is likely an
294 additional reason why the NH $EF_{alltime}$ of 16 is much larger than the SH $EF_{alltime}$ of 4.

295 Inter-hemispheric trends in atmospheric Hg have been previously investigated^{65,66}. Observed mean
296 atmospheric Hg⁰ concentrations across monitoring networks for the modern, 1990-2010 period were 1.8
297 ng m⁻³ in the NH and 1.2 ng m⁻³ in the SH^{67,68}. Modern-day SH Hg⁰ concentrations are therefore higher
298 than what would be expected based on estimates of modern NH and SH primary Hg emissions of 1.6 and
299 0.7 Gg yr⁻¹ (Table 3). Inter-hemispheric transport of NH Hg⁰ to the SH potentially contributes to the high
300 SH Hg⁰ concentrations. A key difference between the NH and SH is the land-ocean distribution, with the
301 SH being only 19% land covered and the NH 39%. The land-ocean distribution plays an important role in
302 atmospheric boundary layer Hg dynamics. A study on atmospheric Hg⁰ seasonality, which is more
303 pronounced in the NH and quasi-absent in the SH, suggested that the vegetation Hg pump, i.e. the foliar
304 uptake of Hg⁰ and sequestration in soils, is an important driver of NH atmospheric Hg⁰ seasonality²⁴. The
305 SH has a smaller terrestrial vegetation and soil pool, and we speculate that the SH has relatively higher
306 atmospheric Hg⁰ due to a weaker vegetation Hg pump. In addition coupled ocean-atmosphere Hg
307 chemistry and transport models find stronger marine Hg⁰ evasion in the SH than in the NH, mainly due to
308 upwelling of Hg rich deep waters in the Southern Ocean^{19,69}. The model studies suggest that SH
309 atmospheric Hg⁰ is largely controlled by these SH marine Hg⁰ emissions^{8,19}. These findings were recently
310 confirmed by long-term observations on Hg⁰ seasonality at the Cape Point, South-Africa monitoring station
311⁷⁰. The 2-fold higher SH natural background HgAR in peat therefore echoes the higher than expected
312 modern SH atmospheric Hg⁰ concentrations, and both can potentially be explained by the hemispheric
313 land-ocean distribution.

314 We use peat $EF_{mod/bck}$ for both hemispheres (Table 2) to estimate what natural atmospheric Hg⁰
315 concentrations may have been during pre-1450AD times. Dividing modern-day mean NH and SH
316 atmospheric Hg⁰ concentrations of 1.8 and 1.2 ng m⁻³ by $EF_{mod/bck}$ yields natural background atmospheric
317 Hg concentrations of 0.2 and 0.4 ng m⁻³ for the NH and SH. In summary, the lower SH enrichment in
318 atmospheric Hg appears to be caused by a combination of lower SH anthropogenic Hg emissions, and
319 higher SH background Hg concentrations. We speculate that the higher SH atmospheric background is
320 driven by a lower SH land/ocean ratio which limits the terrestrial vegetation Hg pump and sustains higher
321 natural marine Hg emissions. Overall, our findings suggest that both background Hg concentrations and
322 all-time Hg enrichment in the NH and SH are different and should be taken into account in environmental
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338 **Author Contributions**

339 J.E.S and F.D.V initiated and designed the project. All authors were involved in field sampling, laboratory
340 analyses, and/or data analysis. C.L. and J.E.S wrote the manuscript on which all authors commented.

341 **Data availability statement**

342 All data generated or analyzed during this study are included in this published article (and its SI
343 Appendix).

344 **References**

- 345 (1) Outridge, P. M.; Mason, R. P.; Wang, F.; Guerrero, S.; Heimbürger-Boavida, L. E. Updated Global
346 and Oceanic Mercury Budgets for the United Nations Global Mercury Assessment 2018. *Environ.*
347 *Sci. Technol.* **2018**, *52* (20), 11466–11477. <https://doi.org/10.1021/acs.est.8b01246>.
- 348 (2) Mason, R. P.; Choi, A. L.; Fitzgerald, W. F.; Hammerschmidt, C. R.; Lamborg, C. H.; Soerensen, A. L.;
349 Sunderland, E. M. Mercury Biogeochemical Cycling in the Ocean and Policy Implications. *Environ.*
350 *Res.* **2012**, *119*, 101–117. <https://doi.org/10.1016/j.envres.2012.03.013>.
- 351 (3) Chen, C.; Amirbahman, A.; Fisher, N.; Harding, G.; Lamborg, C.; Nacci, D.; Taylor, D.
352 Methylmercury in Marine Ecosystems: Spatial Patterns and Processes of Production,
353 Bioaccumulation, and Biomagnification. *Ecohealth* **2008**, *5* (4), 399–408.
354 <https://doi.org/10.1007/s10393-008-0201-1>.
- 355 (4) Sunderland, E. M. Mercury Exposure from Domestic and Imported Estuarine and Marine Fish in
356 the U.S. Seafood Market. *Environ. Health Perspect.* **2007**, *115* (235–242).
- 357 (5) Pirrone, N.; Cinnirella, S.; Feng, X.; Finkelman, R. B.; Friedli, H. R.; Leaner, J.; Mason, R.;
358 Mukherjee, A. B.; Stracher, G. B.; Streets, D. G.; Telmer, K. Global Mercury Emissions to the

- 359 Atmosphere from Anthropogenic and Natural Sources. *Atmospheric Chem. Phys.* **2010**, *10* (13),
360 5951–5964. <https://doi.org/10.5194/acp-10-5951-2010>.
- 361 (6) Pacyna, E. G.; Pacyna, J. M.; Sundseth, K.; Munthe, J.; Kindbom, K.; Wilson, S.; Steenhuisen, F.;
362 Maxson, P. Global Emission of Mercury to the Atmosphere from Anthropogenic Sources in 2005
363 and Projections to 2020. *Atmos. Environ.* **2010**, *44* (20), 2487–2499.
364 <https://doi.org/10.1016/j.atmosenv.2009.06.009>.
- 365 (7) Streets, D. G.; Horowitz, H. M.; Jacob, D. J.; Lu, Z.; Levin, L.; ter Schure, A. F. H.; Sunderland, E. M.
366 Total Mercury Released to the Environment by Human Activities. *Environ. Sci. Technol.* **2017**, *51*
367 (11), 5969–5977. <https://doi.org/10.1021/acs.est.7b00451>.
- 368 (8) Horowitz, H. M.; Jacob, D. J.; Zhang, Y.; Dibble, T. S.; Slemr, F.; Amos, H. M.; Schmidt, J. A.; Corbitt,
369 E. S.; Marais, E. A.; Sunderland, E. M. A New Mechanism for Atmospheric Mercury Redox
370 Chemistry: Implications for the Global Mercury Budget. *Atmospheric Chem. Phys.* **2017**, *17* (10),
371 6353–6371. <https://doi.org/10.5194/acp-17-6353-2017>.
- 372 (9) Engstrom, D. R.; Fitzgerald, W. F.; Cooke, C. A.; Lamborg, C. H.; Drevnick, P. E.; Swain, E. B.;
373 Balogh, S. J.; Balcom, P. H. Atmospheric Hg Emissions from Preindustrial Gold and Silver Extraction
374 in the Americas: A Reevaluation from Lake-Sediment Archives. *Environ. Sci. Technol.* **2014**, *48* (12),
375 6533–6543. <https://doi.org/10.1021/es405558e>.
- 376 (10) Fitzgerald, W. F.; Engstrom, D. R.; Mason, R. P.; Nater, E. A. The Case for Atmospheric Mercury
377 Contamination in Remote Areas. *Environ. Sci. Technol.* **1998**, *32* (1), 1–7.
378 <https://doi.org/10.1021/es970284w>.
- 379 (11) Martinez-Cortizas, A. Mercury in a Spanish Peat Bog: Archive of Climate Change and Atmospheric
380 Metal Deposition. *Science* **1999**, *284* (5416), 939–942.
381 <https://doi.org/10.1126/science.284.5416.939>.
- 382 (12) Schuster, P. F.; Krabbenhoft, D. P.; Naftz, D. L.; Cecil, L. D.; Olson, M. L.; Dewild, J. F.; Susong, D. D.;
383 Green, J. R.; Abbott, M. L. Atmospheric Mercury Deposition during the Last 270 Years: A Glacial
384 Ice Core Record of Natural and Anthropogenic Sources. *Environ. Sci. Technol.* **2002**, *36* (11), 2303–
385 2310. <https://doi.org/10.1021/es0157503>.
- 386 (13) Thomas, R. L. The Distribution of Mercury in the Sediments of Lake Ontario. *Can. J. Earth Sci.*
387 **1972**, *9* (6), 636–651. <https://doi.org/10.1139/e72-054>.
- 388 (14) UNEP. *Global Mercury Assessment 2018*; 2018.
- 389 (15) Fitzgerald, W. F.; Engstrom, D. R.; Mason, R. P.; Nater, E. A. The Case for Atmospheric Mercury
390 Contamination in Remote Areas. *Environ. Sci. Technol.* **1998**, *32* (1), 1–7.
391 <https://doi.org/10.1021/es970284w>.
- 392 (16) Engstrom, D. R.; Fitzgerald, W. F.; Cooke, C. A.; Lamborg, C. H.; Drevnick, P. E.; Swain, E. B.;
393 Balogh, S. J.; Balcom, P. H. Atmospheric Hg Emissions from Preindustrial Gold and Silver Extraction
394 in the Americas: A Reevaluation from Lake-Sediment Archives. *Environ. Sci. Technol.* **2014**, *48* (12),
395 6533–6543. <https://doi.org/10.1021/es405558e>.
- 396 (17) Biester, H.; Bindler, R.; Martinez-Cortizas, A.; Engstrom, D. R. Modeling the Past Atmospheric
397 Deposition of Mercury Using Natural Archives. *Environ. Sci. Technol.* **2007**, *41* (14), 4851–4860.
398 <https://doi.org/10.1021/es0704232>.
- 399 (18) Amos, H. M.; Sonke, J. E.; Obrist, D.; Robins, N.; Hagan, N.; Horowitz, H. M.; Mason, R. P.; Witt, M.;
400 Hedgecock, I. M.; Corbitt, E. S.; Sunderland, E. M. Observational and Modeling Constraints on
401 Global Anthropogenic Enrichment of Mercury. *Environ. Sci. Technol.* **2015**, *49* (7), 4036–4047.
402 <https://doi.org/10.1021/es5058665>.
- 403 (19) Zhang, Y.; Jaeglé, L.; Thompson, L.; Streets, D. G. Six Centuries of Changing Oceanic Mercury:
404 Anthropogenic Mercury in Ocean. *Glob. Biogeochem. Cycles* **2014**, *28* (11), 1251–1261.
405 <https://doi.org/10.1002/2014GB004939>.

