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## Enhanced particulate Hg export at the permafrost boundary, western Siberia

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*Key words: particulate, suspended, landscape, bog, lake, forest, thaw, Siberia*

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### **Bullet points:**

- Particulate Hg (PHg) concentration in western Siberia rivers decreases with watershed size
- PHg correlates with organic carbon and anti-correlates with lithogenic elements
- Hg mobilization from organic (peat) rather than mineral soil layers to the rivers
- Maximum PHg concentration and export fluxes in the sporadic permafrost zone
- Climate warming and permafrost thaw may double PHg export to the Arctic Ocean

### **Capsule:**

Enhanced Hg mobilization from peat soil to the rivers at the permafrost boundary, due to maximal depth of the thawed layer

33 **Abstract**

34 Arctic permafrost soils contain large amounts of organic carbon and the pollutant  
35 mercury (Hg). Arctic warming and associated changes in hydrology, biogeochemistry and  
36 ecology risk mobilizing soil Hg to rivers and to the Arctic Ocean, yet little is known about the  
37 quantity, timing and mechanisms involved. Here we investigate seasonal particulate Hg (PHg)  
38 and organic carbon (POC) export in 32 small and medium rivers across a 1700 km latitudinal  
39 permafrost transect of the western Siberian Lowland. The PHg concentrations in suspended  
40 matter increased with decreasing watershed size. This underlines the significance of POC-rich  
41 small streams and wetlands in PHg export from watersheds. Maximum PHg concentrations and  
42 export fluxes were located in rivers at the beginning of permafrost zone (sporadic permafrost).  
43 We suggest this reflects enhanced Hg mobilization at the permafrost boundary, due to maximal  
44 depth of the thawed peat layer. Both the thickness of the active (unfrozen) peat layer and PHg  
45 run-off progressively move to the north during the summer and fall seasons, thus leading to  
46 maximal PHg export at the sporadic to discontinuous permafrost zone. The discharge-weighted  
47 PHg:POC ratio in western Siberian rivers ( $2.7 \pm 0.5 \mu\text{g Hg: g C}$ ) extrapolated to the whole Ob  
48 River basin yields a PHg flux of  $1.5 \pm 0.3 \text{ Mg y}^{-1}$ , consistent with previous estimates. For current  
49 climate warming and permafrost thaw scenarios in western Siberia, we predict that a northward  
50 shift of permafrost boundaries and increase of active layer depth may enhance the PHg export by  
51 rivers to the Arctic Ocean by a factor of two over the next 10-50 years.

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## 58 1. Introduction

59 The high-latitude critical zone is subject to changes in hydrological regime (Bring et al.,  
60 2016; Wrona et al., 2016) due to widespread permafrost thaw. Climate warming also stimulates  
61 organic carbon (OC) release (Vonk et al., 2015), and many trace compounds including toxic  
62 mercury (Hg) are tightly linked to OC, both in soils and in river waters (Regnel and Hammar,  
63 2004; Zhang et al., 2015; Kohlenberg et al., 2018; Olson et al., 2018; St. Pierre et al., 2018).  
64 Determination of the river Hg flux to the Arctic Ocean has been a topic of active research, since  
65 it is of similar magnitude as Hg atmospheric deposition in the Arctic Ocean Hg budget (Amos et  
66 al., 2014; Dastoor and Durnford, 2014; Fisher et al., 2012). Sonke et al. (2018) recently  
67 constrained a comprehensive pan-arctic river Hg budget that shows equal particulate Hg (PHg)  
68 and dissolved (DHg) fluxes to the Arctic Ocean of 22 Mg y<sup>-1</sup> each. Particulate organic carbon  
69 (POC) contributes substantially to the total OC export from continents to the ocean (Schlesinger  
70 and Melack, 1981; Lal, 2003; Lamoureux et al., 2014; Galy et al., 2015; Li et al., 2017), and a  
71 two-fold increase of Arctic river POC flux by 2100 has been predicted (Gordeev and  
72 Kravchishina, 2009).

73 Consequently, the anticipated increase in POC flux implies that the riverine export of  
74 PHg to the coastal zone of the Arctic Ocean may also strongly increase. However, the magnitude  
75 of this increase is likely region-specific and depends on a number of environmental parameters  
76 such as permafrost coverage, lithology, and the presence of forest, wetlands and lakes in the  
77 watershed. Understanding how these parameters affect POC and PHg dynamics is critical for  
78 predicting of climate change impacts on Hg transport from the land to the Arctic Ocean.

