A revised pan-Arctic permafrost soil Hg pool, based on western Siberia peat Hg and carbon observations

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Key words: mercury, peat soil, landscape, bog, forest, thaw, Siberia

Submitted to *Biogeosciences, after revision, April 29, 2020*

Abstract

Natural and anthropogenic mercury (Hg) emissions are sequestered in terrestrial soils over short, annual, to long, millennial time scales, before Hg mobilization and run-off impacts wetland and coastal ocean ecosystems. Recent studies have used Hg to carbon (C) ratios, R_{HgC}, measured in Alaskan permafrost mineral and peat soils, together with a northern circumpolar permafrost soil carbon inventory, to estimate that these soils contain large amounts, 184 to 755 Gg of Hg in the upper 1 m. However, measurements of R_{HgC} on Siberian permafrost peatlands are largely missing, leaving the size of estimated northern soil Hg budget, and its fate under Arctic warming scenarios uncertain. Here we present Hg and carbon data for 6 peat cores, down to mineral horizons at 1.5 - 4 m depth, across a 1700 km latitudinal (56 to 67°N) permafrost gradient in the western Siberian lowlands (WSL). Mercury concentrations increase from south to north in all soil horizons, reflecting a higher stability of sequestered Hg with respect to re-emission. The R_{HgC} in the WSL peat horizons decreases with depth from 0.38 Gg Pg^{-1} in the active layer to 0.23 Gg Pg^{-1} in continuously frozen peat of the WSL. We estimate the Hg pool (0-1 m) in the permafrost-affected part of the WSL peatlands to be 9.3 ± 2.7 Gg. We review and estimate pan-arctic organic and mineral soil R_{HgC} to be 0.19 and 0.63 Gg Pg^{-1}, and use a soil carbon budget to revise the pan-Arctic permafrost soil Hg pool to be 72 Gg (39-91 Gg, interquartile range (IQR)) in the upper 30 cm, 240 Gg (110-336 Gg) in the upper 1 m, and 597 Gg (384-750 Gg) in the upper 3 m. Using the same R_{HgC} approach, we revise the global upper 30 cm soil Hg pool to contain 1086 Gg of Hg (852-1265 Gg, IQR), of which 6% (72 Gg) resides in northern permafrost soils. Additional soil and river studies in Eastern and Northern Siberia are needed to lower the uncertainty on these estimates, and assess the timing of Hg release to atmosphere and rivers.

1. Introduction

High-latitude organic-rich soils are key ecosystems controlling the transfer of carbon, nutrients, and pollutants between the atmosphere, rivers, lakes and the Arctic Ocean. These soils are most vulnerable to on-going climate change, due to the high potential mobility of carbon stored in the form of peat deposits. Part of the peat layers are currently frozen but may be subjected to fast thawing, especially in discontinuous and sporadic permafrost zones (Romanovsky et al., 2010). Whilst the stock of C in Arctic and subarctic peat and mineral soils is fairly well quantified (472 Pg C (±27 Pg, 95% confidence interval, CI) in the upper 0-1m (Hugelius et al. 2014), this is not true for pollutants such as mercury (Hg). Because of its strong bio-amplification in Arctic marine biota (Morel et al., 1998), and exposure to native Arctic populations (AMAP, 2011), there is a strong interest in understanding Hg biogeochemistry in Arctic environments (Outridge et al., 2008; Steffen et al., 2008; Stern et al., 2012).

Recent advances in quantifying Arctic Hg cycling show that Arctic Hg^{II} wet deposition is generally low (Pearson et al., 2019), and that the vegetation Hg pump drives yearlong net gaseous Hg^0 (and CO_2) deposition, via foliar uptake to Arctic vegetation and litterfall to soils (Obrist et al. 2017; Jiskra et al., 2018, 2019). Soil core analyses in Alaska indicate that large amounts of carbon and Hg have accumulated since the last glacial maximum (Olson et al., 2018; Schuster et al., 2018). Two upscaling approaches reported differing estimates for the 0 to 1 m soil pool of the northern circumpolar permafrost region of 184 Gg (Olson et al., 2018) and 755 Gg (Schuster et al., 2018).

Despite the overall net atmospheric Hg deposition to soils, research has found that Arctic rivers export 37 to 44 Mg y^{-1} of soil Hg, bound to particulate and dissolved organic matter, to the Arctic Ocean (Fisher et al., 2012; Dastoor et al., 2014; Zhang et al., 2015; Sonke et al., 2018; Zolkos et al., 2020). Together with coastal erosion of soils (30 Mg y^{-1}), river Hg inputs constitute a terrestrial Hg flux of 74 Mg y^{-1} to the Arctic Ocean that is of similar magnitude to gross atmospheric

deposition (80 Mg y^{-1}) over the Arctic Ocean (Sonke et al., 2018). Permafrost thawing has been shown to enhance river Hg export from soils to rivers (St Pierre et al., 2018). It is most pronounced in the discontinuous permafrost zone due to thawing of fresh soil organic matter and maximal active layer depth, and has been suggested to potentially double over the next 50 years (Lim et al., 2019). The quantity of atmospheric Hg deposition to northern peat soils that is presently re-emitted to the atmosphere is not well understood. Hg exchange studies indicate temporally limited Hg^0 emission from the Alaskan permafrost tundra at 68°N (Obrist et al., 2017), and year round net Hg^0 emission from Scandinavian peat at 64°N (Osterwalder et al., 2018). Other studies provide evidence for vegetation type (Rydberg et al., 2010) and temperature and insolation control (Fahnestock et al., 2019) on net Hg^0 deposition or emission.

All available data of Hg in permafrost soils originate from N-America or Scandinavia (Jensen et al., 1991; Bailey et al., 2002; Talbot et al., 2017; Schuster et al., 2018; Olson et al., 2018). Except for three studies (Golovatskaya et al., 2009; Lyapina et al., 2009; Vasilevich, 2018), we did not find extensive measurements of Hg in peat profiles from boreal and permafrost regions of the Russian Arctic and Siberia. Recent work used a soil carbon model to estimate the size of the northern permafrost soil Hg inventory to be 755 ±427 Gg (95% CI) in the upper 1 m (Schuster et al., 2018). However, this estimate is based on extrapolation of high Hg to organic carbon (C) ratios, R_{HgC}, of 1.6 Gg Hg per Pg of C (Gg Pg^{-1}) in Alaskan mineral soils to the entire North American and Eurasian permafrost zone. Another study used lower R_{HgC}, of 0.12 to 0.62 Gg Pg^{-1}, derived from observations on both Alaskan organic and mineral soils and literature data, to estimate a lower northern soil 0-1 m Hg inventory of 184 Gg (136-274 Gg, 37.5-62.5 percentiles) (Olson et al., 2018). Both studies provide the Hg inventory by multiplying the respective R_{HgC} with the same C pool estimates for the northern circumpolar permafrost region, covering an area of 17.8×10^6 km^2 (Hugelius et al., 2014).

Direct measurement of soil Hg and carbon profiles in frozen peatlands of Siberia are needed to address these variable R_{HgC} and Hg pool estimates of pan-Arctic permafrost soils. This constitutes the first and main objective of the present study. The second objective was assessing the impact of permafrost type (absent, sporadic, discontinuous and continuous) on Hg concentrations and pools in the active layer, frozen peat and mineral horizons. The third objective was to relate Hg concentration in peat to that of other trace metals in order to reveal possible mechanisms of Hg and other metal pollutant accumulation within the organic and mineral horizons of frozen peatlands. Finally, we revisit the world soil Hg inventory in order to put the pan-Arctic permafrost soil Hg pool into global perspective.

2. Study Site and Methods

2.1.Sampling sites

Soil sampling was performed along a latitudinal transect of the western Siberia lowlands (WSL) that comprised the southern taiga (Plotnikovo, 56°N), the middle taiga (Mukhrino, 60°N), the northern part of the taiga zone (Kogalym, 62°N), forest-tundra (Khanymey 62°N and Pangody, 65°N) and tundra (Tazovsky, 67°N) biomes (**Fig. 1**). In the WSL, the permafrost zones follow the temperature and vegetation distribution over the latitude at otherwise similar relief, lithology and runoff, thus allowing to test the effect of permafrost by analyzing latitudinal features of Hg distribution in soils. Key physico-geographical parameters of studied sites and soil types are listed in **Table S1**. The WSL peat actively formed since the beginning of the Holocene until freezing of bogs in the sub-Boreal period (11-4.5 ky, Kremenetski et al., 2003; Panova et al., 2010; Ponomareva et al., 2012; Loiko et al., 2019). Since 4.5 ky, the rate of peat formation and bog extension in the permafrost-affected part of the WSL have decreased. In the southern part of cryolithozone and permafrost-free part of WSL, peat accumulation and bog extension remained active over the entire Holocene (Kurina and Veretennikova, 2015; Preis and Karpenko, 2015;

127 Kurina et al., 2018). The main mineral substrates underlying frozen peat layers of the WSL are
128 quaternary clays, sands, and alevrolites. In the southern part (sites Plotnikovo and Mukhrino), the
129 typical substrate is carbonate-bearing clays of lake-alluvium origin with rare layers of sandstones
130 (**Table S1**).

131 Mean annual ambient temperature (MAAT) decreases from south to north, being equal to
132 –0.4, –1.2, –4.0, –5.6, –6.4, and –9.1°C at Plotnikovo, Mukhrino, Kogalym, Khanymey, Pandogy
133 and Tazovsky, respectively (Trofimova and Balybina, 2014). The permafrost is absent in
134 Plotnikovo but present at all other sites and ranges from relict to isolated (Mukhrino) and isolated
135 to sporadic (Kogalym) in the south, to discontinuous (Khanymey, Pangody) and continuous
136 (Tazovsky) in the north. At permafrost-affected sites, the average active layer thickness (ALT) at
137 the time of sampling of peat mounds (hummocks) ranged from 90 cm in the south to 45 cm in the
138 north. The peat mounds of ombrotrophic bogs probed in this work are present across the full
139 latitudinal gradient.