- 406 (20) Biester, H.; Bindler, R.; Martinez-Cortizas, A.; Engstrom, D. R. Modeling the Past Atmospheric
407 Deposition of Mercury Using Natural Archives. *Environ. Sci. Technol.* **2007**, *41* (14), 4851–4860.
408 <https://doi.org/10.1021/es0704232>.
- 409 (21) Cooke, C. A.; Hobbs, W. O.; Michelutti, N.; Wolfe, A. P. Reliance on Pb-210 Chronology Can
410 Compromise the Inference of Preindustrial Hg Flux to Lake Sediments. *Environ. Sci. Technol.* **2010**,
411 *44* (6), 1998–2003. <https://doi.org/10.1021/es9027925>.
- 412 (22) Abril, J. M.; Brunskill, G. Evidence That Excess 210Pb Flux Varies with Sediment Accumulation Rate
413 and Implications for Dating Recent Sediments. *J. Paleolimnol.* **2014**, *52*.
414 <https://doi.org/10.1007/s10933-014-9782-6>.
- 415 (23) Enrico, M.; Roux, G. L.; Maruszczak, N.; Heimbürger, L.-E.; Claustres, A.; Fu, X.; Sun, R.; Sonke, J. E.
416 Atmospheric Mercury Transfer to Peat Bogs Dominated by Gaseous Elemental Mercury Dry
417 Deposition. *Environ. Sci. Technol.* **2016**, *50* (5), 2405–2412.
418 <https://doi.org/10.1021/acs.est.5b06058>.
- 419 (24) Jiskra, M.; Sonke, J. E.; Obrist, D.; Bieser, J.; Ebinghaus, R.; Myhre, C. L.; Pfaffhuber, K. A.;
420 Wängberg, I.; Kyllönen, K.; Worthy, D.; Martin, L. G.; Labuschagne, C.; Mkololo, T.; Ramonet, M.;
421 Magand, O.; Dommergue, A. A Vegetation Control on Seasonal Variations in Global Atmospheric
422 Mercury Concentrations. *Nat. Geosci.* **2018**, *11* (4), 244–250. [https://doi.org/10.1038/s41561-](https://doi.org/10.1038/s41561-018-0078-8)
423 [018-0078-8](https://doi.org/10.1038/s41561-018-0078-8).
- 424 (25) Enrico, M.; Le Roux, G.; Heimbürger, L.-E.; Van Beek, P.; Souhaut, M.; Chmeleff, J.; Sonke, J. E.
425 Holocene Atmospheric Mercury Levels Reconstructed from Peat Bog Mercury Stable Isotopes.
426 *Environ. Sci. Technol.* **2017**, *51* (11), 5899–5906. <https://doi.org/10.1021/acs.est.6b05804>.
- 427 (26) European Monitoring and Evaluation Programme. <https://www.emep.int/> 2016.
- 428 (27) Streets, D. G.; Devane, M. K.; Lu, Z.; Bond, T. C.; Sunderland, E. M.; Jacob, D. J. All-Time Releases
429 of Mercury to the Atmosphere from Human Activities. *Environ. Sci. Technol.* **2011**, *45* (24), 10485–
430 10491. <https://doi.org/10.1021/es202765m>.
- 431 (28) Streets, D. G.; Horowitz, H. M.; Lu, Z.; Levin, L.; Thackray, C. P.; Sunderland, E. M. Five Hundred
432 Years of Anthropogenic Mercury: Spatial and Temporal Release Profiles. *Environ. Res. Lett.* **2019**,
433 *14* (8), 084004. <https://doi.org/10.1088/1748-9326/ab281f>.
- 434 (29) Amos, H. M.; Jacob, D. J.; Streets, D. G.; Sunderland, E. M. Legacy Impacts of All-Time
435 Anthropogenic Emissions on the Global Mercury Cycle: GLOBAL IMPACTS OF LEGACY MERCURY.
436 *Glob. Biogeochem. Cycles* **2013**, *27* (2), 410–421. <https://doi.org/10.1002/gbc.20040>.
- 437 (30) Cooke, C. A.; Hintelmann, H.; Ague, J. J.; Burger, R.; Biester, H.; Sachs, J. P.; Engstrom, D. R. Use
438 and Legacy of Mercury in the Andes. *Environ. Sci. Technol.* **2013**, *47* (9), 4181–4188.
439 <https://doi.org/10.1021/es3048027>.
- 440 (31) Streets, D. G.; Horowitz, H. M.; Jacob, D.; Lu, Z.; Levin, L.; ter Schure, A. F. H.; Sunderland, E. M.
441 Total Mercury Released to the Environment by Human Activities. *Environ. Sci. Technol.* **2017**, *51*
442 (11), 5969–5977. <https://doi.org/10.1021/acs.est.7b00451>.
- 443 (32) Amos, H. M.; Sonke, J. E.; Obrist, D.; Robins, N.; Hagan, N.; Horowitz, H. M.; Mason, R. P.; Witt, M.;
444 Hedgecock, I. M.; Corbitt, E. S.; Sunderland, E. M. Observational and Modeling Constraints on
445 Global Anthropogenic Enrichment of Mercury. *Environ. Sci. Technol.* **2015**, *49* (7), 4036–4047.
446 <https://doi.org/10.1021/es5058665>.
- 447 (33) Cooke, C. A.; Martínez-Cortizas, A.; Bindler, R.; Gustin, M. S. Environmental Archives of
448 Atmospheric Hg Deposition – A Review. *Sci. Total Environ.* **2020**, *709*, 134800.
449 <https://doi.org/10.1016/j.scitotenv.2019.134800>.
- 450 (34) Lindberg, S.; Bullock, R.; Ebinghaus, R.; Engstrom, D.; Feng, X.; Fitzgerald, W.; Pirrone, N.; Prestbo,
451 E.; Seigneur, C. A Synthesis of Progress and Uncertainties in Attributing the Sources of Mercury
452 Deposition. *Ambio* **2007**, *36*, 19–32.

- 453 (35) Biester, H.; Kilian, R.; Franzen, C.; Woda, C.; Mangini, A.; Schöler, H. F. Elevated Mercury
454 Accumulation in a Peat Bog of the Magellanic Moorlands, Chile (53°S) – an Anthropogenic Signal
455 from the Southern Hemisphere. *Earth Planet. Sci. Lett.* **2002**, *201* (3–4), 609–620.
456 [https://doi.org/10.1016/S0012-821X\(02\)00734-3](https://doi.org/10.1016/S0012-821X(02)00734-3).
- 457 (36) Guédron, S.; Ledru, M.-P.; Escobar-Torrez, K.; Develle, A. L.; Brisset, E. Enhanced Mercury
458 Deposition by Amazonian Orographic Precipitation: Evidence from High-Elevation Holocene
459 Records of the Lake Titicaca Region (Bolivia). *Palaeogeogr. Palaeoclimatol. Palaeoecol.* **2018**, *511*,
460 577–587. <https://doi.org/10.1016/j.palaeo.2018.09.023>.
- 461 (37) Le Roux, G.; Vleeschouwer, F. Preparation of Peat Samples for Inorganic Geochemistry Used as
462 Palaeoenvironmental Proxies. *Mires Peat* **2011**, *7*.
- 463 (38) Givélet, N.; Le Roux, G.; Cheburkin, A.; Chen, B.; Frank, J.; Goodsite, M.; Kempter, H.; Krachler, M.;
464 Noernberg, T.; Rausch, N.; Rheinberger, S.; Roos-Barraclough, F.; Sapkota, A.; Scholz, C.; Shotyk,
465 W. Suggested Protocol for Collecting, Handling and Preparing Peat Cores and Peat Samples for
466 Physical, Chemical, Mineralogical and Isotopic Analyses. *J. Environ. Monit.* **2004**, *6* (5), 481–492.
467 <https://doi.org/10.1039/b401601g>.
- 468 (39) Li, C.; Sonke, J. E.; Le Roux, G.; Van der Putten, N.; Piotrowska, N.; Jeandel, C.; Mattielli, N.; Benoit,
469 M.; Wiggs, G. F. S.; De Vleeschouwer, F. Holocene Dynamics of the Southern Westerly Winds over
470 the Indian Ocean Inferred from a Peat Dust Deposition Record. *Quat. Sci. Rev.* **2020**, *231*, 106169.
471 <https://doi.org/10.1016/j.quascirev.2020.106169>.
- 472 (40) Goodsite, M. E.; Rom, W.; Heinemeier, J.; Lange, T.; Ooi, S.; Appleby, P. G.; Shotyk, W.; van der
473 Knaap, W. O.; Lohse, C.; Hansen, T. S. High-Resolution AMS ¹⁴C Dating of Post-Bomb Peat Archives
474 of Atmospheric Pollutants. *Radiocarbon* **2001**, *43* (2B), 495–515.
475 <https://doi.org/10.1017/S0033822200041163>.
- 476 (41) Appleby, P. G. Chronostratigraphic Techniques in Recent Sediments. In *Tracking Environmental*
477 *Change Using Lake Sediments*; Last, W. M., Smol, J. P., Eds.; Kluwer Academic Publishers:
478 Dordrecht, 2002; Vol. 1, pp 171–203. https://doi.org/10.1007/0-306-47669-X_9.
- 479 (42) Piotrowska, N. Status Report of AMS Sample Preparation Laboratory at GADAM Centre, Gliwice,
480 Poland. *Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. At.* **2013**, *294*, 176–181.
481 <https://doi.org/10.1016/j.nimb.2012.05.017>.
- 482 (43) Zoppi, U.; Crye, J.; Song, Q.; Arjomand, A. Performance Evaluation of the New AMS System at
483 Accium BioSciences. *Radiocarbon* **2007**, *49* (1), 171–180.
484 <https://doi.org/10.1017/S0033822200041990>.
- 485 (44) Davies, L. J.; Appleby, P.; Jensen, B. J. L.; Magnan, G.; Mullan-Boudreau, G.; Noernberg, T.;
486 Shannon, B.; Shotyk, W.; van Bellen, S.; Zaccone, C.; Froese, D. G. High-Resolution Age Modelling
487 of Peat Bogs from Northern Alberta, Canada, Using Pre- and Post-Bomb ¹⁴C, ²¹⁰Pb and Historical
488 Cryptotephra. *Quat. Geochronol.* **2018**, *47*, 138–162.
489 <https://doi.org/10.1016/j.quageo.2018.04.008>.
- 490 (45) Blaauw, M.; Christen, J. A. Flexible Paleoclimate Age-Depth Models Using an Autoregressive
491 Gamma Process. *Bayesian Anal.* **2011**, *6* (3), 457–474. <https://doi.org/10.1214/11-BA618>.
- 492 (46) Hogg, A. G.; Hua, Q.; Blackwell, P. G.; Niu, M.; Buck, C. E.; Guilderson, T. P.; Heaton, T. J.; Palmer, J.
493 G.; Reimer, P. J.; Reimer, R. W.; Turney, C. S. M.; Zimmerman, S. R. H. SHCal13 Southern
494 Hemisphere Calibration, 0–50,000 Years Cal BP. *Radiocarbon* **2013**, *55* (4), 1889–1903.
495 https://doi.org/10.2458/azu_js_rc.55.16783.
- 496 (47) Hua, Q.; Barbetti, M.; Rakowski, A. Z. Atmospheric Radiocarbon for the Period 1950–2010.
497 *Radiocarbon* **2013**, *55* (4), 2059–2072. https://doi.org/10.2458/azu_js_rc.v55i2.16177.
- 498 (48) Lamborg, C. H.; Fitzgerald, W. F.; Damman, A. W. H.; Benoit, J. M.; Balcom, P. H.; Engstrom, D. R.
499 Modern and Historic Atmospheric Mercury Fluxes in Both Hemispheres: Global and Regional

- 500 Mercury Cycling Implications: MODERN AND HISTORIC FLUXES OF ATMOSPHERIC MERCURY. *Glob.*
501 *Biogeochem. Cycles* **2002**, *16* (4), 51-1-51–11. <https://doi.org/10.1029/2001GB001847>.
- 502 (49) Cooke, C. A.; Balcom, P. H.; Kerfoot, C.; Abbott, M. B.; Wolfe, A. P. Pre-Colombian Mercury
503 Pollution Associated with the Smelting of Argentiferous Ores in the Bolivian Andes. *AMBIO* **2011**,
504 *40* (1), 18–25. <https://doi.org/10.1007/s13280-010-0086-4>.
- 505 (50) Ribeiro Guevara, S.; Meili, M.; Rizzo, A.; Daga, R.; Arribére, M. Sediment Records of Highly
506 Variable Mercury Inputs to Mountain Lakes in Patagonia during the Past Millennium. *Atmospheric*
507 *Chem. Phys.* **2010**, *10* (7), 3443–3453. <https://doi.org/10.5194/acp-10-3443-2010>.
- 508 (51) Cooke, C. A.; Balcom, P. H.; Biester, H.; Wolfe, A. P. Over Three Millennia of Mercury Pollution in
509 the Peruvian Andes. *Proc. Natl. Acad. Sci.* **2009**, *106* (22), 8830–8834.
510 <https://doi.org/10.1073/pnas.0900517106>.
- 511 (52) Guerero, S. Chemistry as a Tool for Historical Research: Identifying Paths of Historical Mercury
512 Pollution in the Hispanic New World. *Bull Hist Chem* **2012**, *37*, 61–70.
- 513 (53) Roser, M.; Ritchie, H.; Ortiz-Ospina, E. World Population Growth. Published Online at
514 OurWorldInData.Org. Retrieved from: “<https://Ourworldindata.Org/World-Population-Growth>.”
515 2013.
- 516 (54) Bagnato, E.; Tamburello, G.; Avar, G.; Martinez-Cruz, M.; Enrico, M.; Fu, X.; Sprovieri, M.; Sonke,
517 J. E. Mercury Fluxes from Volcanic and Geothermal Sources: An Update. *Geol. Soc. Lond. Spec.*
518 *Publ.* **2015**, *410* (1), 263–285. <https://doi.org/10.1144/SP410.2>.
- 519 (55) Andres, R. J.; Kasgnoc, A. D. A Time-Averaged Inventory of Subaerial Volcanic Sulfur Emissions. *J.*
520 *Geophys. Res. Atmospheres* **1998**, *103* (D19), 25251–25261. <https://doi.org/10.1029/98JD02091>.
- 521 (56) Carn, S. A.; Fioletov, V. E.; McLinden, C. A.; Li, C.; Krotkov, N. A. A Decade of Global Volcanic SO₂
522 Emissions Measured from Space. *Sci. Rep.* **2017**, *7* (1), 44095. <https://doi.org/10.1038/srep44095>.
- 523 (57) Kocman, D.; Horvat, M.; Pirrone, N.; Cinnirella, S. Contribution of Contaminated Sites to the
524 Global Mercury Budget. *Environ. Res.* **2013**, *125*, 160–170.
525 <https://doi.org/10.1016/j.envres.2012.12.011>.
- 526 (58) Agnan, Y.; Le Dantec, T.; Moore, C. W.; Edwards, G. C.; Obrist, D. New Constraints on Terrestrial
527 Surface–Atmosphere Fluxes of Gaseous Elemental Mercury Using a Global Database. *Environ. Sci.*
528 *Technol.* **2016**, *50* (2), 507–524. <https://doi.org/10.1021/acs.est.5b04013>.
- 529 (59) Fitzgerald, W.F. and Lamborg, C.H.,. Geochemistry of Mercury. Treatise on Geochemistry: Volume
530 9: Environmental Geochemistry; 2004.
- 531 (60) Lamborg, C. H.; Von Damm, K. L.; Fitzgerald, W. F.; Hammerschmidt, C. R.; Zierenberg, R. Mercury
532 and Methylmercury in Fluids from Sea Cliff Submarine Hydrothermal Field, Gorda Ridge.
533 *Geophys. Res. Lett.* **2006**, *33* (17), L17606. <https://doi.org/10.1029/2006GL026321>.
- 534 (61) Bowman, K. L.; Hammerschmidt, C. R.; Lamborg, C. H.; Swarr, G. J.; Agather, A. M. Distribution of
535 Mercury Species across a Zonal Section of the Eastern Tropical South Pacific Ocean (U.S.
536 GEOTRACES GP16). *Mar. Chem.* **2016**, *186*, 156–166.
537 <https://doi.org/10.1016/j.marchem.2016.09.005>.
- 538 (62) Demers, J. D.; Blum, J. D.; Zak, D. R. Mercury Isotopes in a Forested Ecosystem: Implications for
539 Air-Surface Exchange Dynamics and the Global Mercury Cycle. *Glob. Biogeochem. Cycles* **2013**, *27*
540 (1), 222–238. <https://doi.org/10.1002/gbc.20021>.
- 541 (63) Zheng, W.; Obrist, D.; Weis, D.; Bergquist, B. A. Mercury Isotope Compositions across North
542 American Forests: Mercury Isotopes Across U.S. Forests. *Glob. Biogeochem. Cycles* **2016**, *30* (10),
543 1475–1492. <https://doi.org/10.1002/2015GB005323>.
- 544 (64) Hersbach, H.; Bell, B.; Berrisford, P.; Hirahara, S.; Horányi, A.; Muñoz-Sabater, J.; Nicolas, J.;
545 Peubey, C.; Radu, R.; Schepers, D.; Simmons, A.; Soci, C.; Abdalla, S.; Abellan, X.; Balsamo, G.;
546 Bechtold, P.; Biavati, G.; Bidlot, J.; Bonavita, M.; De Chiara, G.; Dahlgren, P.; Dee, D.; Diamantakis,
547 M.; Dragani, R.; Flemming, J.; Forbes, R.; Fuentes, M.; Geer, A.; Haimberger, L.; Healy, S.; Hogan,