79 Despite recent efforts in characterizing PHg fluxes in large Arctic Rivers (Coquery et al.,  
80 1995; Amos et al., 2014; Kirk and Louis, 2009; Leitch et al., 2007; Carrie et al., 2012;  
81 Quemerais et al., 2009; Sonke et al., 2018), these studies do not identify the factors controlling  
82 PHg fluxes in high latitude rivers, which depend on the size of the river watershed and its

83 landscape (physio-geographical) parameters. In this regard, large continental plains such as the  
84 western Siberia Lowland (WSL) which contains huge reservoir of frozen and thawed organic  
85 carbon and metals such as Hg (Sheng et al. 2004; Stepanova et al., 2015; Schuster et al., 2018)  
86 may be especially useful in assessing environmental controls on PHg export to the Arctic Ocean.  
87 Because of the vast amounts of frozen peat with an average thickness of 1 to 3 m, extending  
88 over a territory of more than 1 million km<sup>2</sup>, the permafrost thaw in this region can strongly affect  
89 the coastal Arctic system via enhanced export of organic-rich river suspended matter (RSM).  
90 Due to the homogeneity of the WSL landscape, lithology, and topography, one can use the  
91 natural north-south gradient of permafrost zone distribution to assess the direct impact of  
92 permafrost conditions on PHg in river waters. The export of Hg by rivers is known to be  
93 strongly controlled by organic carbon (Schuster et al., 2011; Kirk et al., 2012; Sonke et al.,  
94 2018). In western Siberia, there is a maximum of organic carbon concentration and export  
95 fluxes in the sporadic to isolated permafrost zone (Krickov et al., 2018). This zone corresponds  
96 to the boundary of permafrost occurrence, and the thawing front of frozen peatlands. We  
97 therefore hypothesized a maximum of PHg export by rivers located at the appearance of  
98 permafrost (sporadic to isolated zone).

99 To test this hypothesis and to improve current understanding of magnitude and  
100 seasonality of PHg export, we quantified concentrations of PHg across a 1700 km latitudinal  
101 gradient with special emphasis on permafrost-bearing zones during three main hydrological  
102 periods, the peak of spring flood (early June), the summer base flow (August), and the autumn  
103 high flow before the ice on (October). This latitudinal gradient corresponds to that of  
104 temperature and permafrost coverage and also reflects the decrease in active layer thickness  
105 (ALT) from S to N. We aimed at assessing the effects of latitude (temperature), permafrost  
106 coverage and basic landscape features (proportion of bogs, lakes and forest at the watershed) as  
107 well as the size of the river watershed on PHg concentration, PHg:POC ratio, and PHg run-off

108 and flux. Particular attention has been paid to the impact of the size of the river watershed, given  
109 that the small rivers are strongly understudied in terms of their suspended load (Holmes et al.,  
110 2002). While small rivers allow better assessment of landscape parameters impact on export  
111 fluxes, such rivers are hardly accessible in pristine regions, and at the same time, small rivers are  
112 often anthropogenically contaminated in populated regions. The WSL allows us to overcome  
113 these obstacles due to well-developed road infrastructure in an otherwise pristine region. Finally,  
114 we will use our observations to predict changes in PHg fluxes under climate warming, landscape  
115 changes and progressive permafrost thaw in the largest frozen peatland province in the world.

116

## 117 **2. Study Site and Methods**

### 118 *2.1. Geographical setting*

119 The 32 rivers were sampled in western Siberia Lowland, a large (about 2.6 million km<sup>2</sup>),  
120 peatland and forest zone (**Fig. 1**). The WSL territory encompasses the taiga, forest-tundra and  
121 tundra biomes, the positions of which follow the decrease of mean annual air temperature  
122 (MAAT) from -0.5°C in the south to -9.5°C in the north. The permafrost distribution also  
123 follows the MAAT and changes from absent, isolated and sporadic in the south to discontinuous  
124 and continuous in the north. The active layer thickness (ALT) corresponding to the depth of the  
125 seasonal thaw of peat layers is equal to 55±10 cm in continuous permafrost zone, 70±20 cm in  
126 discontinuous permafrost zone and 160±50 cm in sporadic to isolated zone (Raudina et al.,  
127 2017). Further details of WSL physio-geographical settings, peat and lithological description of  
128 the territory are provided elsewhere (Kremenetski et al., 2003; Stepanova et al., 2015;  
129 Pokrovsky et al., 2015; Raudina et al., 2018) and listed in **Table S1A** of the Supplement. For  
130 each biome (taiga, forest-tundra and tundra), several rivers of different watershed size were  
131 chosen, following the previous strategy of dissolved WSL river load sampling along the  
132 latitudinal transect (Pokrovsky et al., 2015, 2016; Vorobyev et al., 2017). The open water period