140 The vegetation of three studied types of bogs (polygonal, flat-mound and ridge-hollow) is
141 essentially oligotrophic (poor in nutrients) which indicates the ombrotrophic (rain and snow water
142 fed) conditions, i.e., the lack of groundwater input and lateral surface influx of nutrients. The flat-
143 mound palsa is covered by dwarf shrubs (*Ledum decumbens, Betula nana, Andromeda polifolia,*
144 *Vaccinium ssp., Empetrum nigrum),* lichens (*Cladonia ssp., Cetraria, Ochrolechia)* and mosses
145 (*Dicranum ssp., Polytrichum ssp., Sphagnum angustifolium, S. lenense*). At southern sites, the
146 pine *Pinus sylvestris* is abundant on ridges (Peregon et al., 2008, 2009) whereas the two taiga sites
147 are dominated by *Pinus sylvestris* with minor but permanently present *Betula pubescens* and *Pinus*
148 *sibirica*. Dwarf shrubs are dominated by *Ledum palustre, Chamaedaphne calyculata, Vaccinium*
149 *vitis-idaea.* The moss layer is dominated by *S. fuscum, S. angustifolium* with the presence of *S.*
150 *magellanicum, S. capillifolium* and boreal forest moss species like *Pleurozium schreberi*.
151

2.2. Sampling procedure, analyses and data treatment

Peat core samples were collected in August, when the depth of unfrozen layer was at its maximum (i.e., see Raudina et al., 2017). Based on measurements by temperature loggers over the summer period, the in-situ temperature of studied soil profile ranged from 15±5°C in the top soil (0-20 cm) to 4±2°C at the permafrost boundary (40-80 cm). Field logistics and financial resources did not make it possible to study multiple cores from each climate zone. The physical, chemical and botanical properties of several peat cores collected in the homogeneous palsa region in the north and ridgre-ryam complex in the south are highly similar among different peat mounds, suggesting that the cores we obtained are representative for the WSL (Velichko et al., 2011; Stepanova et al., 2015).

Peat cores were extracted using a Russian sediment/peat corer and the frozen part was sampled using a motorized Russian peat corer (UKB-12/25 I, Russia) with a 4-cm diameter corer sterilized with 40% ethanol prior to each extraction. We collected the full active (unfrozen) layer peat column, the frozen peat column and some 10 to 30 cm of frozen mineral horizons using clean powder-free vinyl gloves. Peat or mineral soil samples were divided in 5-10 cm segments using a sharp sterile single-use plastic knife. Soil samples were placed in sterile PVC doubled-zipped bags and kept at -20°C during transport and storage. To avoid contamination of peat from external surroundings, we separated the part of the core for geochemical analysis exclusively from the interior of the core (> 1 cm from the core liner) following conventional procedures (Wilhelm et al., 2011).

Total Hg concentration, THg, in freeze-dried and ground slices of peat cores was determined using a direct mercury analyzer (DMA-80 - Milestone, Italy). Analysis of reference material BCR-482 (lichen, 480±28 ng g^{-1}), MESS-3 (sediment, 91±9 ng g^{-1}) and NIST SRM 1632d (coal, 93±3 ng g^{-1}) showed good reproducibility (mean±1σ) of 467±28 ng g^{-1}, 80±6 ng g^{-1} and 98±8 ng g^{-1}, respectively. The average uncertainty on duplicate sample analysis did not exceed

5% (1σ). The carbon (C) and nitrogen (N) concentration were measured using catalytic combustion with Cu-O at 900°C with an uncertainty of ≤ 0.5% using Thermo Flash 2000 CN Analyzer, and aspartic acid (C 36.1%±1.5%; N 10.5%±0.5%) and soil reference material (C 2.29%±0.07%; N 0.21%±0.01%) as standards. Analyses of total C before and after sample treatment with HCl did not yield more than 1 % of inorganic C; therefore our total C-determination represents organic carbon. For trace and major element analysis, soil samples were subjected to full acid digestion in the clean room following ICP-MS (inductively coupled plasma mass spectrometry, Agilent 7500 ce) analyses as described previously (Morgalev et al., 2017).

The Shapiro Wilk normality test was used to asses THg, elemental and R_{HgC} distributions, and statistical data descriptors adjusted accordingly. All statistical tests used a significance level of 95% ($\alpha = 0.05$). Spearman rank order correlations (significant at $p < 0.05$) were performed to characterize the link of Hg with C, N and other major and trace elements. The differences in Hg concentration between the active- and frozen peat layer were tested using the Mann-Whitney U test for paired data at a significance level of 0.05.

C pools of different soil classes reported by Hugelius et al. (2014) were divided into two categories, organic and mineral soils. Histosols and Histels were defined as organic soils. Turbels and Orthels were considered as organic soils for the 0 – 0.3 m interval and as mineral soils for the 0.3 – 3m interval. All other soils were considered as mineral soils. To estimate the pan-Arctic permafrost soil Hg pool, C pools were multiplied with the respective R_{HgC} derived for organic (>20% C) and mineral (<20% C) soil data from Eurasia and North America (excluding Alaska). To calculate the global Hg pool, a simpler approach was used and one single R_{HgC} was considered for 5 climate zones which were defined by latitude (arctic: > 67°, boreal: 50° - 67°, temperate: 35° - 50°, subtropical: 23.45° - 35°, tropical: <23.45°) according to FAO and ITPS (2018). The uncertainty was assessed with a Monte carlo approach using the *rnorm* and *rlnorm* function of R (version 3.6.1.) and is reported as the interquartile range (25^{th} and 75^{th} percentile) of 100,000

simulations. For the pan-Arctic permafrost soil Hg pool, final uncertainties incorporate the uncertainties on the C stock from Hugelius et al. (2014) assuming normal distribution and uncertainties of R_{HgC} assuming log-normal distribution.

3. Results

3.1. Depth (vertical) distribution of Hg in WSL peat profiles

Hg concentration in peat cores of the WSL are illustrated in **Fig. 2** and primary data on soil chemical composition and Hg concentration are listed in **Table S2** of the Supplement. The upper 0-20 cm layer is 2 to 3 times enriched in Hg compared to the rest of the peat core in permafrost-affected sites (Khanymey, Pangody and Tazovsky). This is not the case, however, for the sporadic permafrost zone (Kogalym) and isolated permafrost zone (Mukhrino), where a local maximum at ca. 35 cm depth was detected but no enrichment of upper 10-20 cm horizons occurred. In the most southern, permafrost-free site of the WSL (Plotnikovo, southern taiga), the Hg concentration profile in the peat was fairly constant with a local minimum at 100 cm depth. The mean, depth-integrated Hg concentrations in active layer, permafrost and mineral horizons are illustrated in **Fig. 3** and summarized in **Table 1**. The latitudinal trend of Hg concentration in peat consists of a systematic increase northward, both in permafrost and active peat layers. The dominant ground vegetation (lichens) analyzed at 5 sites out of 6 (Plotnikovo, Kogalym, Khanymey, Pangody and Tazovsky) did not show significantly different (U test Mann-Whitney) Hg concentrations relative to the peat cores (**Fig. 3**). Mercury concentrations in the six WSL peat cores ranged from 7 to 284 ng g^{-1} with a median (± IQR) of 67±57 ng g^{-1}. The Hg concentration in the thawed, active layer was generally comparable to that in the frozen layer, supported by a Mann-Whitney test, which did not show significant difference in Hg concentration between frozen and thawed peat in all permafrost-affected sites. Within the latitudinal transect from south to north,

226 the Hg concentrations in peat are higher (Plotnikovo, Kogalym, Khanymey, Pangody) or
227 comparable (Tazovsky) to those in the mineral horizons.

228 The ratio of Hg:C (R_{HgC}, μg g^{-1}, corresponding to Gg Pg^{-1}) ranged between 0.05 and 2.0
229 over the peat columns, and was 5 to 10 times higher in mineral horizons compared to frozen peat
230 and active layers (**Fig. 4**). The R_{HgC} in the active layer and in the mineral horizons increased 3-
231 fold from the south (56°N) to the north (67°N). In the frozen peat horizon, the R_{HgC} ratio increased
232 two-fold from sporadic and isolated to continuous permafrost zone.

233

234 *3.2. Regional and total pools of Hg in the WSL peat and mineral layers*

235 The mass of Hg per area of soil in the active- and frozen peat layer as well as in the top 30
236 cm of frozen mineral horizons of the six studied WSL peat profiles was calculated by multiplying
237 bulk soil peat and mineral layer densities (range from 0.01 to 0.38 g/cm^3, **Table S2**) by Hg
238 concentration and integrating over the corresponding depths. The surface area - normalized Hg
239 stock systematically increased from south to north (ca. 0.3 to 6.0 mg Hg m^{-2} and ca. 0.8 to 13.7
240 mg Hg m^{-2}, in the 0-30 and the 0-100 cm peat layer, respectively (**Fig. 5 A**). This northward
241 increase was most pronounced for the active layer, was less evident for frozen peat, and
242 insignificant for the upper 30 cm of mineral horizon located under the peat (**Fig. 5 B**). Taking into
243 account the proportion of bogs (peatlands) in each zone (1° latitudinal grid) from Sheng et al.
244 (2004), we calculated the pool of Hg in permafrost-free and permafrost-affected WSL peatlands
245 (**Fig. 6**). The total pool of Hg in the 0-100 cm layer of peat bogs exhibits a maximum (356-580
246 Mg) in the discontinuous permafrost zone.