- 548 R. J.; Hólm, E.; Janisková, M.; Keeley, S.; Laloyaux, P.; Lopez, P.; Lupu, C.; Radnoti, G.; de Rosnay,
549 P.; Rozum, I.; Vamborg, F.; Villaume, S.; Thépaut, J.-N. The ERA5 Global Reanalysis. *Q. J. R.*
550 *Meteorol. Soc.* **2020**, *146* (730), 1999–2049. <https://doi.org/10.1002/qj.3803>.
- 551 (65) Slemr, F.; Seiler, W.; Schuster, G. Latitudinal Distribution of Mercury over the Atlantic Ocean. *J.*
552 *Geophys. Res.* **1981**, *86* (C2), 1159. <https://doi.org/10.1029/JC086iC02p01159>.
- 553 (66) Bieser, J.; Slemr, F.; Ambrose, J.; Brenninkmeijer, C.; Brooks, S.; Dastoor, A.; DeSimone, F.;
554 Ebinghaus, R.; Gencarelli, C. N.; Geyer, B.; Gratz, L. E.; Hedgecock, I. M.; Jaffe, D.; Kelley, P.; Lin, C.-
555 J.; Jaegle, L.; Matthias, V.; Ryjkov, A.; Selin, N. E.; Song, S.; Travnikov, O.; Weigelt, A.; Luke, W.;
556 Ren, X.; Zahn, A.; Yang, X.; Zhu, Y.; Pirrone, N. Multi-Model Study of Mercury Dispersion in the
557 Atmosphere: Vertical and Interhemispheric Distribution of Mercury Species. *Atmospheric Chem.*
558 *Phys.* **2017**, *17* (11), 6925–6955. <https://doi.org/10.5194/acp-17-6925-2017>.
- 559 (67) Sprovieri, F.; Pirrone, N.; Bencardino, M.; D'Amore, F.; Carbone, F.; Cinnirella, S.;
560 Mannarino, V.; Landis, M.; Ebinghaus, R.; Weigelt, A.; Brunke, E.-G.; Labuschagne, C.; Martin, L.;
561 Munthe, J.; Wängberg, I.; Artaxo, P.; Morais, F.; Barbosa, H. de M. J.; Brito, J.; Cairns, W.;
562 Barbante, C.; Diéguez, M. del C.; Garcia, P. E.; Dommergue, A.; Angot, H.; Magand, O.; Skov, H.;
563 Horvat, M.; Kotnik, J.; Read, K. A.; Neves, L. M.; Gawlik, B. M.; Sena, F.; Mashyanov, N.; Obolkin,
564 V.; Wip, D.; Feng, X. B.; Zhang, H.; Fu, X.; Ramachandran, R.; Cossa, D.; Knoery, J.; Maruszczak, N.;
565 Nerentorp, M.; Norstrom, C. Atmospheric Mercury Concentrations Observed at Ground-Based
566 Monitoring Sites Globally Distributed in the Framework of the GMOS Network. *Atmospheric*
567 *Chem. Phys.* **2016**, *16* (18), 11915–11935. <https://doi.org/10.5194/acp-16-11915-2016>.
- 568 (68) Zhang, Y.; Jacob, D. J.; Horowitz, H. M.; Chen, L.; Amos, H. M.; Krabbenhoft, D. P.; Slemr, F.; St.
569 Louis, V. L.; Sunderland, E. M. Observed Decrease in Atmospheric Mercury Explained by Global
570 Decline in Anthropogenic Emissions. *Proc. Natl. Acad. Sci.* **2016**, *113* (3), 526–531.
571 <https://doi.org/10.1073/pnas.1516312113>.
- 572 (69) Strode, S. A.; Jaeglé, L.; Selin, N. E.; Jacob, D. J.; Park, R. J.; Yantosca, R. M.; Mason, R. P.; Slemr, F.
573 Air-Sea Exchange in the Global Mercury Cycle: MERCURY AIR-SEA EXCHANGE. *Glob. Biogeochem.*
574 *Cycles* **2007**, *21* (1). <https://doi.org/10.1029/2006GB002766>.
- 575 (70) Bieser, J.; Angot, H.; Slemr, F.; Martin, L. *Atmospheric Mercury in the Southern Hemisphere*
576 *– Part 2: Source Apportionment Analysis at Cape Point Station, South Africa*; preprint;
577 Gases/Field Measurements/Troposphere/Chemistry (chemical composition and reactions), 2020.
578 <https://doi.org/10.5194/acp-2020-63>.

581 **Figures Captions**

582 Figure 1. Profiles of Hg accumulation rates (HgAR) in the peat cores from Amsterdam Island (AMS), Falkland
583 Islands (SCB, Islas Malvinas), Andorra and Harberton (AND, HBT, Tierra del Fuego). Vertical dashed lines
584 operationally separate the natural background (pre-1450AD), pre-industrial (1450-1880AD), the extended
585 20th century maximum HgAR (20Cmax, grey bars) and modern (post-1990AD) reference periods, following
586 reference ¹⁵).

587 Figure 2. Review of published Hg accumulation rates (HgAR) and enrichment factors (EF) in NH and SH peat
588 and sediment cores for different reference time periods. HgAR ($\mu\text{g m}^{-2} \text{yr}^{-1}$) and EF in peat (A), (C) and
589 sediment (B), (D) profiles during different periods: Natural background (pre-1450AD), pre-industrial (1450-
590 1880AD), extended 20th century maximum (20Cmax, defined as the broad 20th century HgAR peak, and
591 modern period (post-1990AD). $\text{EF}_{\rho/b}$: EF from natural background to pre-industrial period. $\text{EF}_{\text{preind}}$: EF from
592 pre-industrial to 20Cmax. $\text{EF}_{\text{alltime}}$: EF from natural background to 20Cmax.

593 Figure 3. Hg enrichment factors between different reference time periods and peat background Hg
594 accumulation rate. Enrichment factors (EF) in Hg accumulation rates for A) 20th century industrial relative
595 to pre-industrial periods ($\text{EF}_{\text{pre-ind}}$, 1450-1880AD). B) 20th century industrial relative to natural background
596 periods ($\text{EF}_{\text{alltime}}$, pre-1450AD century). Circles represent peat cores, and crosses sediment cores. C) Natural
597 background Hg accumulation rate (pre-1450AD HgAR) in peat cores as a function of latitude. For details
598 see Extended Data 2.

599 **Table 1. Hg accumulation rate (HgAR) enrichment factor observed in the peat profiles from this study.**
 600 AMS, Amsterdam Island; SCB, the Falkland Islands; AND, HBT, Andorra and Harberton, Argentina. 'Pre-ind',
 601 pre-industrial; '20Cmax', extended 20th century maximum HgAR (see Methods); 'p/b', pre-
 602 industrial/background.

	Pre-ind/ background (EF _{p/b})	20Cmax/Pre-ind (EF _{preind})	20Cmax/background (EF _{alltime})
AMS	1.6	1.7	2.7
SCB	0.6	2.5	1.5
AND		3.0	
HBT	1.4	5.3	7.3

603
 604 **Table 2. Summary of Hg accumulation rate (HgAR) enrichment factors (EF) in global peat and sediment**
 605 **records.** 'Pre-ind', pre-industrial; '20Cmax', extended 20th century maximum HgAR (see Methods); 'p/b',
 606 pre-industrial/background; 'modern/back', 'modern/background'; NH, northern hemisphere; SH,
 607 southern hemisphere.

	Pre-ind /background (EF _{p/b})		20Cmax/pre-ind (EF _{preind})		20Cmax/background (EF _{alltime})		Modern/ background (EF _{mod/bck})	
Global-sediment	1.6	n=13	2.9	n=103	4.3	n=14	5.0	n=10
Global-peat	2.5	n=17	4.3	n=30	14.5	n=25	10.3	n=18
NH-sediment+peat	3.9	n=18	3.3	n=110	16.1	n=26	10.5	n=17
SH-sediment+peat	1.3	n=11	1.9	n=21	4.0	n=13	3.5	n=11
NH-sediment	3.7	n=5	3.1	n=84	12.8	n=5	19.3	n=4
NH-peat	3.9	n=14	4.6	n=25	16.2	n=21	12.3	n=14
SH-sediment	1.4	n=8	1.8	n=17	3.8	n=97	5.0	n=8
SH-peat	1.2	n=3	3.1	n=4	6.0	n=4	3.1	n=4

608 ¹the number of records, n, do not always add up due to the 2 σ outlier tests applied, for ex. SH sediment,
 609 n=8, SH peat, n=3, but SH sediment+peat, n=10. See Methods and Extended Data 2 for details on outlier
 610 tests.

611 **Table 3. Summary of natural and anthropogenic Hg emissions to the atmosphere (mean \pm 1 σ)**

	NH	1 σ	SH	1 σ
passive volcanic degassing (this study) Mg y ⁻¹	92	20	87	19
eruptive volcanic degassing (this study) Mg y ⁻¹	10	10	10	10
crustal degassing ^{57,58} Mg y ⁻¹	91	27	44	13
anthropogenic 20Cmax emissions ⁷ Mg y ⁻¹	2000	500	480	20
Mean EF _{emission}	11.2	4.6	4.4	1.5
Median EF _{alltime}	16.1	10-30 IQR	4.0	2-6 IQR

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619 **Graphic files**

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621 Unequal anthropogenic enrichment of mercury in Earth's northern and
622 southern hemispheres.