133 lasts about 6 months, from May to October. Most small rivers located within the permafrost  
134 zone are frozen during winter period. The 6-months winter discharge is  $\leq 10\%$  of annual water  
135 flow (Pokrovsky et al., 2015).

136

## 137 *2.2. Sampling*

138 Sampling was performed during three main hydrological seasons: spring flood (17 May –  
139 15 June 2016), summer baseflow (1 – 29 August 2016) and autumn baseflow before ice on (24  
140 September – 13 October 2016). During sampling campaigns, we moved from south to north in  
141 spring and from north to south in autumn over 2 to 4-weeks period of time, following the natural  
142 change of seasons. This allowed to sample all rivers of the transect at approximately the same  
143 hydrological stage, after ice off and before ice on. Compared to a 30-year climate record (1985-  
144 2015), the year 2016 was normal for western Siberia in terms of spring, summer and autumn  
145 precipitation but the temperature was 4 and 2.7°C higher than normal in spring and summer,  
146 respectively, and not different from the average temperature in autumn (Rosgidromet, 2017).

147 Ultraclean sampling and handling was used during river suspended matter (RSM)  
148 collection (Krickov et al., 2019). Large volume water samples were taken from the middle of  
149 the river at 0.5 m depth in pre-cleaned polypropylene jars (30 to 50 L) from a PVC boat and  
150 were decanted during 2 to 3 days to allow the particle settling. The jars were pre-cleaned by  
151 detergent, river water, 1 M HCl, MilliQ water, and finally by filling them completely 5 times by  
152 river water before collecting the sample for decantation. Some 10% headspace was left in the  
153 jars and the lids were not tightened to allow aeration. Note that all river waters sampled in this  
154 work were atmospheric oxygen-saturated. As such, the redox conditions in the jars were similar  
155 to those of the river water. The jars were protected from daylight to avoid any phytoplankton  
156 development and photodegradation of DOM, or photoreduction of Hg. The decantation was  
157 done at the river bank, in shadowed areas at the temperature close to that of the river water.

158 During this decantation, the samples were not subjected to biological transformation as  
159 particulate organic C, N and P concentrations remained constant and were found to be identical  
160 to those determined after immediate filtration and freezing of filters with particulate organic  
161 matter (see Krickov et al., 2018 for details). Further, a number of trace metals also demonstrated  
162 reasonable agreement in their concentrations in RSM obtained after 2-3 days of decantation and  
163 direct filtration (Krickov et al., 2019). Nevertheless, some biogeochemical transformation of  
164 PHg bioavailability and speciation during sampling could not be excluded.

165 The water of the bottom layer of the jars (approx. 30% of the initial volume) was  
166 centrifuged on-site during 20 min at 3500 rpm using 4 pre-cleaned 50-mL HDPE Nalgene tubes,  
167 the particles were recovered on the bottom of these tubes, immediately frozen at -18°C and  
168 freeze-dried later in the laboratory. The cleaning of Nalgene tubes for RSM centrifugation,  
169 storage and freeze-drying was performed by rinsing with 1N HCl and MilliQ, in a clean room  
170 class A 10,000. For determination of RSM bulk concentration, smaller volumes of freshly  
171 collected river water (1-2 L) were filtered on-site (at the river bank or in the boat) using pre-  
172 weighed acetate cellulose filters (47 mm, 0.45 µm) and Nalgene 250-mL polystyrene filtration  
173 units using a Mityvac® manual vacuum pump. This method provided the RSM concentration  
174 (mg L<sup>-1</sup>) which was in reasonable (±10-20%) agreement with the value estimated from total  
175 RSM recovery from big jars; see details in Krickov et al. (2018).