247 We estimate the total organic soil Hg pool in the WSL from the Hg stock (mg Hg m^{-2} over
248 0-100 cm depth) for permafrost and permafrost-free zones (**Fig. 7 A**), extrapolated to the full
249 average thickness of peat in the WSL (280 cm, Sheng et al., 2004), assuming that Hg concentration
250 in the upper 0-100 cm peat layer is the same as in 100-280 cm of peat and multiplied by the area

of bogs in each latitudinal grid (S, m^2) as shown in **Fig 7 B**. This yields 1.7 Gg Hg in the permafrost-free zone and 7.6 Gg Hg in the permafrost-bearing zone with a total Hg pool of 9.3 Gg in the WSL. For this calculation we did not take into account the mineral horizons and we used variable active layer thickness across the latitudinal gradient of WSL, as estimated at our sampling sites (**Table S1**). The amount of Hg in permafrost-bearing zone within the active (unfrozen) peat layer (0-160 cm in the south and 0-20 cm in the north) of the WSL is 2.0 Gg, and that in the frozen (160-280 cm in the south and 20-280 cm in the north) layer) is 5.6 Gg.

Alternatively, to calculate the total pool of Hg in WSL bogs, we used the R_{HgC} inferred from our data across the gradient of permafrost and biomes (Table 1). Taking into account the C pool in the WSL (70.2 Pg C of 0-280 cm depth layer, Sheng et al., 2004) and the median R_{HgC} of 0.133 Gg Pg^{-1} in the WSL, we calculated Hg for the full depth of the peat layer in each zone. This also gives 9.3 Gg Hg for a total area of 592,440 km^2.

3.3. Correlation of Hg with other elements in the peat cores

Spearman rank order correlations of Hg with other elements demonstrated significant positive relationships ($R > 0.60$; $p < 0.05$) with K, Rb, Cs, P, As, W, V, Cr, Cu in the active (unfrozen) layer (**Table S3** of Supplment). However, these relationships were less pronounced in the frozen peat, where only Mg, Ca, Sr, Mn, N, P, As, Cu, Ni, Sb and some REE demonstrated minor ($0.40 < R < 0.55$) positive correlations with Hg. Finally, in the mineral layer, significant ($R > 0.70$) positive correlations of Hg were observed with Li, Ca, Sr, P, N, Mn, Ni, Co, Cr, Cd. A positive ($R = 0.60$) relationship between Hg and C was observed in mineral horizons, whereas no correlation was detected in both frozen and thawed peat. This is consistent with some studies of peat soil in Brazil (Roulet et al., 1998) and Arctic tundra soils (Olson et al., 2018). At the same time, there was a positive correlation of Hg with nitrogen (N) in the active layer, frozen peat and

mineral horizons (R = 0.50, 0.47 and 0.75, respectively). Stronger and more stable correlation of Hg with N compared to C was also noted by Roulet (2000).

4. Discussion

4.1. Hg association with other elements in peat

Stronger accumulation of Hg relative to C in mineral horizons in the north (Tazovsky**, Fig. 4**) may be linked to the clay nature of mineral layers (Roulet et al., 1998; Baptista-Salazar et al., 2017) in these regions (**Table S1**), but also to the presence of specific host phases of Hg (see examples of peat minerals in Rudmin et al., 2018). Dissolved oxygen measurements in soil porewaters at the Tazovsky site indicate that mineral gleysoils and peat histosols, which often overlay former lake sediments, are anoxic (Raudina et al., 2017; Loiko et al., 2019). The Hg host phases in these soils are therefore likely sulfide minerals. Indeed, known Hg carriers in peat deposits are Fe and Zn sulfide minerals or organic-bound sulfide functional groups (Smieja-Król et al., 2010; 2014; Prietzel et al., 2009; Skyllberg et al., 2003, Bates et al., 1998; Steinmann and Shotyk, 1997).

In the peat active layer, Hg was positively correlated with K, Rb, Cs, P, As, V, Cr, Cu (**Table S3**). In the frozen part of the peat core, Hg was positively correlated with Ca, N, Mn, Sr, Mg, P (**Table S3**). Indeed, atmospheric particles in snow across the WSL exhibit strong enrichment in Mo, W, As, Sb, Ni, Cu, Zn, Cd, Pb, Mg, Ca, and Na (Shevchenko et al., 2017). The strong positive correlation of Hg with these elements in peat soils of WSL suggests a common atmospheric origin. Note however, that the cited elements deposit with particles, rainfall and snowfall, whereas atmospheric Hg transfer to peat occurs mainly via the vegetation pump, with tundra and taiga vegetation actively taking up atmospheric gaseous Hg^0 through foliage (Obrist et al., 2017; Jiskra et al., 2018).

4.2. Estimating the pan-Arctic permafrost soil Hg pool

A recent study used a median R_{HgC} value of 1.6 Gg Pg^{-1}, observed mainly in mineral soil samples (median SOC of 3%, IQR=1.7 to 8.7 %) along a transect in Alaska, to estimate a pan-Arctic permafrost soil Hg pool of 755±427 Gg in the upper 0-100 cm, and 1656±962 Gg in the upper 0-300 cm (Schuster et al. 2018). In the case of western Siberia, this high R_{HgC} value overestimates the Hg pool 12-fold, given that the median R_{HgC} in WSL peat is only 0.13±0.12 Gg Pg^{-1} (median±IQR) (**Table 1, Fig. 4**). The extrapolation based on Alaskan R_{HgC} for the whole Northern Hemisphere permafrost region also suggests that the WSL contains large amounts of Hg in the upper 0-30 cm (20-40 mg Hg m^{-2}) and in the upper 0-100 cm (40-80 mg Hg m^{-2}). These numbers are much higher than the direct measurements in this study: 0.3 mg Hg m^{-2} in 0-30 cm and 0.8-1.3 mg Hg m^{-2} in 0-100 cm layer in the permafrost-free zone (Plotnikovo and Mukhrino sites); 0.5 mg Hg m^{-2} in 0-30 cm and 3.0 mg Hg m^{-2} in 0-100 cm layer in the sporadic zone (Kogalym site); 1.8-4.0 mg Hg m^{-2} in 0-30 cm and 9.6-11.9 mg Hg m^{-2} in 0-100 cm layer in the continuous to discontinuous permafrost zone (Khanymey and Pangody sites), and 6.0 mg Hg m^{-2} in 0-30 cm and 13.7 mg Hg m^{-2} in 0-100 cm layer in continuous permafrost zone (Tazovsky). It is worth noting that the recent data of Talbot et al. (2017) for Ontario (Canada) bogs (S_{area} = 1,133,990 km²; 18.8 Gg Hg for the 277 ± 123 cm depth) are consistent with the results of the present study in the WSL (9.3 Gg Hg for 592,440 km² for the 280 ± 100 cm depth).

A revised value of the Hg pool in the pan-Arctic permafrost soils was recently provided by Olson et al. (2018) who combined measured R_{HgC} values for Alaskan tundra soils (Dalton Highway, Noatak National Preserve, 8 Mile Lake Observatory) with literature data (Table S4), and derived R_{HgC} of 0.12 Gg Pg^{-1} for 0-30 cm (organic) and 0.62 Gg Pg^{-1} for 30-100 cm (mineral) layers. Note that Olson et al. Table 1 has an incorrect organic soil R_{HgC} of 0.274 Gg Pg^{-1}, which should be 0.119 Gg Pg^{-1}; the typo did not affect their soil Hg budgets. Olson et al. (2018) estimate pan-Arctic permafrost soil Hg pools of 26 Gg (0-30 cm) and 158 Gg (30-100 cm), which combined

(184 Gg) is 4 times lower than the number of 755 Gg (0-100 cm) by Schuster et al. (2014). Both studies report R_{HgC} measurements from Alaskan soils, due to relatively easy road access to the sampling sites along the Dalton Highway. The bedrock along the Dalton Highway contains relatively high geogenic Hg levels (mean concentration: 32 ng/g), resulting in a high geogenic contribution in mineral soils (39% for B horizons and 20% for A horizons, Obrist et al., 2017). It is clear that any upscaling calculation of pan-Arctic permafrost Hg depends critically on the R_{HgC} of the 0-30 and 30-100 cm peat layers, as Eurasian sporadic to continuous permafrost represents 54% of the northern soil C inventory (**Table 2**). We combined our new WSL data (n=202) with literature data covering Russia (n=42), Scandinavia (n=97), Canada (n=122) and Alaska (n=703) to build a pan-arctic database for RHgC (Table S4). Setting Alaska aside, as a geographic region with high geogenic Hg, we find that Eurasian and North American (excluding Alaska) mineral (<20% SOC) soil R_{HgC} was lower (μ = 0.67 Gg Pg^{-1}, median= 0.64 Gg Pg^{-1} (IQR = 0.33 to 0.79 Gg Pg^{-1}), n=130) than R_{HgC} reported for mineral soils along the Dalton highway in Alaska (median= 1.64 Gg Pg^{-1} (IQR = 0.91 to 2.93 Gg Pg^{-1}), n=589) (Schuster et al., 2018) (**Figure 8**). The R_{HgC} in organic soils (>20% SOC, including data from Alaska) was approximately 4 times lower (μ= 0.19 Gg Pg^{-1}, median= 0.15 Gg Pg^{-1} (IQR = 0.09 to 0.25 Gg Pg^{-1}), n=446) than that in mineral soils of Eurasia and North America (excluding Alaska) (**Figure 8**), consistent with the observed difference in WSL mineral and organic soils. Higher R_{HgC} observed in mineral soils may originate from a contribution of geogenic Hg from the weathered bedrock (independent of C stock) and/or a higher mineralization rate of C (preferential C over Hg loss) in predominantly oxic mineral soils compared to anoxic peat soils.