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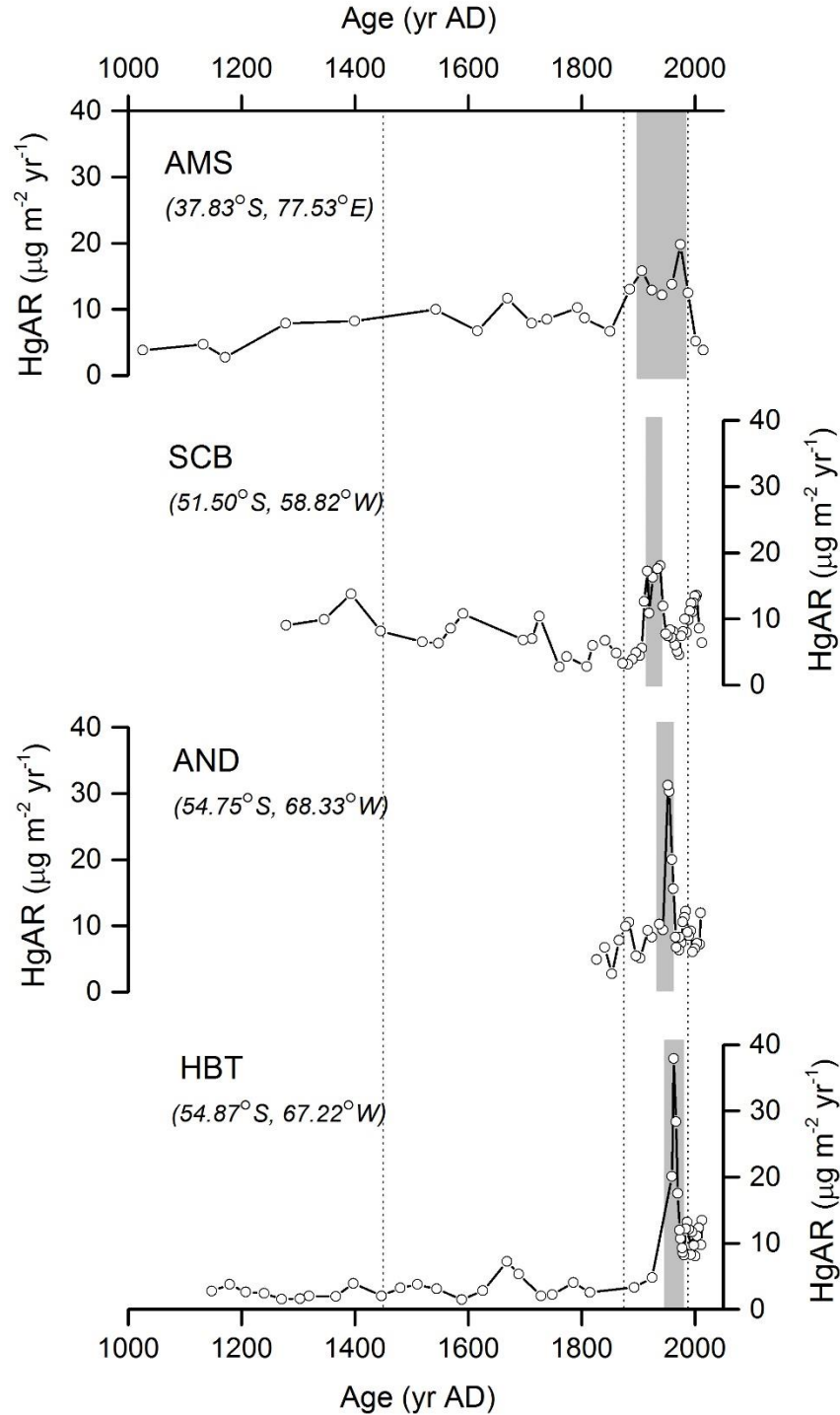
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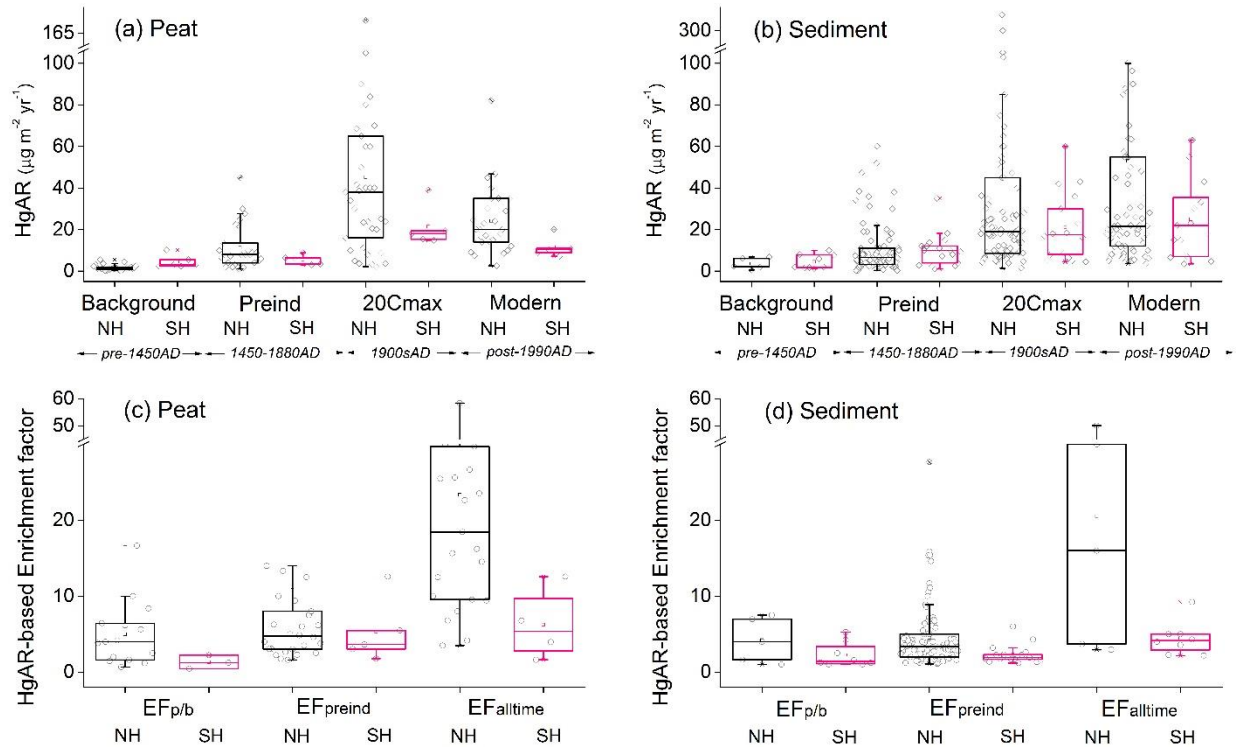
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642
 643 **Figure 1. Profiles of Hg accumulation rates (HgAR) in the peat cores from Amsterdam Island (AMS),**
 644 **Falkland Islands (SCB, Islas Malvinas), Andorra and Harberton (AND, HBT, Tierra del Fuego).** Vertical
 645 dashed lines operationally separate the natural background (pre-1450AD), pre-industrial (1450-1880AD),
 646 the extended 20th century maximum HgAR (20Cmax, grey bars) and modern (post-1990AD) reference
 647 periods, following reference ¹⁵).

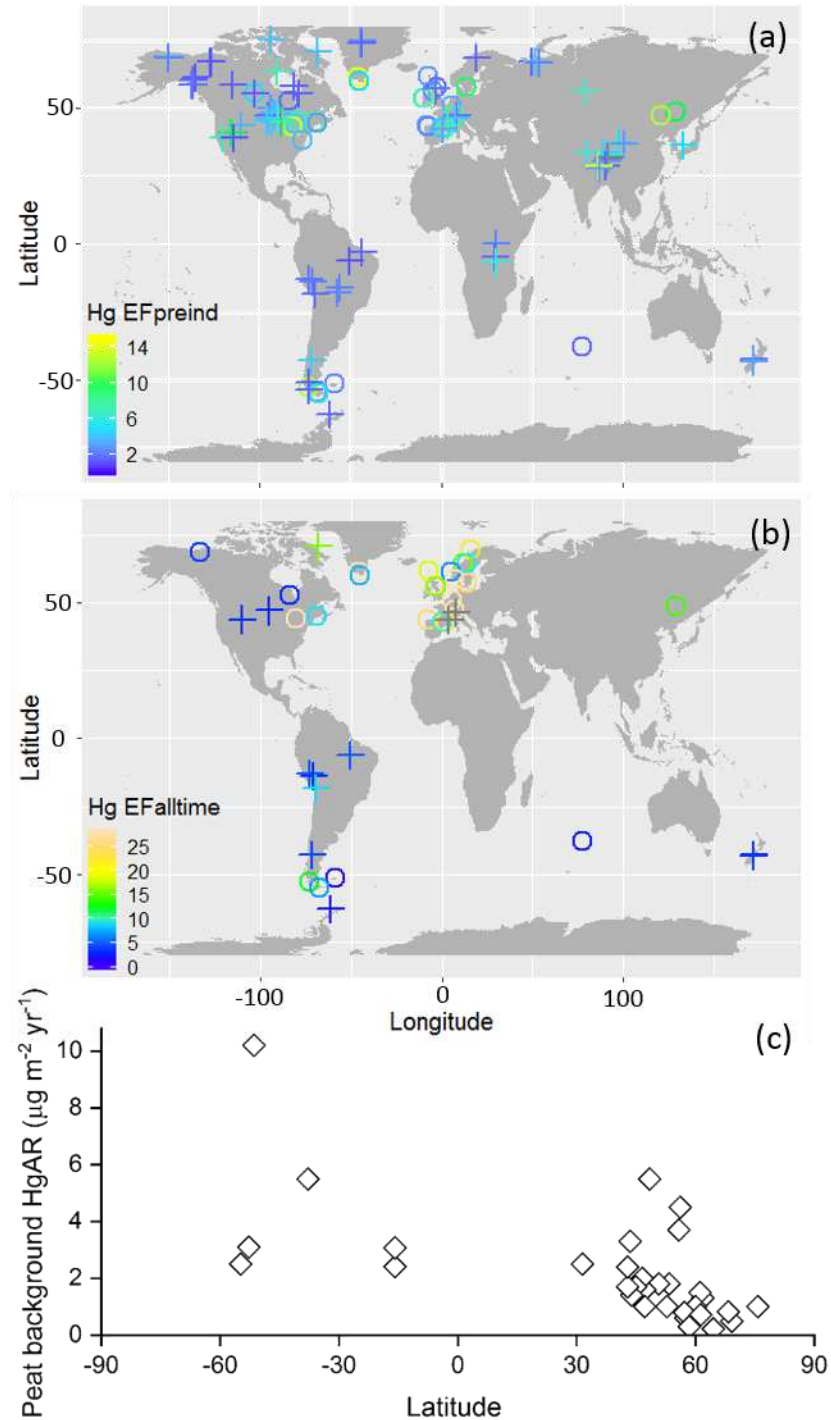


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650 **Figure 2. Review of published Hg accumulation rates (HgAR) and enrichment factors (EF) in NH and SH**
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 654 **and modern period (post-1990AD). $\text{EF}_{\text{p/b}}$: EF from natural background to pre-industrial period. $\text{EF}_{\text{preind}}$: EF**
 655 **from pre-industrial to 20Cmax. $\text{EF}_{\text{alltime}}$: EF from natural background to 20Cmax.**

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657
 658 **Figure 3. Hg enrichment factors between different reference time periods and peat background Hg**
 659 **accumulation rate.** Enrichment factors (EF) in Hg accumulation rates for a) 20th century industrial relative
 660 to pre-industrial periods ($EF_{\text{pre-ind}}$, 1450-1880AD). b) 20th century industrial relative to natural background
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668
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 671 pre-industrial/background; 'modern/back', 'modern/background'; NH, northern hemisphere; SH,
 672 southern hemisphere.