176

### 177 2.3. Analyses

178 Total Hg concentration in RSM (PHg<sub>RSM</sub>, ng g<sup>-1</sup>) was determined using a direct mercury  
179 analyzer (DMA-80 - Milestone, Italy). Total mercury concentration was measured by  
180 combustion of the sample at 750 °C, pre-concentration of the Hg<sup>0</sup> vapor released on a gold trap,  
181 and detection by atomic absorption. Analysis of reference materials BCR-482 (lichen, 480 ng g<sup>-1</sup>)  
182 and MESS-3 (sediment, 91 ng g<sup>-1</sup>) showed good recoveries of 450±23 ng g<sup>-1</sup> (n=6, 1σ) and



183 89±6 ng g<sup>-1</sup> (n=7, 1σ) respectively. The average uncertainty on duplicate sample analysis did not  
184 exceed 5% (1σ). Other measured components included total C and N concentration using  
185 catalytic combustion with Cu-O at 900°C with an uncertainty of ≤ 0.5% using Thermo Flash  
186 2000 CN Analyzer at Tomsk University and all major and trace elements. For the latter, the  
187 RSM samples were subjected to full acid digestion with bi-distilled HNO<sub>3</sub> and HF in the clean  
188 room following ICP-MS (Agilent 7500 ce) analyses as described previously for OC-rich natural  
189 samples (Stepanova et al., 2015). The LKSD sediment and 2711a Montana II Soil international  
190 standard reference materials (organic-rich sediment and soil) were processed together with RSM  
191 samples for quality control (see details in Shevchenko et al., 2017). The Hg<sub>RSM</sub> (ng g<sup>-1</sup>) and total  
192 RSM concentration in water (mg L<sup>-1</sup>) allowed calculating PHg concentration in the river water  
193 (PHg<sub>water</sub>, ng L<sup>-1</sup>).

194

#### 195 *2.4. Data treatment*

196 In order to assess the specific impact of the permafrost on PHg concentration, we  
197 separated all the sampled rivers into five categories: permafrost-free (south of 61°N, 6 rivers),  
198 isolated (61 to 63.5°N, 4 rivers); sporadic (63.5 to 65°N, 5 rivers); discontinuous (65 to 66°N, 13  
199 rivers), and continuous permafrost zones (north of 66°N, 4 rivers). Statistical analysis of the  
200 RSM chemistry and fluxes in WSL rivers followed the methodology elaborated for particulate  
201 C, N and P (Krickov et al., 2018). Because of the non-normal distribution of the PHg<sub>RSM</sub> and  
202 PHg<sub>water</sub> concentration data, we use median ± interquartile range (IQR) when reporting  
203 uncertainty. All statistical tests used a significance level of 5% (α = 0.05). The differences in  
204 PHg between different seasons and between adjacent permafrost zones were tested using the  
205 Mann-Whitney U test for paired data at a significance level of 0.05. For unpaired data, the non-  
206 parametric H-criterion Kruskal-Wallis test was performed for all watershed sizes and all  
207 permafrost zones. Further, Spearman rank order correlations (significant at p < 0.05) were

208 performed for assessing the effect of latitude, watershed area ( $S_{\text{watershed}}$ ), percentage of bogs,  
209 forest, lakes at the watershed, the permafrost coverage and bulk concentration of RSM on  
210  $\text{PHg}_{\text{RSM}}$  and  $\text{PHg}_{\text{water}}$ , as well as to characterize the link of PHg with C, N, other major and trace  
211 elements. In order to identify the variable(s) responsible for PHg, constrained principal  
212 component analysis (PCA) was performed.

213 The seasonal  $\text{PHg}_{\text{water}}$  export by rivers was calculated separately for spring (May and  
214 June), summer (July and August) and autumn (September and October) and integrated for each  
215  $2^\circ$  - wide latitudinal belt of the WSL territory, following the hydrological approach developed  
216 for C, major and trace elements (Pokrovsky et al., 2015, 2016). This approach was also used in  
217 the recent estimate of particulate C, N and RSM fluxes of WSL rivers (Krickov et al., 2018).  
218 Given the small number of measurements for each individual river, the combined uncertainties  
219 on these fluxes are estimated as high as  $\pm 30\%$ , which stems mainly from uncertainties on water  
220 discharges (Pokrovsky et al., 2015; Krickov et al., 2018). However, the integral assessment of  
221 riverine export from the WSL territory is robust due to large number of sampled rivers.