In **Table 2** we revisit the full 0-300 cm pan-Arctic permafrost soil Hg inventory. For this, we used Eurasian and North-American (excluding Alaska) R_{HgC} based on the literature data compilations of Olson et al. (2018) and Schuster et al. (2018), and our observed WSL R_{HgC} for Eurasia, multiplied by estimated organic C pools for the northern permafrost region covering 17.8

350 $\times 10^6$ km^2 from Hugelius et al. (2014). The systematic underestimation made by neglecting high
351 R_{HgC} in Alaskan mineral soils is small, on the order of 2.5 Gg Hg, due to relatively small Alaskan
352 C pool of 2.6 Pg C (Tarnocai et al., 2009). We estimate the pan-Arctic permafrost soil Hg pool to
353 be 72 Gg (39-91 Gg, IQR) in the upper 30 cm, 240 Gg (110-336 Gg) in the upper 1 m, and 597
354 Gg (384-750 Gg) in the upper 3 m (**Table 3**). Note that our revised value in the 0 - 1m range (240
355 Gg) is similar to that of Olson et al. (184 Gg), but sizably lower than that of Schuster et al. (755
356 Gg). We find that Hg stocks in organic soils (>20% SOC) represent 52% and 21% of the total Hg
357 stock in the 0-30 cm and 0-100 cm depth range, respectively (**Table 2**). The rest of the pan-arctic
358 Hg is associated with C in mineral soils (<20% SOC) for which relatively sparse data exists
359 (n=131). In particular, turbel and orthel mineral soils, which are estimated to contain 49 to 62%
360 of total arctic C (Hugelius et al., 2014) and 36 to 85% of Hg at the various depth intervals need to
361 be further investigated.
362
363 *4.3. Estimating Earth's global soil (0-30 cm) Hg pool*
364 To assess Earth's global soil Hg pool, we combined the more detailed Arctic pool estimate
365 (separating organic and mineral soils) with a more basic approach for the other climate zones,
366 where we derived bulk R_{HgC} for the 0-30 cm surface soils based on published literature data. Core
367 dataset for this analysis was the global literature compilation by Schuster et al. 2018 with 11,000
368 datapoints, of which 6,224 datapoints were from 0-30 cm range. This global dataset was
369 complemented by soil Hg data from the boreal (Olson et al., 2018) and tropical (Campbell et al.,
370 2003; Almeida, 2005; Almeida et al. 2005; Melendez-Perez et al., 2014) zone. The R_{HgC} for each
371 climate zone (arctic, boreal, temperate, subtropical and tropical) were then multiplied with the
372 respective C stock estimates from the global soil organic carbon map (FAO and ITPS, 2018). The
373 R_{HgC} of all 0 to 30 cm soil data increases from cold climate zones to warmer climate, from 0.36
374 Gg Pg^{-1} for Arctic soils (excluding Alaska), to 1.8 Gg Pg^{-1} in subtropical and tropical soils (**Figure**

375 **9, Table 4**). This latitudinal trend in R_{HgC} likely reflects a combination of low C mineralization
376 rates in colder north and additional Hg sorption to Fe(oxi)hydroxides in old tropical soils. Taking
377 into account the variation in R_{HgC} and C stocks across the climate zones, we estimate a global Hg
378 stock of 1086 Gg (852 – 1265 Gg, IQR) for the top 0 - 30 cm (**Table 4**). Previous global Hg soil
379 pool estimates vary between 232 and 1150 Gg (Selin et al., 2008; Smith-Downey et al., 2010;
380 Amos et al., 2013, 2015; Hararuk et al., 2013; Wang et al., 2019). Schuster et al. (2018) concluded
381 that Arctic permafrost soils store nearly twice the amount of Hg as all other soils, the ocean and
382 atmosphere combined, but in doing so they compared different depth ranges for global (typically
383 0-15 cm or 0-20 cm) and Arctic soil estimates (0 - 300 cm). Our revised estimate of the pan-Arctic
384 permafrost and global soil pool suggests that, for a similar depth range of 0-30 cm, permafrost
385 soils contain 6% (72 Gg) of the global soil Hg pool (1086 Gg).
386
387 *4.4. Northern soil Hg sequestration and Hg loss*
388 Olson et al. (2018) recognized that the large 0-100 cm northern soil Hg pool is the result
389 of thousands of years of net atmospheric Hg deposition. The latitudinal trend of northward
390 increasing peat Hg concentration in the WSL (**Fig. 4, 5**) illustrates that this net Hg deposition is a
391 fine balance between the vegetation Hg pump, which sequesters Hg^0 in soils via foliar uptake and
392 litterfall, and Hg^0 emission during biomass degradation. Annual gross Hg sequestration by
393 vegetation, via the vegetation pump, likely scales with primary productivity and therefore
394 decreases northward as insolation and growing season decrease. However, in the north,
395 degradation rates of vegetation biomass are lower than in the south: the moss biomass losses
396 during decomposition in the forest tundra zone (5-6% over 1st year and 10-12% over 2 years) is
397 lower than that in the southern taiga (10-20% over 1st year and 20-40% over 2 years), based on in-
398 situ biomass degradation experiments across the WSL gradient of biomes (Vishnyakova and
399 Mironycheva-Tokareva, 2018). The net result is a higher preservation of soil Hg in the north,

where less emission of Hg^0 during plant decay occurs. The dependence of this balance, between Hg^0 sequestration and Hg^0 re-emission, on climate explains qualitatively the contrasting observations made in Toolik (AK, USA, 68°N, MAAT = -7°C, Obrist et al., 2017) and Degerö Stormyr (Sweden, 64°N, MAAT = 2°C, Osterwalder et al., 2018). At Toolik, net Hg^0 deposition by vegetation and soil uptake occurs on an annual basis, whereas at Degerö Stormyr higher temperatures result in net annual Hg^0 emission. More research is needed to quantify the climate dependence of Hg^0 sequestration (as soil Hg^{II}) and Hg^0 re-emission before we can predict and model northern soil Hg loss to the atmosphere due to global warming trajectories.

5. Conclusions

Western Siberian peatlands contain a large amount of Hg in frozen and thawed peat; the lateral pools of peat palsa bogs range from 1-2 mg Hg m^{-2} in the south to 10-15 mg Hg m^{-2} in the north. This northward increase of Hg concentration and pools can be explained by better preservation of organic-bound Hg, i.e. lower Hg re-emissions, due to colder temperatures and shorter active period in the continuous permafrost zone compared to the discontinuous and sporadic zones. We revisited the full 0-300 cm pan-Arctic permafrost soil Hg inventory, based on published R_{HgC} and our observed WSL R_{HgC}, together with estimated pan-Arctic permafrost soil organic C pools for 0-300 cm from Hugelius et al. (2014). We estimate the 0-300 cm pan-Arctic permafrost soil Hg inventory to be 597 Gg (384-750 Gg, IQR), which is three times lower than a previous estimate of 1656 ± 962 Gg Hg for the same depth range (Schuster et al., 2018). We estimate the global soil Hg pool to be 1086 Gg for the 0-30 cm depth range. The permafrost Hg pool for the same 0 - 30 cm depth range is 77 Gg, and while large compared to the 3 Gg of Hg residing in the Arctic Ocean (Soerensen et al., 2016), it represents only 6% of the global soil Hg pool.

We document large regional differences in R_{HgC} driven by (1) different contributions of geogenic Hg in mineral soils, e.g. resulting in higher R_{HgC} in Alaska than in other areas of the pan-

Arctic permafrost region and (2) the stability of Hg with respect to emission from organic soils, e.g. resulting in a gradient with increasing R_{HgC} towards the north of the WSL. These systematic differences illustrate the limitations of the poolsize estimation approach where C inventories are multiplied with average R_{HgC} values and emphasize the need for spatially resolved sampling and pool size estimates, similar to the pan-Arctic permafrost C pool estimates (Hugelius et al., 2014). In particular, to estimate the release of Hg to aquatic ecosystems, e.g. coastal erosion and transfer to rivers, and Hg evasion to the atmosphere, spatially resolved Hg soil pools will be valuable.

Data availability. Hg and C concentration data of the WSL soil samples are available in the supplement. A summary of pan-Arctic literature data is provided in the supplement. The permafrost data from Schuster et al. 2018 and a global compilation of R_{HgC} data is available at (https://agupubs.onlinelibrary.wiley.com/doi/full/10.1002/2017GL075571, last access: 6 December 2019). The Arctic and boreal soil data from the Olson et al. 2018 study is available from the corresponding author upon request. Note that Olson et al. Table 1 has an incorrect organic soil R_{HgC} of 0.274 Gg Pg^{-1}, which should be 0.119 Gg Pg^{-1}. The data from the tropical climate zone was compiled from original publications of Almeida (2005); Almeida et al. (2005); Campbell et al. (2003); Melendez-Perez et al. (2014).

Author contributions. OSP and SVL designed the study. SVL, AGL, and NK performed field sampling. AGL and OSP did all laboratory analysis. MJ and JES did the northern soil and global soil Hg pool calculations. All authors contributed to writing of the manuscript.

We declare no competing interests

Acknowledgements:

Fieldwork and data analysis was carried out under support from the Russian Science Foundation (project №18-77-10045). Partial support from the Russian Fund for Basic Research, via grant no. 19-29-05209-mk and 19-55-15002-NCNI_a, and from the CNRS Chantier Arctique Français, via the PARCS project, and the H2020 ERA-PLANET (689443) iGOSP and iCUPE programmes are acknowledged. M.J. acknowledges funding by the Swiss National Science Foundation (grant no. PZ00P2_174101). We are thankful to Luiz D. Lacerda for sharing Hg and C data from Brazil.