	Pre-ind /background (EF _{p/b})		20Cmax/pre-ind (EF _{Preind})		20Cmax/background (EF _{alltime})		Modern/ background (EF _{modern/back})	
Global-sediment	1.6	n=13	2.9	n=101	4.3	n=11	5.0	n=11
Global-peat	2.5	n=17	4.3	n=30	14.5	n=25	10.3	n=18
NH-sediment+peat	4.0	n=20	3.3	n=111	16.2	n=27	12.2	n=20
SH-sediment+peat	1.3	n=10	1.9	n=19	4.3	n=11	3.5	n=9
NH-sediment	4.2	n=5	3.2	n=86	19.0	n=5	22.5	n=5
NH-peat	3.9	n=14	4.6	n=25	16.2	n=21	12.3	n=14
SH-sediment	1.4	n=8	1.8	n=15	3.8	n=7	5.0	n=6
SH-peat	1.2	n=3	3.1	n=4	6.0	n=4	3.1	n=4

673 ¹the number of records, n, do not always add up due to the 2 σ outlier tests applied, for ex. SH sediment,
 674 n=8, SH peat, n=3, but SH sediment+peat, n=10. See Methods and Extended Data 2 for details on outlier
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anthropogenic 20Cmax emissions (7) Mg y ⁻¹	2000	500	480	20
Mean EF _{emission}	11.2	4.6	4.4	1.5
Median EF _{alltime}	16.1	10-30 IQR	4.0	2-6 IQR

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681 **Supporting Information**

682

683 Unequal anthropogenic enrichment of mercury in Earth's northern and
684 southern hemispheres.

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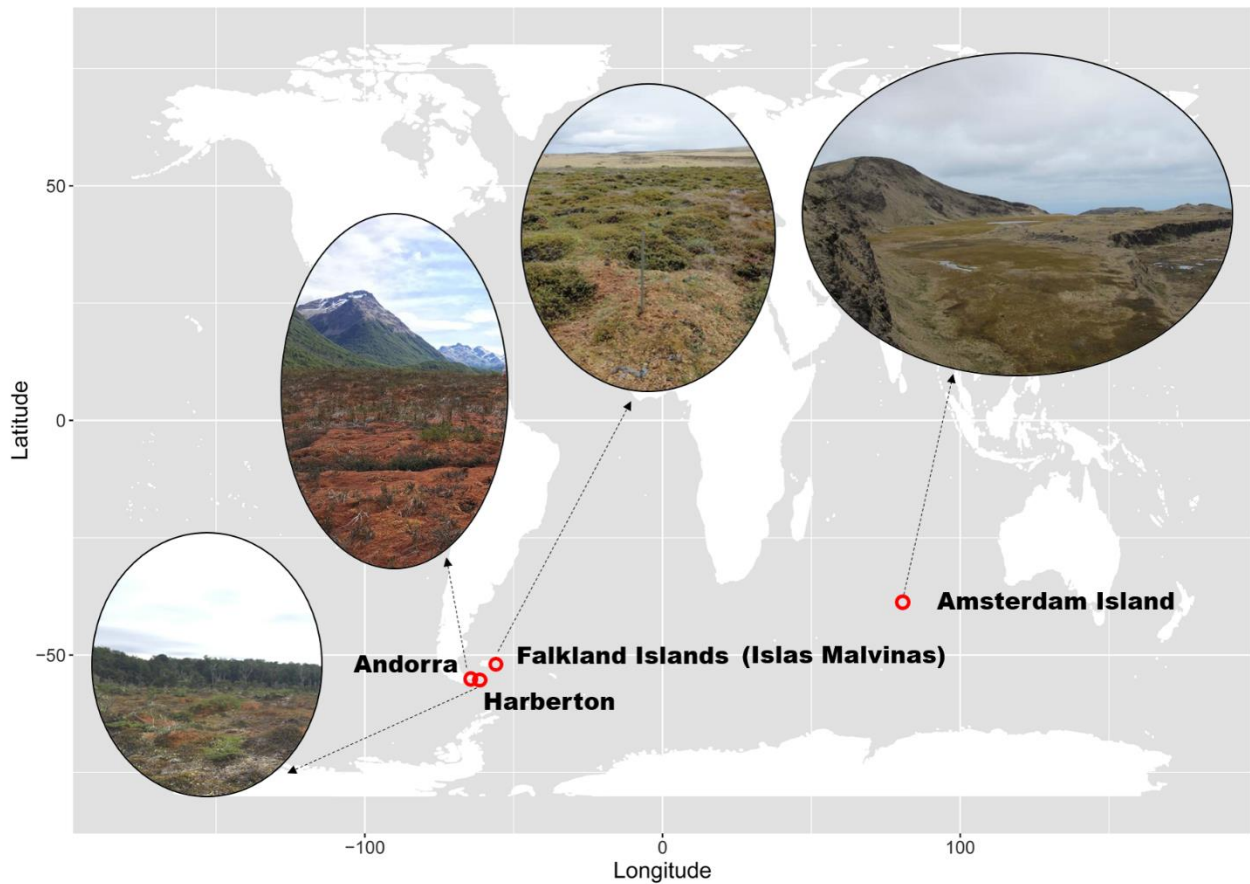
703 This SI contains Table S1, S2, Text S1, Figures S1, S2, S3, S4, S5, S6.

704

705 **Table S1. Details of the coring sites in this investigation**

Location	Site name	coordinates	Elevation (m a.s.l)	Precipitation (mm yr ⁻¹)	Coring date	Label core	core length (m)
Amsterdam Island	Central plateau	37.83°S, 77.53°E	738	1124	11/2014	AMS14-PB01	5
Falkland Islands (Islas Malvinas)	San Carlos bog	51.50°S, 58.82°W	8	575	2013	SCB13-PB01C	1.7
Valle de Andorra	Andorra	54.75°S, 68.22°W	198	450-600	02/2012	AND12-PB01W1	0.77
Estancia Harberton	Harberton	54.87°S, 67.22°W	26	600	02/2012	HBT12-PB01W1	0.92

706



708 **Figure S1. Location of Amsterdam Island (AMS), Falkland Islands (SCB, Islas Malvinas), Andorra (AND)**
 709 **and Harberton (HBT).**

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712

713 **Text S1 Core sites:**

714 **Amsterdam Island (AMS):** A 5 m-long peat sequence (AMS14-PB01A) was collected from the most
715 elevated area of the peatland at 738 m a.s.l. in December 2014 using a stainless steel Russian D-corer of
716 10 cm internal diameter and 50 cm length. The mean annual temperature at the meteorological station
717 (27 m a.s.l.) is 14°C and annual precipitation is about 1100 mm yr⁻¹ (ref¹). For details about AMS coring site
718 see ref². The vegetation at the coring site is characterized by bryophytes (brown mosses together with
719 liverworts and some *Sphagnum* species), *Blechnum penna-marina*, *Scirpus aucklandicus*, *Trisetum insularis*
720 and scattered stands of *Agrostis delislei*. Based on low resolution plant macrofossil data for the last 1000
721 years of a peat core taken close to the AMS14-PB01A core, with an independent age-depth model, the
722 macrofossil record is dominated by higher plant epidermis (c. 70%) until about 400 cal yr BP. For the last
723 400 years, bryophytes are dominant (70-80%), mainly composed of brown mosses and liverworts, with
724 little occurrence of *Sphagnum* spp. Ash content is <2wt% throughout the core and, together with major
725 element profiles, suggests the site to be ombrotrophic to at least 3.5m depth.

726
727 **The Falkland Islands (SCB, Islas Malvinas):** 'San Carlos bog' is located on the western side of East Falkland
728 Islands (SCB13-PB01C). The native vegetation is treeless and dominated by mosses, grasses and dwarf
729 shrubs^{3,4}. A 1.7 m-long peat sequence was collected from a hummock with an upper monolith section (0
730 - 50 cm) and lower Russian core section⁵. The surface vegetation of the bog is dominated by *Sphagnum*
731 *magellanicum*, *Hymenophyllum caespitosum*, *Gaultheria pumila*, *Oreobulis obtusangulus*, *Gunnera*
732 *magellanica* and *Myrteola nummularia*. *Sphagnum* is found to be more than 80% to a depth of 65 cm and
733 followed by herbaceous compacted peat to the bottom. The annual precipitation and temperature are
734 575 mm yr⁻¹ and 7°C, respectively (data sources from the Falkland Islands Government reported in ref⁴).

735 **Andorra (AND):** An ombrotrophic peat monolith (0.72 m length, AND12-PB01W1) was collected at
736 Andorra bog using a stainless steel Wardenaar corer⁶. The AND peat profile is dominated >96% by
737 *Sphagnum magellanicum*. The annual precipitation and temperature are 450-600 mm yr⁻¹ and 6°C,
738 respectively⁷.

739 **Harberton (HBT):** An ombrotrophic peat monolith (0.73 m length, HBT12-PB01W1) was sampled at
740 Harberton Bog by a stainless steel Wardenaar corer⁶. The bog surface is dominated >80% by *Sphagnum*
741 *magellanicum* with a sparse cover of *Marsippospermum grandiflorum* and *Empetrum rubrum*⁸. The annual
742 precipitation and temperature are around 600 mm yr⁻¹ and 6°C, respectively⁸. We are aware of limited
743 gold mining from 1883 to 1906 on Chilean Islands South of the Beagle Channel, but this is hundreds of kms
744 away from our sites, and late 20th century peaks in HgAR at HBT do not correspond in terms of timing.