222 Note that there is no direct Hg emission by anthropogenic activity in the studied S-N  
223 latitudinal transect of the WSL because there are no industrial centers or coal-fired power plants  
224 in the vicinity (50-100 km) of sampling sites. Low Hg contributions from combustion to  
225 atmospheric particles is consistent with low concentrations of black carbon in snow collected  
226 across the WSL, despite detectable contributions of gas flaring in the northern part of the WSL  
227 (Evangelidou et al., 2018).

228

### 229 **3. Results**

#### 230 *3.1. PHg seasonality and watershed size*

231 Median  $\text{PHg}_{\text{water}}$  concentration in the WSL river waters progressively decreased from the  
232 beginning to the end of open water seasons and were equal to  $1.7 \pm 2.2$ ,  $0.9 \pm 1.5$  and  $0.7 \pm 1.4$  ng L<sup>-1</sup>

233 <sup>1</sup> in spring, summer and autumn, respectively (Table 1). The corresponding Hg<sub>RSM</sub>  
234 concentrations were equal to 237, 124, and 96 ng g<sup>-1</sup>. Particulate Hg concentrations in individual  
235 rivers (**Table S2**) demonstrated that, regardless of watershed area, PHg followed the order  
236 spring > summer > autumn. The effect of watershed size on both PHg<sub>water</sub> and PHg<sub>RSM</sub> was most  
237 pronounced in spring (**Fig. 2A and B**). Although we collected one sample for each river, the  
238 sampling was representative of the riverine particles because: 1) we visited all rivers 3 times per  
239 year; 2) the sample volume was large (30 to 50 L), and 3) the river water chemical composition  
240 in western Siberia is highly stable over full transects of the river channel and on a time scale of 2  
241 to 3 days (Vorobyev et al., 2019). The typical RSM concentrations in WSL rivers range from 3  
242 to 10 mg L<sup>-1</sup> with a median value of 7.5 mg L<sup>-1</sup> (Krickov et al., 2018).

243 Statistical analysis confirmed that PHg<sub>RSM</sub> depended on watershed area (H= 7.72, p =  
244 0.021) in spring whereas both PHg<sub>RSM</sub> and PHg<sub>water</sub> depended on watershed area in autumn  
245 (H=7.13, p = 0.028; H=8.05, p=0.018, respectively) as shown by the H-criterion of the Kruskal-  
246 Wallis test and U-test of Mann-Whitney (**Table S3**). This was also reflected by significant (p <  
247 0.05) R<sub>Spearman</sub> rank order correlations of Hg<sub>RSM</sub> and size of the watershed (-0.49, -0.39 and -0.45  
248 in spring, summer and autumn, respectively, **Table S4**).

### 249 250 *3.2. Role of landscape parameters and permafrost distribution on PHg*

251 The latitude and permafrost coverage positively correlated with PHg<sub>RSM</sub> in summer  
252 (R<sub>Spearman</sub> = 0.48 and 0.51, respectively, **Table S4**). The bog coverage exhibited significant (p <  
253 0.05) control on PHg<sub>RSM</sub> which was detectable in spring (R<sub>Spearman</sub> = 0.56). The effect of forest  
254 and lake coverage of watersheds was not significant ( p < 0.05). There was a peak of both  
255 PHg<sub>water</sub> and PHg<sub>RSM</sub> in rivers of sporadic permafrost zone in spring (**Fig. 3 A, B**) and a less  
256 pronounced maximum of PHg<sub>water</sub> in rivers of discontinuous permafrost zone in summer (**Fig. 3**  
257 **B**). The spring PHg<sub>RSM</sub> concentrations in absent, isolated and sporadic permafrost zones were ca.