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743 **Table 1.** Mean (± SD) concentrations and stocks of C and Hg in 6 studied sites of Western
744 Siberia Lowlands (WSL) peatbogs.
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Horizons	C, %	Hg, ng g^{-1}	kg C m^{-2}	mg Hg m^{-2}	R_{HgC} (Gg Pg^{-1})
Plotnikovo (Pl), Southern taiga, 56.9°N*					
ALT (0-140 cm)	45±2	36±12	24	2.8	0.08±0.03
Mineral (140-150 cm)	13	53	23	9.0	0.40
Total (150 cm)	43±8	37±12	47	11.8	0.10±0.09
0-30 cm	44±2	44±5	3	0.3	0.10±0.01
0-100 cm	45±2	36±10	9	0.8	0.08±0.02
Mukhrino (Mh), Middle taiga, 60.9°N*					
ALT (0-360 cm)	53±7	26±13	67	4.3	0.05±0.03
Mineral (360-380 cm)	15±19	24±19	51	8.3	0.46±0.46
Total (380 cm)	51±10	26±19	118	12.6	0.07±0.12
0-30 cm	50±0.4	29±7	4	0.3	0.06±0.02
0-100 cm	52±5	32±17	18	1.3	0.06±0.04
Kogalym (Kg), Northern taiga, 62.3°N*					
ALT (0-175 cm)	48±4	48±30	93	8.7	0.10±0.06
Mineral (175-190 cm)	10±13	12±10	17	2.3	0.34±0.36
Total (190 cm)	45±12	45±30	110	11	0.12±0.12
0-30 cm	45±1	65±19	3	0.5	0.14±0.04
0-100 cm	47±2	49±34	38	3	0.11±0.07
Khanymey (Kh), Northern taiga, 63.8°N					
ALT (0-34 cm)	44±2	64±43	17	2.1	0.15±0.10
PF1 (34-100 cm)	50±2	47±13	78	7.6	0.09±0.02
PF2 (34-138 cm)	48±6	47±11	119	11.8	0.10±0.02
Mineral (138-147 cm)	1±1	4±1	2	0.5	0.31±0.13
Total (147 cm)	42±16	47±28	138	14.4	0.13±0.09
0-30 cm	43±1	71±46	13	1.8	0.17±0.11
0-100 cm	47±4	54±29	95	9.6	0.12±0.07
Pangody (Pg), Forest tundra, 65.9°N					
ALT (0-40 cm)	50±4	78±25	38	5.3	0.16±0.07
PF1 (40-100 cm)	53±4	61 ±25	54	6.6	0.11±0.04
PF2 (40-155 cm)	48±10	67±23	78	11.0	0.15±0.07
Mineral (155-185 cm)	3±1	24±11	15	12.5	0.88±0.28
Total (185 cm)	41±19	62±28	130	28.8	0.27±0.30
0-30 cm	50±5	83±26	26	4.0	0.17±0.07
0-100 cm	52±4	68±25	92	11.9	0.13±0.06
Tazovsky (Tz), Southern tundra, 67.4°N					
ALT (0-40 cm)	49±3	186±110	22	7.4	0.38±0.20
PF1 (40-100 cm)	46±3	109±28	27	6.3	0.23±0.06
PF2 (40-380 cm)	47±4	104±39	156	35.0	0.22±0.08
Mineral (380-405 cm)	14±5	152±65	60	64.7	1.24±0.66
Total (405 cm)	45±9	115±57	238	107.0	0.30±0.31
0-30 cm	49±4	209±120	16	6.0	0.42±0.21
0-100 cm	47±3	140±80	48	13.7	0.29±0.15

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747 **Footnote**: ALT is Active Layer Thickness; PF1 is frozen peat, (ALT-100 см); PF2 is frozen peat
748 (ALT to mineral layer); 'Mineral' is mineral layer; 'Total' is total Hg content averaged over full
749 sampled depth. *In permafrost-free zone, the ALT extends from the surface to the mineral layer.
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Table 2. Estimated pan-Arctic permafrost soil Hg inventory (Gg) for different depth ranges down to 300 cm. Hg pool uncertainties are reported as the interquartile range (IQR), i.e. the 25th to 75th percentiles of the Hg pool distribution estimates by a Monte Carlo method. Soil organic carbon (SOC) pools are from Hugelius et al. (2014).

depth range	soils	SOC	Hg Pool	IQR		% of total
		Pg	Pg			
0-30 cm	organic (>20% SOC)	172	35	13	45	48
	mineral (<20% SOC)	45	37	15	47	52
	total	**217**	**72**	**39**	**91**	
0-100 cm	organic (>20% SOC)	253	50	26	65	21
	mineral (<20% SOC)	219	191	59	287	79
	total	**472**	**240**	**110**	**336**	
0-200 cm	organic (>20% SOC)	366	72	44	92	16
	mineral (<20% SOC)	461	392	196	529	84
	total	**827**	**464**	**269**	**601**	
0-300 cm	organic (>20% SOC)	427	85	55	106	14
	mineral (<20% SOC)	607	512	300	666	86
	total	**1034**	**597**	**384**	**750**	

Table 3. Comparison of estimated pan-Arctic permafrost soil Hg pools by different studies.

	Hugelius et al. 2014		Schuster et al. 2018		Olson et al. 2018		this study			
depth range	SOC	95% CI	Hg Pool	95% CI	Hg Pool	25% CI	Hg Pool	IQR		
	Pg	Pg	Gg	Gg	Gg	Gg	Gg	Gg		
0-30 cm	217	12	347	±196	26	21 - 42	72	39	-	91
0-100 cm	472	27	755	±301	184	136 - 274	240	110	-	336
0-200 cm	827	108	1323	±764			464	269	-	601
0-300 cm	1034	150	1656	±962			597	384	-	750

[1] Confidence interval (CI) corresponding to the 37.5th to 62.5th percentile

[2] interquartile range (IQR), i.e. the 25th to 75th percentiles

Table 4. Estimated soil Hg pool (0 to 30 cm interval) for different climate zones based on reported R_{HgC} and carbon pools. Hg pool uncertainties are reported as the interquartile range (IQR), i.e. the 25th to 75th percentiles of the Hg pool distribution estimates by a Monte Carlo method.

Climate zone	C pool Pg	median R_{HgC} (Gg/Pg)	mean R_{HgC} (Gg/Pg)	Hg pool Gg	IQR Gg	
tropics	208	1.85	2.14	446	268	- 556
subtropics	102	1.83	2.13	217	128	- 271
temperate	191	1.37	1.55	296	183	- 366
boreal	140	0.35	0.41	55	35	- 67
arctic	217	(0.15, 0.64)	(0.19, 0.67)	72	39	- 91
total	**858**			**1086**	**852**	**1265**

[1] Carbon pools are from FAO and ITPS (2018).
[2] The arctic R_{HgC} and Hg pool are from Table 2.
[3] The arctic carbon pool is from Hugelius et al. (2014)