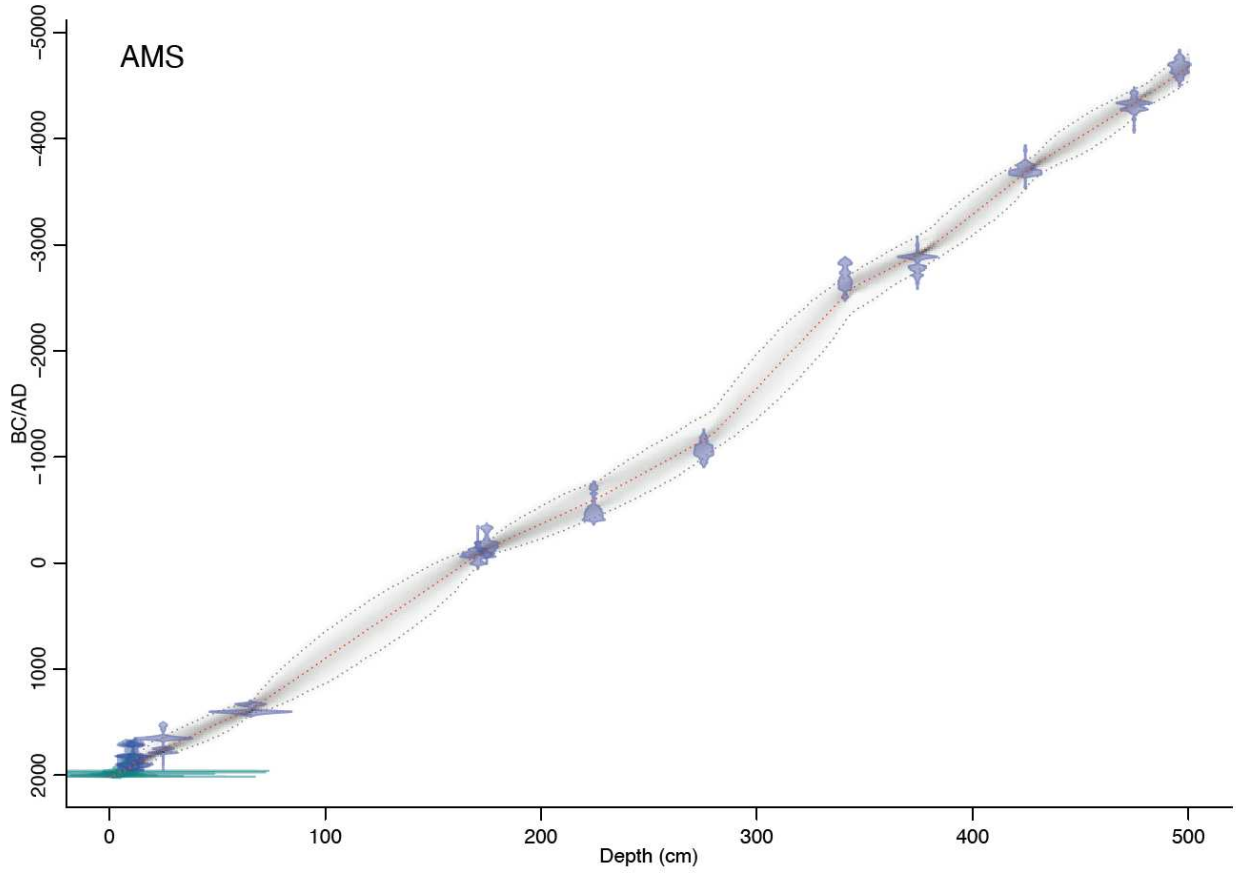
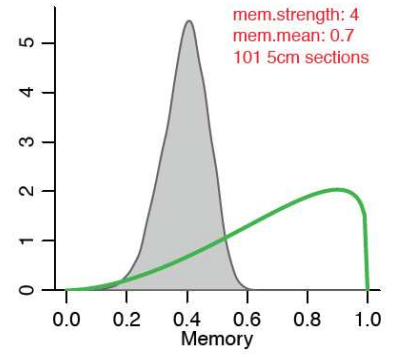
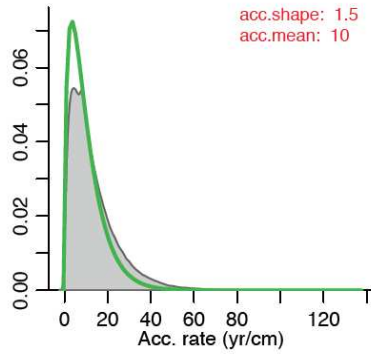
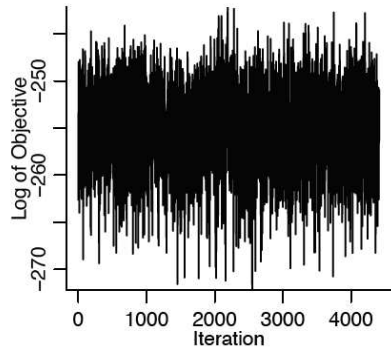
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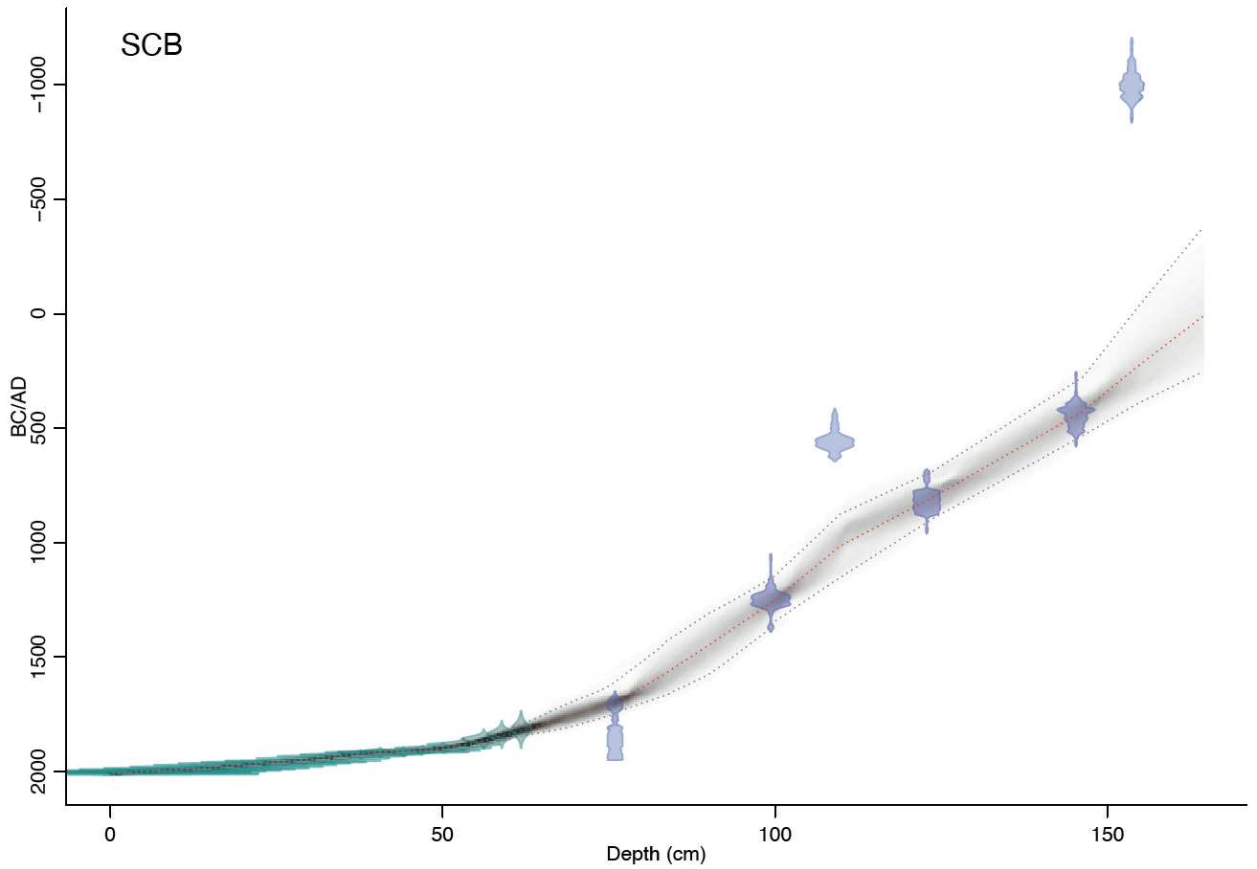
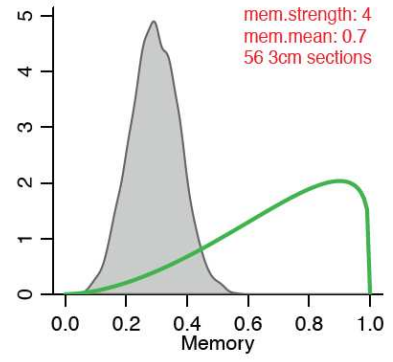
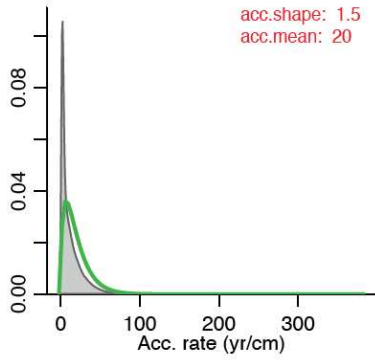
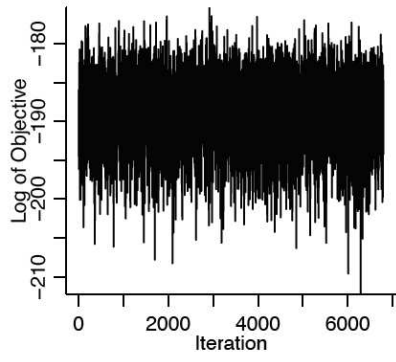
Table S2 Accelerator Mass Spectrometry ¹⁴C dating of plant macrofossils from all the four peat cores.

Core name	Lab ID	Mid-Point Depth (cm)	material	Conventional ¹⁴ C Age (yr BP, ± 1σ)	Calibrated age (median, AD/BC)	Modelled age AD/BC (95.4% probability range)
AMS*	SacA50049	2.0	<i>Chorisondontium/Dicranoloma</i> stems + leaves	-557 ± 21	2008 AD	1997-2001 AD
AMS*	SacA50050	3.5	Brown moss stems	-1489 ± 20	1987 AD	1985-1987 AD
AMS*	SacA50051	4.9	Brown moss + liverworts stems	-3052 ± 18	1974 AD	1973-1979 AD
AMS*	SacA50052	6.4	Brown moss + liverworts stems	-1248 ± 20	1960 AD	1956-1962 AD
AMS*	SacA50053	7.8	Brown moss stems	135 ± 30	1942 AD	1938-1950 AD
AMS*	SacA50054	9.4	Brown moss stems	115 ± 30	1928 AD	1917-1937 AD
AMS*	SacA50055	10.8	Brown moss stems + leaves	80 ± 30	1912 AD	1895-1923 AD
AMS*	SacA50056	12.0	Brown moss stems + <i>Chorisondontium/Dicranoloma</i> leaves	160 ± 30	1893 AD	1854-1917 AD
AMS*	SacA50057	13.2	brown moss stems	70 ± 30	1885 AD	1823-1915 AD
AMS*	GdA-4136	24.9	brown moss stems	275 ± 25	1752 AD	1640-1800 AD
AMS*	GdA-4558	65.4	Residue (<i>Sphagnum</i> dominated)	595 ± 25	1389 AD	1310-1440 AD
AMS*	GdA-4560	170.7	Brown moss stems	2100 ± 25	78 BC	155 BC-30 AD
AMS*	GdA-4137	174.8	brown moss stems	2170 ± 30	126 BC	195-55 BC
AMS*	GdA-4138	224.4	brown moss stems	2430 ± 30	580 BC	750-415 BC
AMS*	GdA-4139	275.4	brown moss stems	2925 ± 30	1142 BC	1380-980 BC
AMS*	GdA-4561	340.9	brown moss stems	4145 ± 35	2535 BC	2965-2275 BC
AMS*	GdA-4140	374.4	<i>Sphagnum</i>	4285 ± 30	2900 BC	3075-2750 BC
AMS*	GdA-4141	424.4	<i>Sphagnum</i> + brown moss	4960 ± 30	3680 BC	3795-3550 BC
AMS*	GdA-4142	474.8	<i>Sphagnum</i> stems	5515 ± 35	4330 BC	4460-4190 BC
AMS*	GdA-4143	495.9	<i>Sphagnum</i> stems	5860 ± 35	4615 BC	4750-4470 BC
SCB	SUERC-51676	76.5	<i>Sphagnum</i>	153 ± 37	1694 AD	1597-1737 AD
SCB	GdA-3755	99.9	Undefined peat macrofossils	814 ± 41	1256 AD	1147-1345 AD
SCB	GdA-4744	109.8	Charcoal + Monocotyledons undifferentiated (leaf bases)	1553 ± 25	1009 AD	876-1152 AD
SCB	GdA-4745	123.7	Monocotyledons undifferentiated (leaf bases)	1261 ± 21	804 AD	688-896 AD
SCB	GdA-4746	146.3	Monocotyledons undifferentiated (leaf bases)	1661 ± 25	428 AD	277-532 AD
SCB	GdA-4742	154.3	Charcoal + Monocotyledons undifferentiated (leaf bases)	2882 ± 22	252 AD	19 BC-396 AD
SCB	GdA-3756	164.3	Undefined peat macrofossils	11582 ± 50	36 AD	376 BC-254 AD
AND	SacA50058	0.6	<i>Sphagnum</i>	-594 ± 19	2004 AD	2007-2014 AD
AND	SacA50059	13.1	<i>Sphagnum</i>	-1749 ± 19	1983 AD	1985-2000 AD
AND	SacA50060	34.3	<i>Sphagnum</i>	-2839 ± 17	1974 AD	1969-1976 AD
AND	SacA50061	41.0	<i>Sphagnum</i>	-2695 ± 18	1964 AD	1961-1967 AD
AND	SacA50062	47.6	<i>Sphagnum</i>	-67 ± 21	1954 AD	1947-1958 AD
AND	SacA50063	54.6	<i>Sphagnum</i>	120 ± 30	1926 AD	1902-1942 AD
AND	SacA50064	61.9	<i>Sphagnum</i>	140 ± 30	1893 AD	1856-1919 AD
AND	SacA50065	68.8	<i>Sphagnum</i>	160 ± 30	1863 AD	1814-1893 AD
AND	GdA-3032	73.2	<i>Sphagnum</i>	193 ± 23	1843 AD	1787-1876 AD
AND	SacA50066	76.1	<i>Sphagnum</i>	150 ± 30	1831 AD	1769-1865 AD
HBT	SacA42507	0.3	<i>Sphagnum</i>	-424 ± 21	2010 AD	2010-2019 AD
HBT	SacA42508	4.7	<i>Sphagnum</i>	-606 ± 22	2004 AD	2002-2012 AD
HBT	SacA42509	6.9	<i>Sphagnum</i>	-677 ± 21	2002 AD	1999-2008 AD
HBT	SacA42510	9.1	<i>Sphagnum</i>	-788 ± 21	1999 AD	1996-2005 AD