258 3-5 times higher compared to those in the discontinuous and continuous permafrost zone in  
259 spring, whereas the highest  $\text{PHg}_{\text{RSM}}$  were observed in discontinuous and continuous permafrost  
260 zones in summer, when  $\text{PHg}_{\text{RSM}}$  was a factor of 2 to 4 higher than in absent, isolated and  
261 sporadic permafrost (**Table 1, Fig. 3 B**). Consistent with that, a non-parametric Kruskal-Wallis  
262 H-test yielded significant differences in  $\text{PHg}_{\text{water}}$  and  $\text{PHg}_{\text{RSM}}$  among permafrost-free and 4  
263 permafrost-bearing zones in spring ( $H=15.8$ ,  $p = 0.0032$ ;  $H=12.3$ ,  $p = 0.016$ , respectively) and  
264 autumn ( $H = 10.2$ ,  $p = 0.038$  for  $\text{PHg}_{\text{water}}$ , **Table S5 A**). The U-test of Mann-Whitney for  
265  $\text{PHg}_{\text{water}}$  and  $\text{PHg}_{\text{RSM}}$  between all adjacent permafrost zones yielded significant differences in  
266 spring and for  $\text{PHg}_{\text{water}}$  in autumn ( $p < 0.04$ , **Table S5 B**). Finally, the Principal Component  
267 Analysis (PCA) demonstrated the presence of 3 factors acting on  $\text{PHg}_{\text{water}}$ ,  $\text{PHg}_{\text{RSM}}$  and the  
268  $\text{PHg}_{\text{flux}}$  (**Fig. S1** of Supplement). The F1 (30% variance) included latitude, MAAT and  
269 permafrost coverage. The F2 (15% variance) comprised the proportion of lakes and forest at the  
270 watersheds, C and N. Finally, the F3 (13%) included discharge and the watershed area.

271

272

### 273 *3.3. Correlations of Hg with C, N and other major and trace elements in RSM*

274 There were significant ( $p < 0.05$ ) positive relationships of  $\text{PHg}_{\text{RSM}}$  with OC and N  
275 ( $R_{\text{Spearman}} = 0.74$  and  $0.71$ , respectively) during spring period. In contrast, negative correlations  
276 between  $\text{PHg}_{\text{RSM}}$  and mineral-originated insoluble elements (Al, Ti, Zr, Th, Nb, Ga, Rare Earth  
277 Elements ( $R_{\text{Spearman}} = -0.60$  to  $-0.74$ ) were observed (**Table S6**). There was no correlation  
278 between  $\text{PHg}_{\text{RSM}}$  and other elements in RSM in summer, whereas in autumn, a number of heavy  
279 metals and aerosol-deposited elements demonstrated significant ( $p < 0.05$ ) positive correlation  
280 with  $\text{PHg}_{\text{RSM}}$  ( $R_{\text{Spearman}} = 0.41$ ,  $0.37$ ,  $0.47$ ,  $0.43$ , and  $0.59$  and  $0.42$  for Pb, Zn, Cu, Sb, and Mo,  
281 respectively).

282 The Hg:C ratio ( $\mu\text{g Hg} : \text{g C}$ ) in the particulate load of WSL rivers (**Table 1**) was  
283 independent of watershed area and generally 3 times higher in spring compared to summer and  
284 autumn (**Fig. 4 A**). The Hg:C ratio was similar across all permafrost zones in spring (**Fig. 4 B**).  
285 In summer and autumn, the Hg:C ratio was significantly ( $p < 0.05$ ) higher in discontinuous and  
286 continuous permafrost zone compared to permafrost-free, isolated and sporadic zones (**Fig. 4 B**).  
287 The average discharge-weighted Hg:C ratio in particles of WSL rivers is equal to  $2.7 \pm 0.5 \mu\text{g g}^{-1}$   
288 which integrates both permafrost-free and permafrost-bearing zones.

289

#### 290 *3.4. PHg export flux from the WSL to the Arctic Ocean*

291 An open-water period (May to October) export flux of  $\text{PHg}_{\text{water}}$  across the WSL territory  
292 demonstrated a significant ( $p < 0.01$ , non-parametric Mann-Witney U-test and H-criterion of  
293 Kruskal-Wallis) maximum at  $61\text{-}62^\circ\text{N}$ , in the sporadic permafrost zone (**Fig. 5 A**). In this  
294 latitudinal belt, the riverine PHg fluxes were 2.3 and 1.9 times higher than those in permafrost-  
295 free and discontinuous/continuous permafrost zones, respectively. The 2-month spring period  
296 provided the maximal contribution to open water PHg fluxes in sporadic permafrost zone,  
297 whereas the contribution of summer and autumn was the highest in discontinuous and  
298 continuous permafrost zone (**Fig. 5 A**). The average PHg export yield from WSL river  
299 watersheds during open water period of the year (6 months) ranged from 0.2 to  $0.5 \text{ g PHg km}^{-2}$   
300 and exhibited a maximum at  $61\text{-}62^\circ\text{N}$ , within the sporadic permafrost zone (**Table S2, Fig. 5 B**).  
301 This zone provided the highest contribution to the overall PHg export flux from permafrost-  
302 affected territory of WSL (56 %, **Fig. 5 C**). Note that the winter period contributes less than 10%  
303 of RSM, POC and PHg fluxes in Arctic Rivers (McClelland et al., 2016; Sonke et al., 2018). The  
304 mean annual air temperature (MAAT), which could be constrained for each permafrost zone but  
305 not for individual rivers, shows a maximum in PHg export yield by WSL rivers that corresponds  
306 to the sporadic permafrost zone (**Fig. 5 D**). Note that the observed maximum in PHg export was