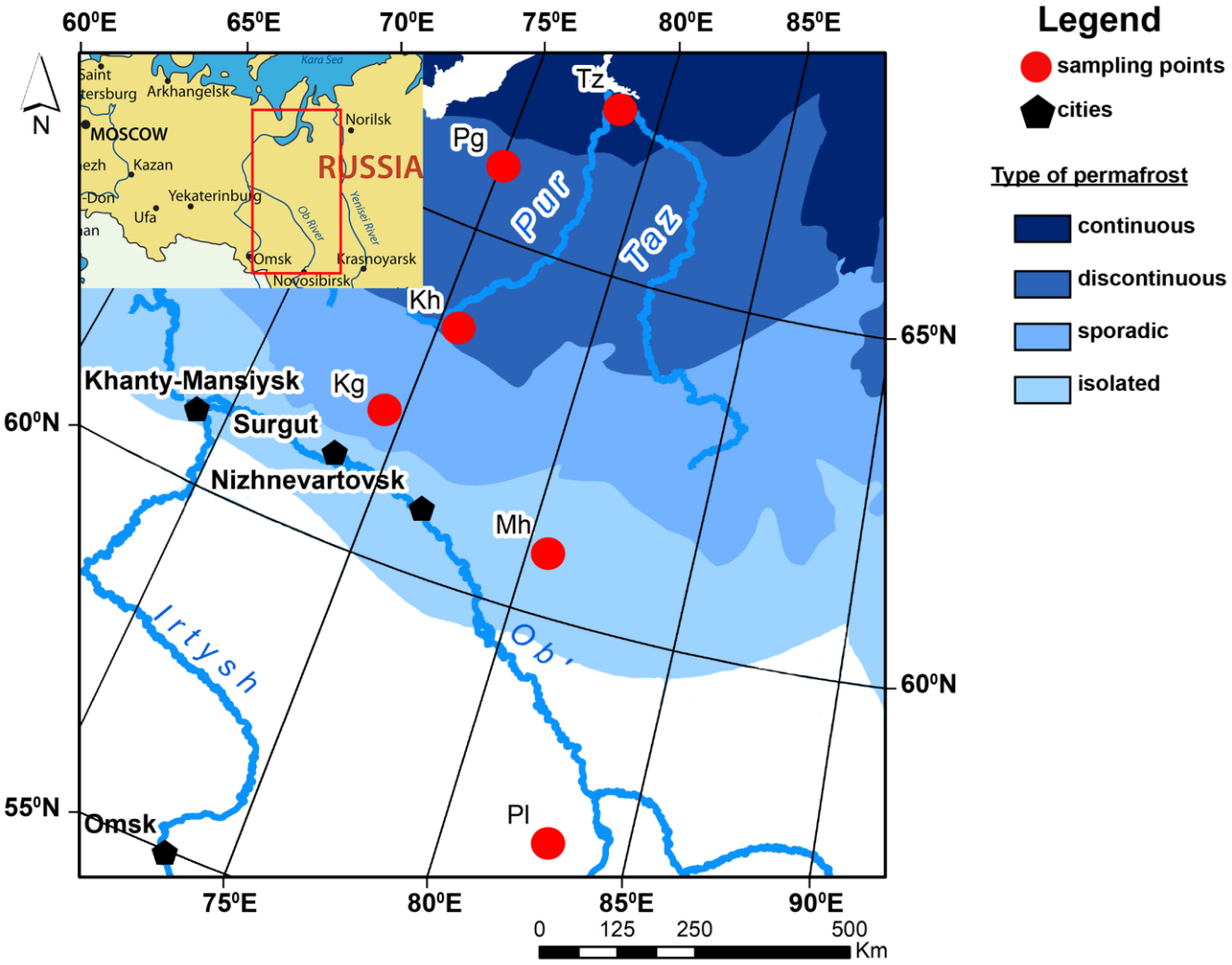
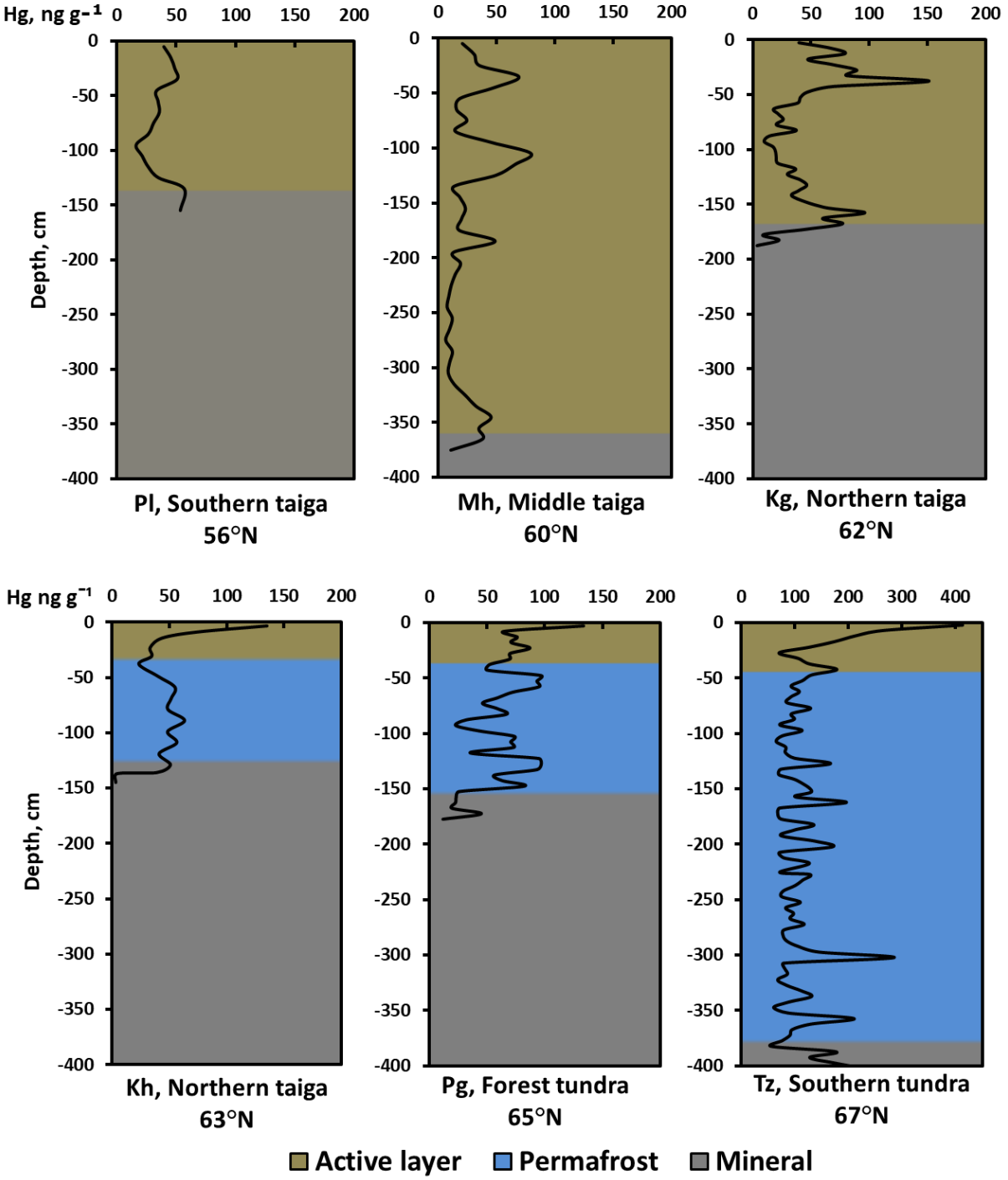


Fig. 1. Sampling sites and permafrost boundaries (modified after Brown et al., 2001) of WSL territory investigated in this work. The climate and soil parameters of 6 sampling sites (Tazovsky Tz, Pangody Pg, Khanymey Kh, Kogalym Kg, Mukhrino Mh, and Plotnikovo Pl) are listed in Supplementary Table S1.



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800 **Fig. 2.** Vertical depth profile distribution of total Hg (THg) in 6 peat cores across a 1700 km
801 latitudinal transect of the WSL. Site location and physio-geographical parameters are shown in
802 Fig. 1 and Table S1.
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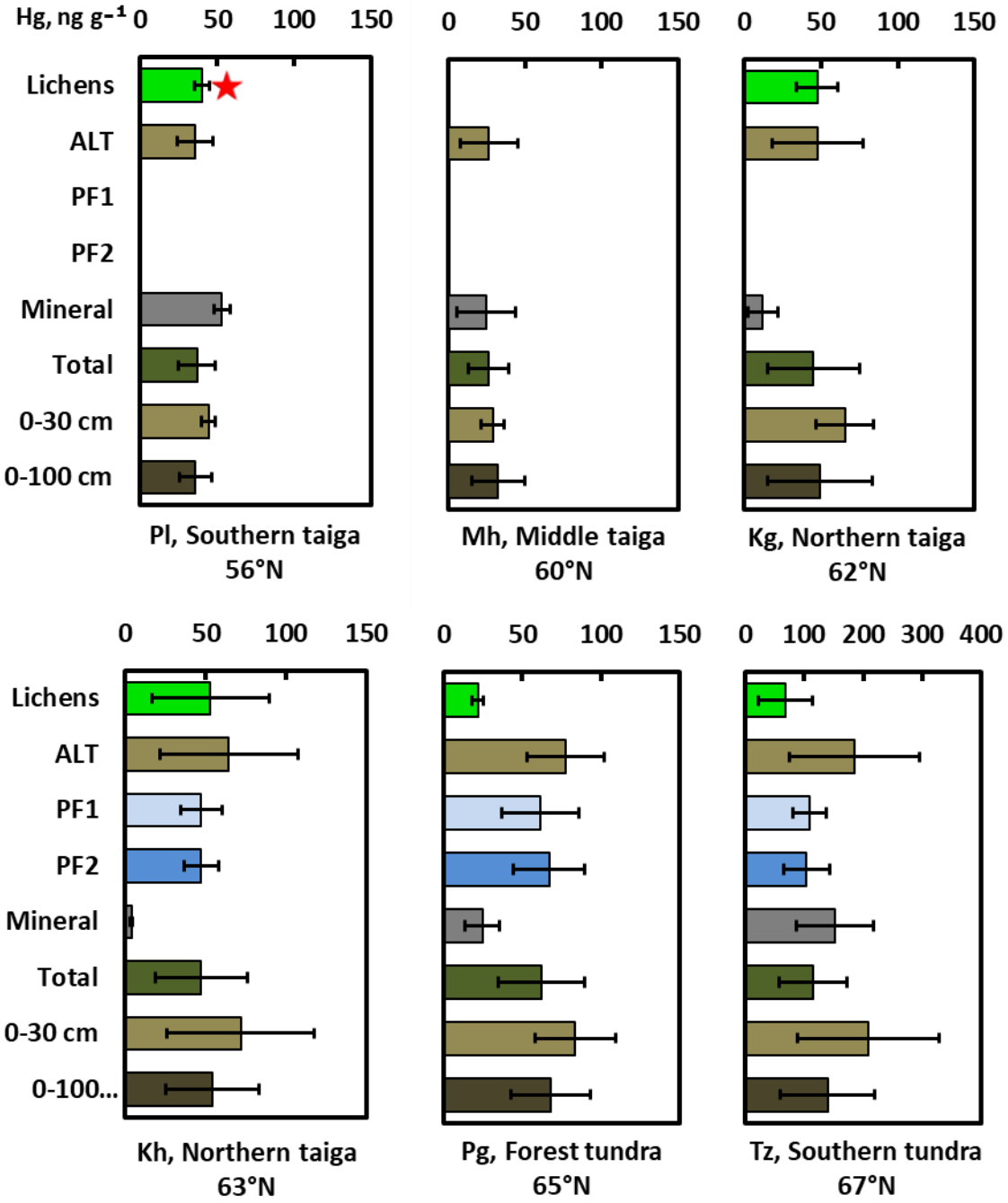


Fig. 3. Mean (±SD), depth-integrated Hg concentrations in peat columns and mineral layers of 6 studied sites. Red asterisk represents the data from Lyapina et al. (2009).

Footnote: ALT is Active Layer Thickness; PF1 is frozen peat, (ALT-100 см); PF2 is frozen peat (ALT to mineral layer); 'Mineral' is mineral layer; 'Total' is total Hg content averaged over full sampled depth. *In permafrost-free zone, the ALT extends from the surface to the mineral layer.