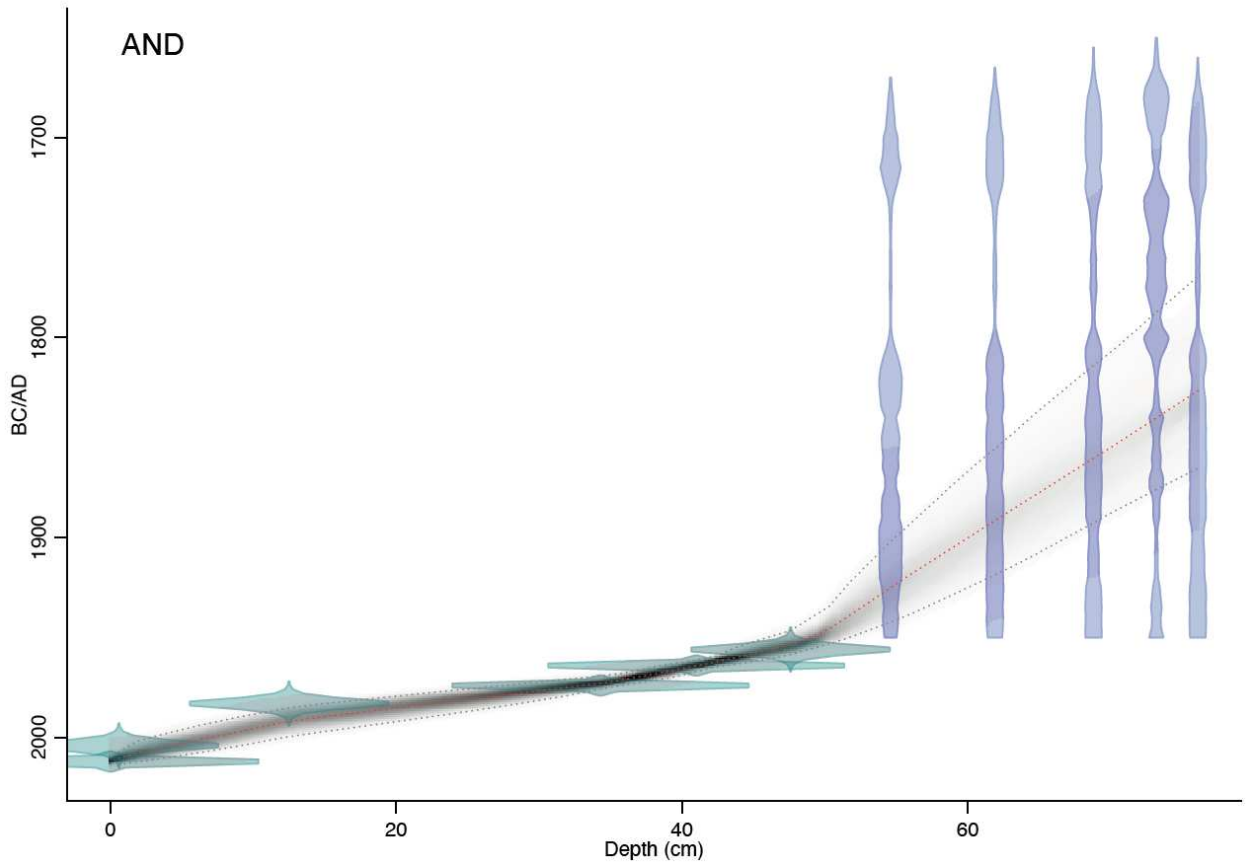
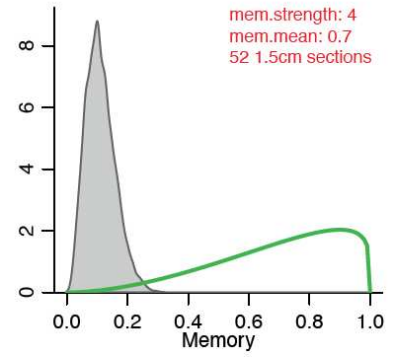
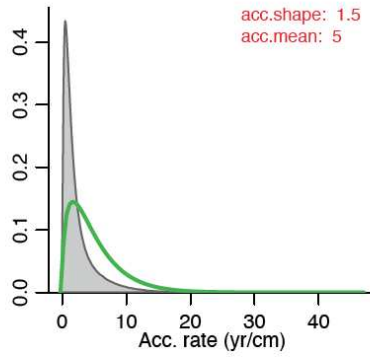
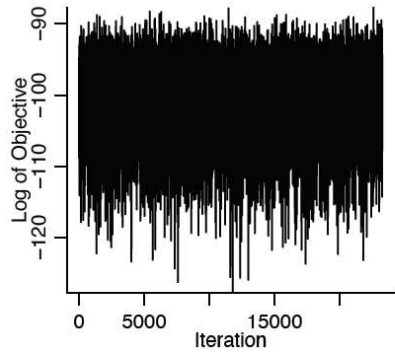
HBT	SacA42511	11.3	<i>Sphagnum</i>	-838 ± 22	1998 AD	1994-2002 AD
HBT	SacA42512	13.6	<i>Sphagnum</i>	-914 ± 21	1996 AD	1991-1999 AD
HBT	SacA44490	15.8	<i>Sphagnum</i>	-1092 ± 22	1992 AD	1988-1996 AD
HBT	SacA44491	19.2	<i>Sphagnum</i>	-1333 ± 21	1988 AD	1984-1992 AD
HBT	SacA44492	21.3	<i>Sphagnum</i>	-1513 ± 20	1985 AD	1981-1989 AD
HBT	SacA44493	26.6	<i>Sphagnum</i>	-2186 ± 21	1979 AD	1974-1982 AD
HBT	SacA44494	31.7	<i>Sphagnum</i>	-2715 ± 20	1975 AD	1964-1976 AD
HBT	SacA44495	37.0	<i>Sphagnum</i>	-2462 ± 27	1964 AD	1930-1964 AD
HBT	SacA44496	43.5	<i>Sphagnum</i>	214 ± 23	1815 AD	1736-1885 AD
HBT	SacA44497	56.2	<i>Sphagnum</i>	407 ± 25	1608 AD	1518-1631 AD
HBT	SacA44498	90.7	<i>Sphagnum</i>	984 ± 24	1148 AD	1063-1216 AD

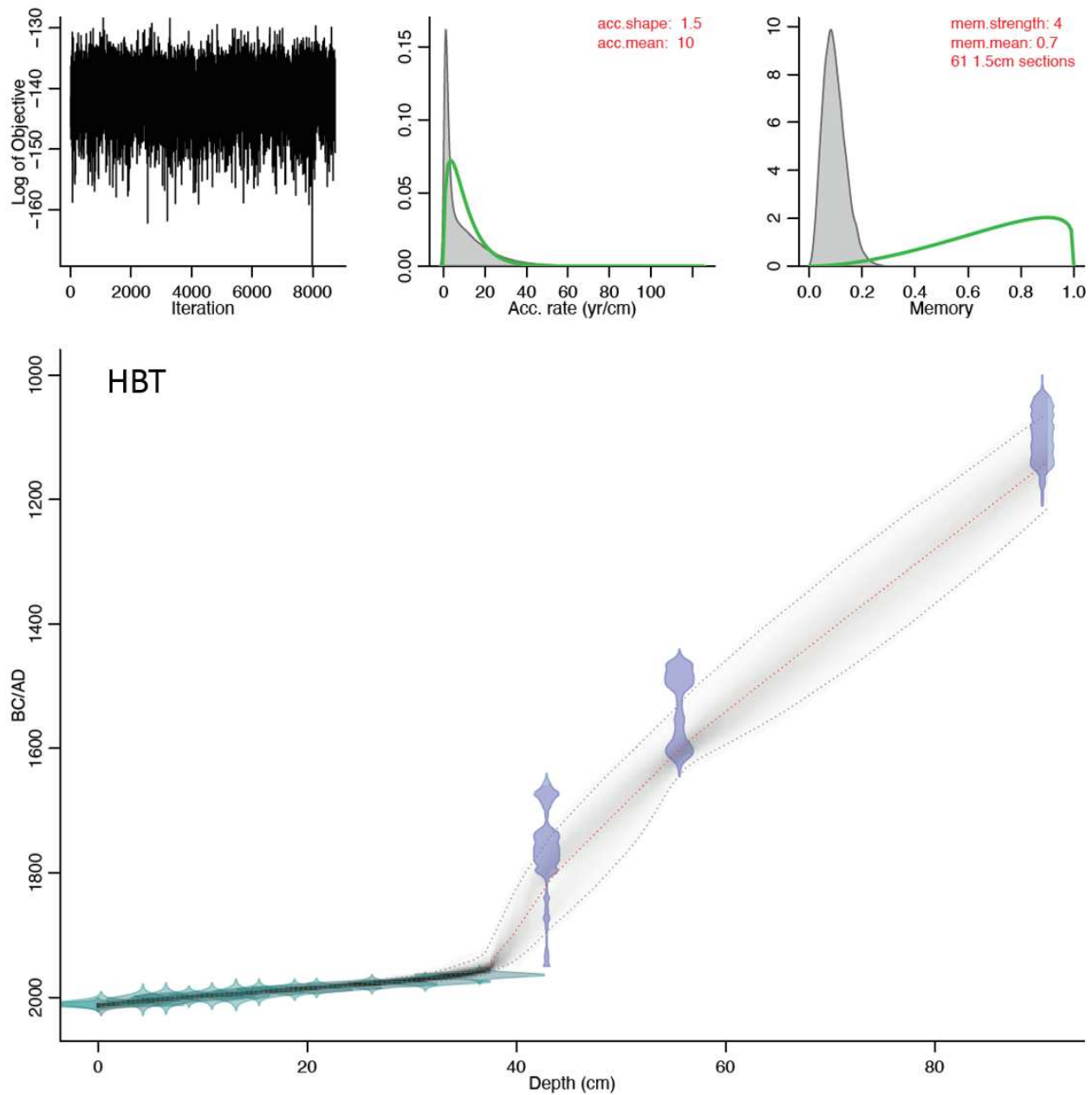
*Data are from ref ²⁸.





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755 **Figure S2. Age models of peat cores from AMS, SCB, AND and HBT using Bacon.** Calibrated ^{14}C dates show
 756 in transparent blue and ^{210}Pb dates show in transparent green. Red curve indicates single best-fit model
 757 based on the weighted mean age for each depth. Darker greys represent more likely calendar ages with
 758 95% confidence intervals shown by grey stippled lines. Diagnostic plots in upper left panels confirm
 759 appropriate performance of the models. Settings for accumulation rate and memory are shown in middle
 760 and right upper panels (green line—prior, grey shade—posterior distribution), along with thickness and
 761 number of sections used for modelling. Prior settings for accumulation rates described by gamma
 762 distribution with shape 1.5 and acc.mean 10 or 20 yr/cm, for memory the default beta distribution with
 763 parameters mem.strength=4 and mem.mean=0.7 was used.

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765 **Table S3 Summary of Hg measurements in standard reference materials.**

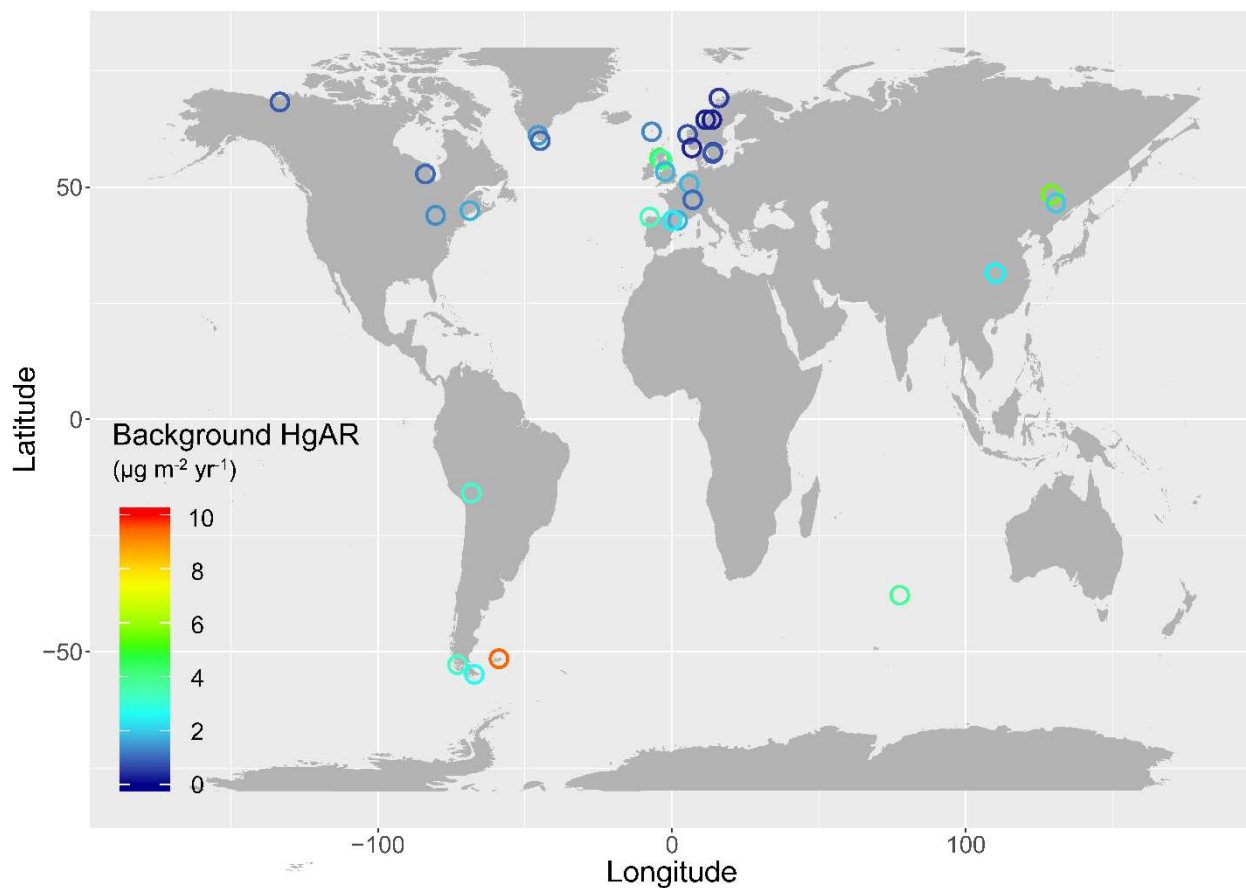
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SRM	materials	Measured value (mean \pm 1 σ , ng g ⁻¹)	Certified value (mean \pm 2 σ , ng g ⁻¹)
IPE 176	Reed/ <i>Phragmites communis</i>	35.1 \pm 6.3 (n=143)	37.9 \pm 2.9
NIST 1632d	Coal	91.3 \pm 7.0 (n=9)	92.8 \pm 3.3
BCR 482	Lichen	481.3 \pm 8.7 (n=5)	480 \pm 20