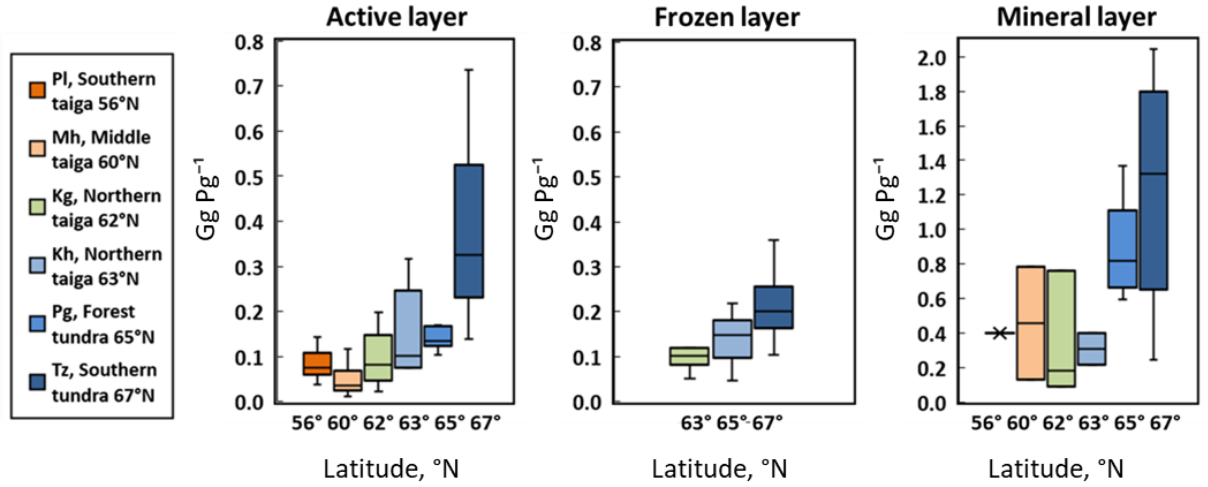


Fig. 4. The ratio Hg:C (Gg Pg^{-1}), median±IQR, in the active layer, frozen peat and mineral horizons across the WSL latitudinal transect.

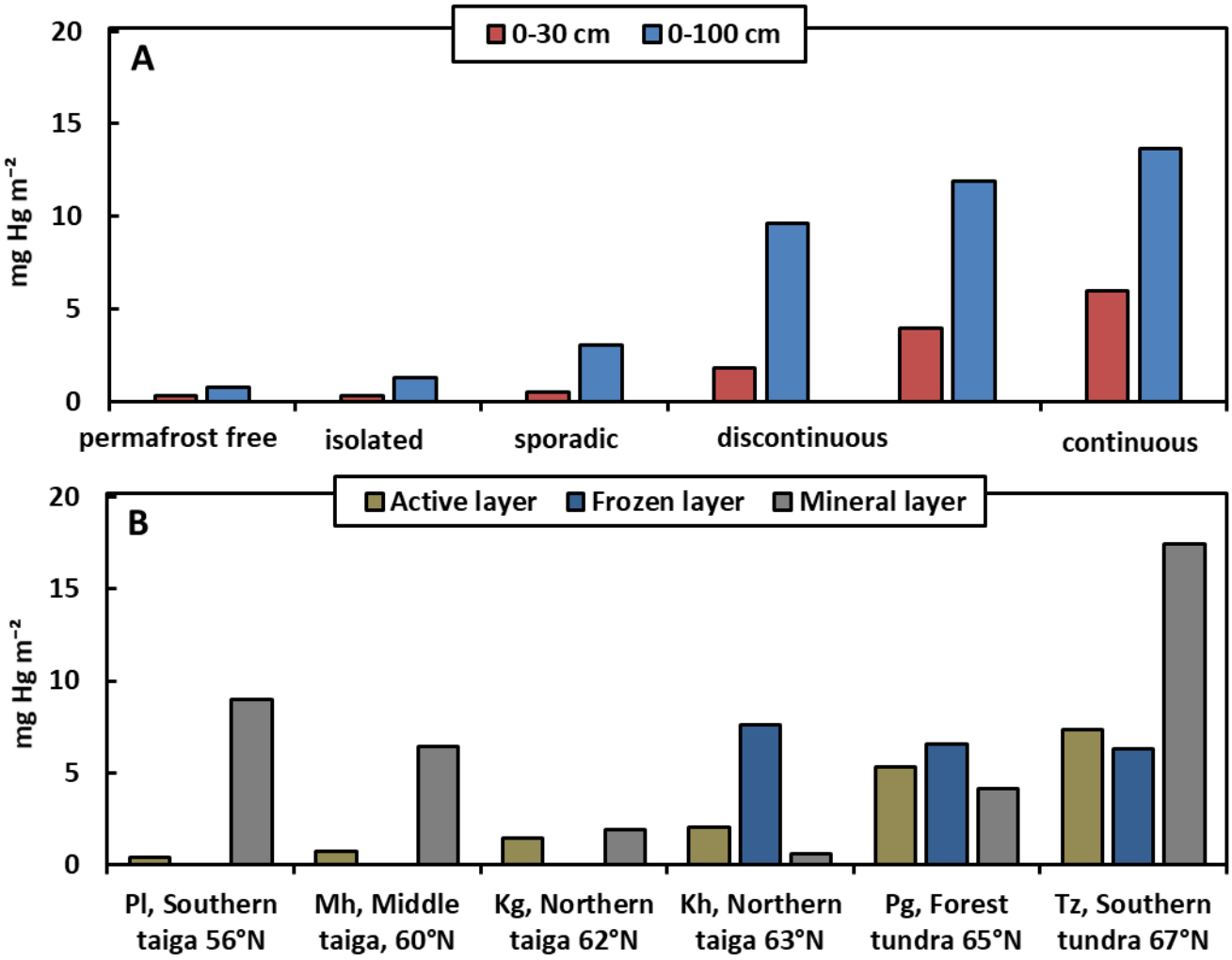


Fig. 5. Latitudinal variation in WSL soil Hg storage (mg Hg m^{-2}) in the 0-30 and 0-100 cm peat layer (**A**) and in the active, frozen and mineral layers (**B**). In the permafrost-free zone, the first 40 cm were used to calculate Hg storage in the active layer. The permafrost peat layer is fixed from the lower boundary of the active (unfrozen) layer down to 100 cm. Finally, for the mineral layer we considered only the first 10 cm below peat deposits across the full latitudinal gradient of the WSL peatland.

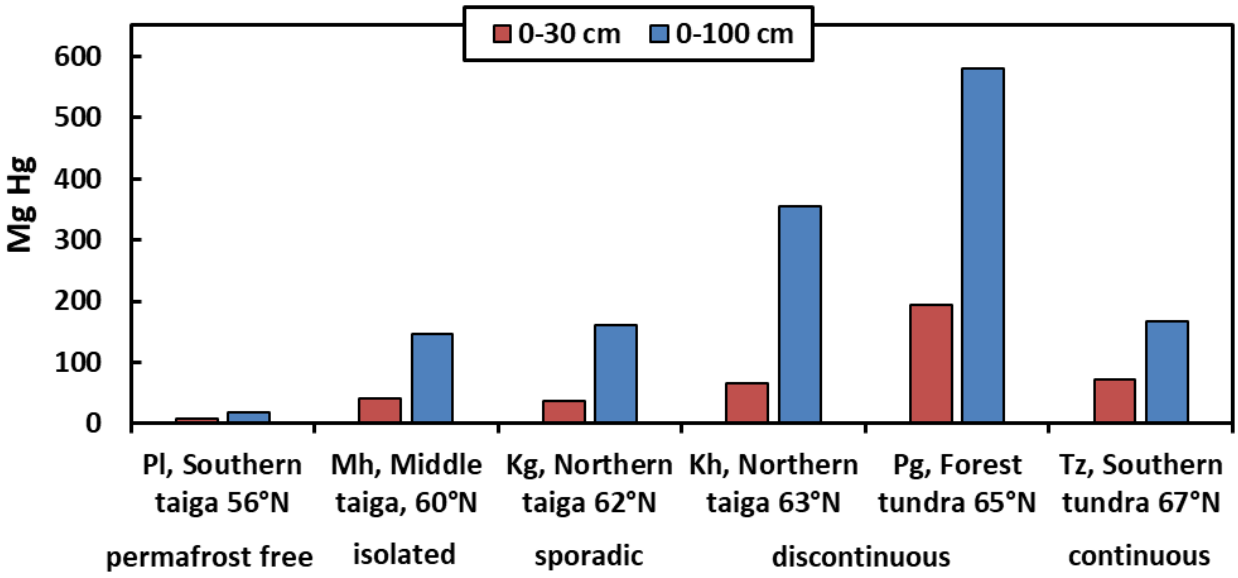
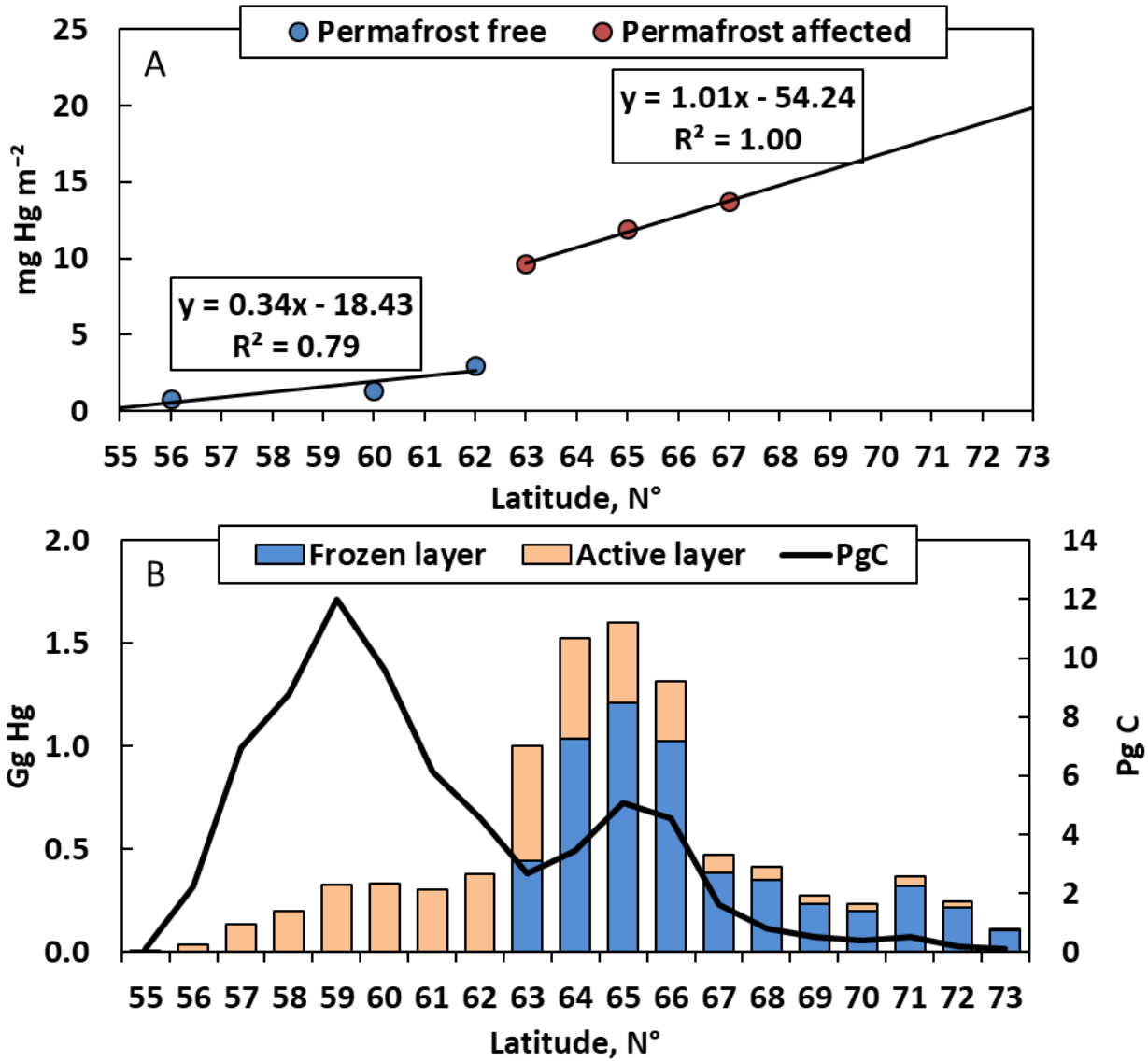


Fig. 6. Total, depth-integrated pools of Hg mass (Mg) in the upper 0-30 and 0-100 cm (red and blue columns, respectively) of WSL frozen peatlands in each permafrost zone. The stocks are calculated assuming the areal proportion of bogs from the landscape inventory across the WSL (Sheng et al., 2004).



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865 **Fig. 7**. Estimated Hg storage (mg Hg m^{-2}) in the 0-100 cm soil layer of the WSL (**A**) and
866 latitudinal distribution of the Hg pool (Gg) in active and frozen peat layers of the WSL (**B**).
867 Solid black line represents the WSL C pool (Pg) from Sheng et al. (2004).

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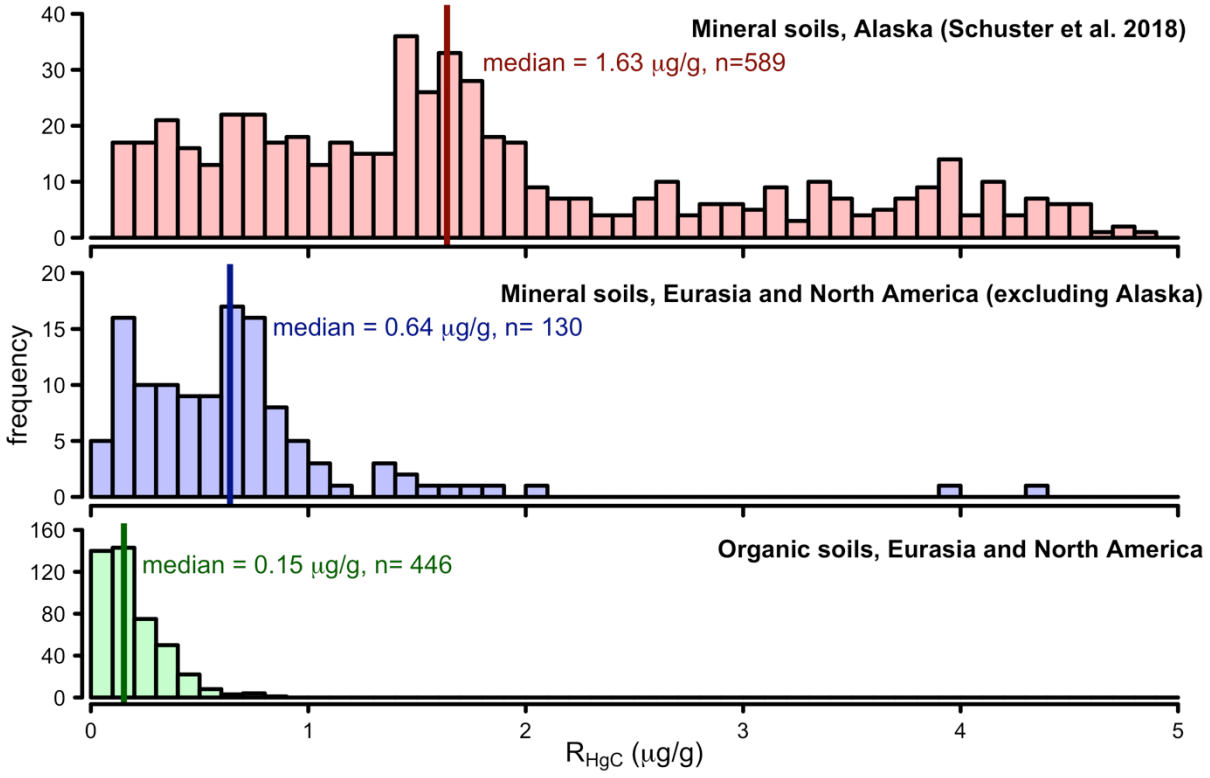

Fig. 8. Histogams of published (Table S4) and WSL (this study) R_{HgC} data used for estimating the northern circumpolar permafrost region soil Hg pool: top: Alaskan mineral soils (Schuster et al., 2018), middle: mineral soils from Eurasia and North America (excluding Alaska), and bottom: organic soils from Eurasia and North America (including Alaska).

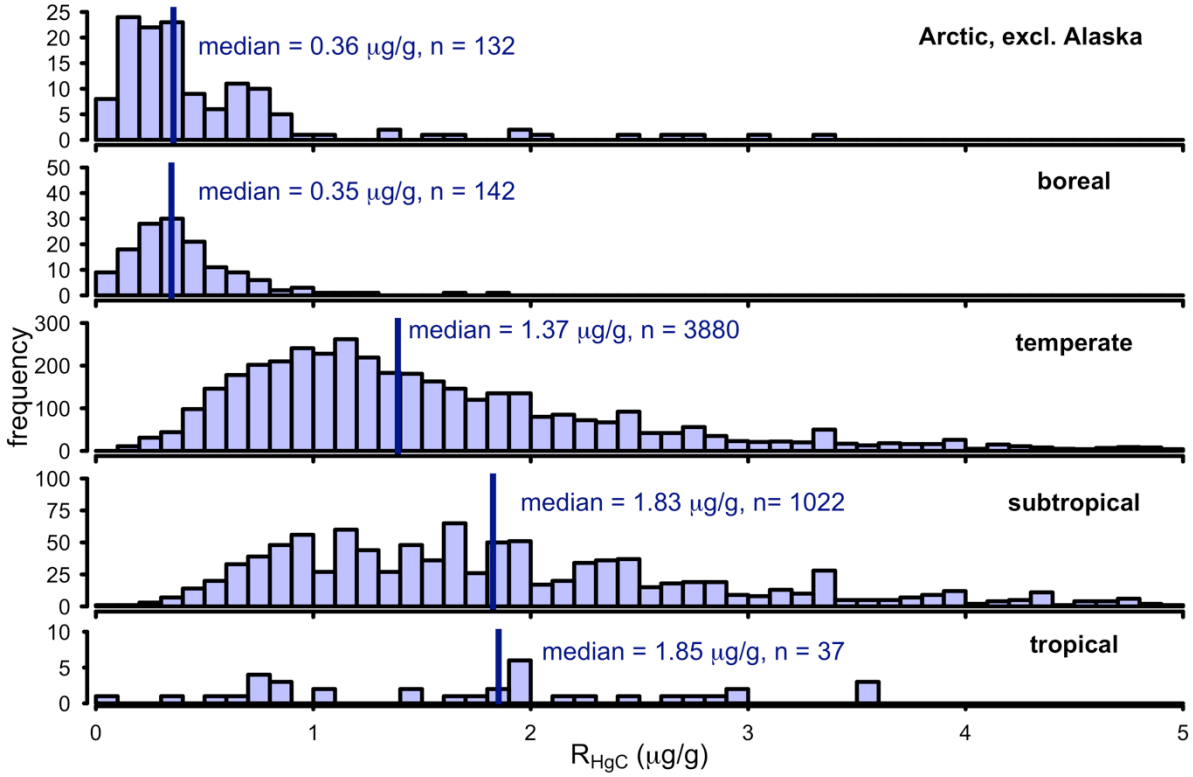

Fig. 9. Histograms and median R_{HgC} for the 0 - 30 cm soil interval for different global climate zones.