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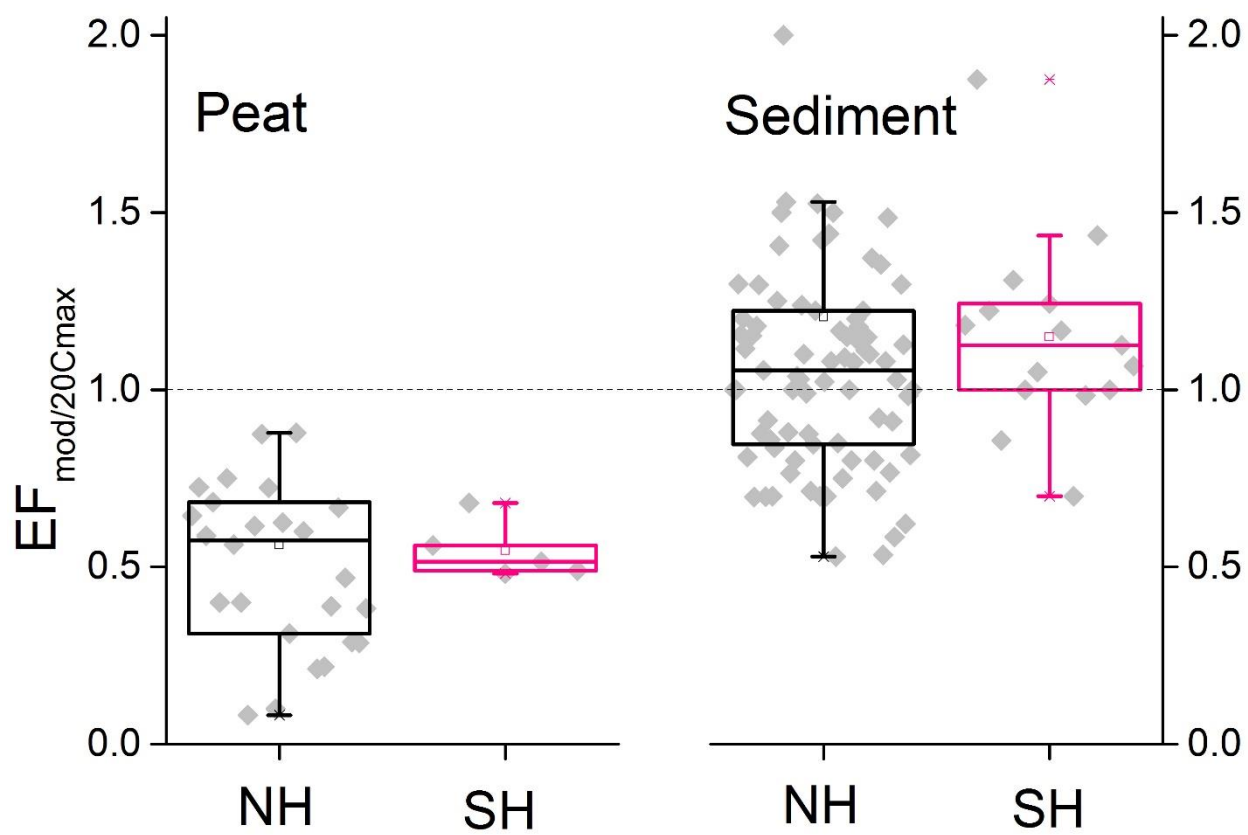
771 **Figure S3. Natural background Hg accumulation rates ($\mu\text{g m}^{-2} \text{yr}^{-1}$) derived from natural peat archives.**

772 **Details see Extended Data 2.**

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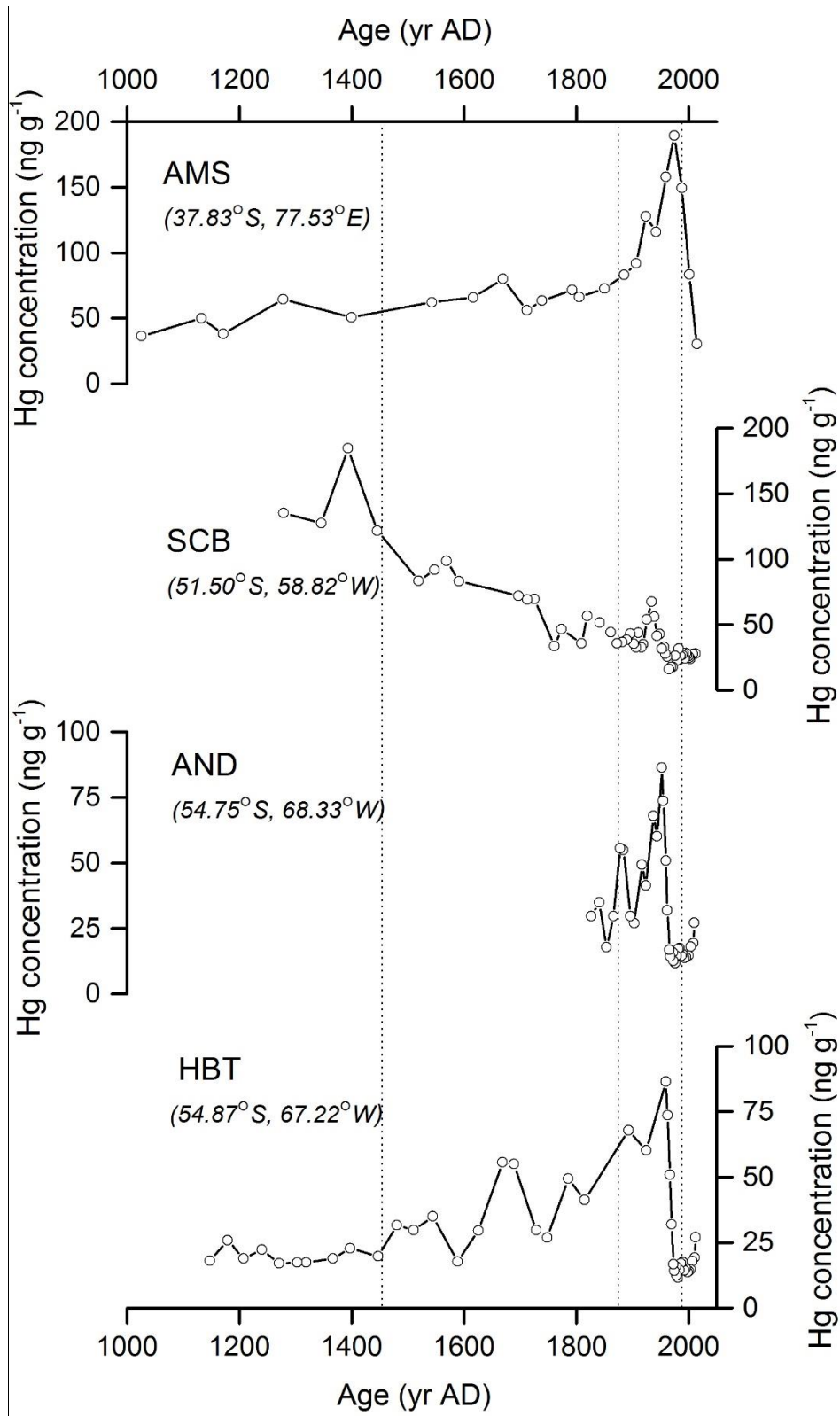
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Figure S4. Profiles of HgAR enrichment factor of modern (post-1990) to extended 20th century maximum ($EF_{\text{mod}/20\text{Cmax}}$) from Northern Hemisphere (NH) and Southern Hemisphere (SH) peat and sediment records. Dashed line indicates $EF=1$.

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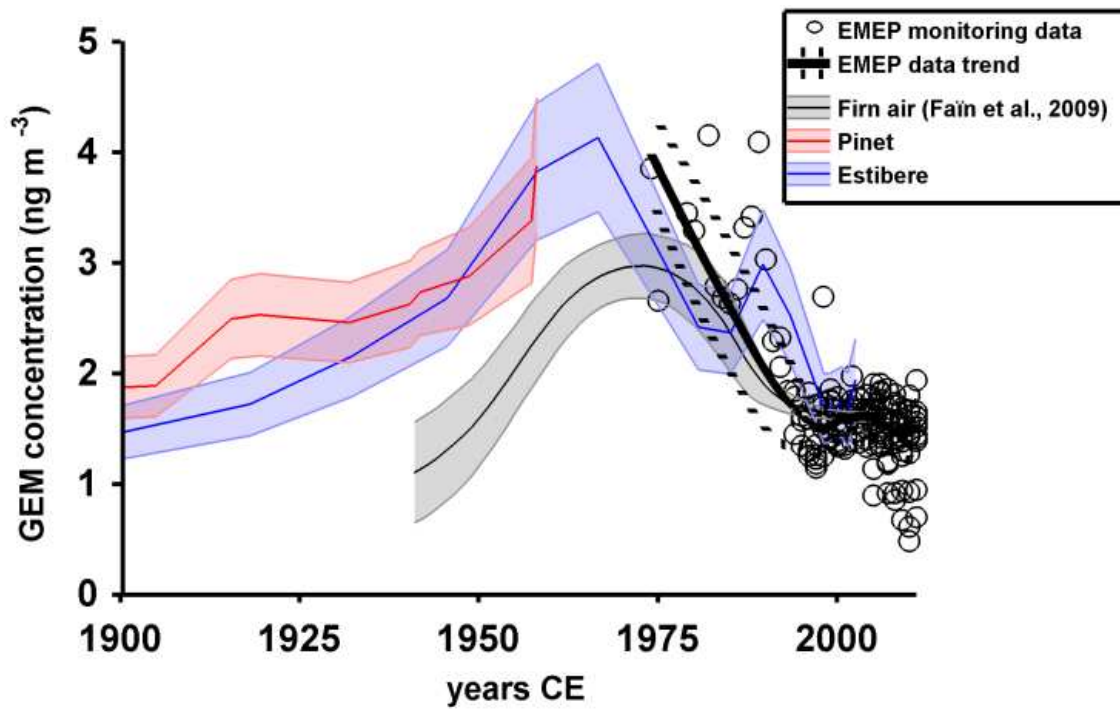


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Figure S5. Profiles of Hg concentration (ng g⁻¹) in the peat cores from AMS, SCB, AND and HBT.

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 787 **Figure S6. Historical atmospheric Hg monitoring observations and reconstructed Hg levels.** Figure
 788 reproduced from Enrico et al. 2017, ES&T with permission⁹. Atmospheric gaseous elemental Hg⁰ (GEM)
 789 monitoring data (circles) are from EMEP¹⁰.
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791 **Supporting Information references**

- 792 1. Lebouvier, M. and Frenot, Y. Conservation and management in the French sub-Antarctic islands and
793 surrounding seas. In *Papers and proceedings of the royal society of Tasmania*. 207, Vol. 141, No. 1, pp. 23-28.
- 794 2. Li, C.; Le Roux, G.; Sonke, J.; van Beek, P.; Souhaut, M.; Van der Putten, N.; De Vleeschouwer, F. Recent
795 ²¹⁰Pb, ¹³⁷Cs and ²⁴¹Am accumulation in an ombrotrophic peatland from Amsterdam Island (Southern Indian
796 Ocean). *Journal of environmental radioactivity*. 2017, 175, pp.164-169.
- 797 3. Moore, D.M., 1968. The vascular flora of the Falkland Islands. *The vascular flora of the Falkland Islands.*, (60).
- 798 4. Bokhorst, S.; Convey, P.; Huiskes, A.; Aerts, R. Dwarf shrub and grass vegetation resistant to long-term
799 experimental warming while microarthropod abundance declines on the Falkland Islands. *Austral Ecology*. 2017,
800 42(8), pp.984-994.
- 801 5. Belokopytov, I.E. and Beresnevich, V.V. Giktorf's peat borers. *Torfyanaya Promyshlennost*. 1955, 8(9), p.10.
- 802 6. Wardenaar, E.C.P. A new hand tool for cutting soil monoliths. *Canadian journal of Soil science*. 1987, 67(2),
803 pp.405-407.
- 804 7. Mauquoy, D.; Blaauw, M.; Van Geel, B.; Borrromei, A.; Quattrocchio, M.; Chambers, F.M.; Possnert, G. Late
805 Holocene climatic changes in Tierra del Fuego based on multiproxy analyses of peat deposits. *Quaternary Research*.
806 2004, 61(2), pp.148-158.
- 807 8. Vanneste, H., De Vleeschouwer, F.; Martínez-Cortizas, A.; Von Scheffer, C.; Piotrowska, N.; Coronato, A.; Le
808 Roux, G. Late-glacial elevated dust deposition linked to westerly wind shifts in southern South America. *Scientific*
809 *reports*. 2015, 5, p.11670.
- 810 9. Enrico M., et al., Holocene Atmospheric Mercury Levels Reconstructed from Peat Bog Mercury Stable
811 Isotopes. *Environ. Sci. Technol*. 2017, 51, 5899–5906.
- 812 10. European Monitoring and Evaluation Programme (2016). <https://www.emep.int/>

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