307 not linked to latitudinal variation in river runoff. The latter remained fairly constant over full  
308 range of studied sites and independent on latitude ( $R^2 = 0.08$ , **Fig. 5 E**).

309

310

## 311 **4. Discussion**

### 312 *4.1. The influence of watershed size on PHg concentration*

313 The WSL river  $\text{PHg}_{\text{water}}$  concentrations (typically 0.5 to 5.0  $\text{ng L}^{-1}$ ) are similar to other Arctic  
314 rivers of low runoff which drain through boreal zone such as Ob (7.5  $\text{ng L}^{-1}$ , Coquery et al.,  
315 1995), Severnaya Dvina (4 to 11  $\text{ng L}^{-1}$ , Sonke et al., 2018), Yenisey (0.8 to 20  $\text{ng L}^{-1}$ , Sonke et  
316 al., 2018) and N. American rivers (0.4 to 3.7  $\text{ng L}^{-1}$ , Schuster et al., 2011). The  $\text{PHg}_{\text{RSM}}$  values of  
317 the WSL rivers (100 to 1000  $\text{ng g}^{-1}$ ) are in agreement with observations on the Yenisey (60 to  
318 600  $\text{ng g}^{-1}$ , Sonke et al., 2018) but higher than N. American rivers (10 to 40  $\text{ng g}^{-1}$ , Schuster et  
319 al., 2011). A possible reason for high  $\text{PHg}_{\text{RSM}}$  in WSL rivers is high POC concentration,  
320 reflecting the dominance of peatlands in Siberian watersheds.

321 Indeed, small WSL streams ( $S_{\text{watershed}} < 1,000 \text{ km}^2$ ) exhibited significantly higher  
322 concentrations of  $\text{PHg}_{\text{RSM}}$  compared to larger rivers (**Fig. 2**). The small streams of WSL have the  
323 highest POC concentration in the RSM (Krickov et al., 2018). This suggests enhanced mobilization  
324 of PHg in the form of peat debris. Consistently, histosols, the dominant ( $\geq 50\%$ ) soil group of  
325 WSL, can provide large amounts of organic particles, given the high susceptibility of peat to  
326 physical disintegration compared to mineral sediments (Rezanezhad et al., 2016). The  
327 enrichment of the river water in OC-rich particles may occur both at the river banks (especially  
328 in small rivers flowing through the wetlands) as well as within extensive floodplains via  
329 remobilization of organic-rich sediments at high flow (Krickov et al., 2018; Vorobyev et al.,  
330 2019). The floodplains of the Ob river and other rivers of the WSL extend more than 10 times  
331 the width of the main channel (Vorobyev et al., 2015). Small rivers also exhibited the largest

332 scatter in  $\text{PHg}_{\text{RSM}}$  concentrations, probably due to the very short transit time in the watershed,  
333 leading to a fast response of river particulate load to minor variations in surface hydrology such  
334 as local storm events (e.g., Jeong et al., 2012; Lee et al., 2016).

335 It is also possible that autochthonous processes of suspended matter transformation in the  
336 river channel such as photo- (Mayer et al., 2006; Riggsbee et al., 2008) or bio-degradation  
337 (Richardson et al., 2013; Gladyshev et al., 2015; Attermeyer et al., 2018) of particulate organic  
338 matter (POM) are capable of modifying  $\text{PHg}_{\text{RSM}}$  to a greater extent in large rivers relative to small  
339 streams. The large rivers of WSL have longer water residence times compared to small rivers (Ala-  
340 aho et al., 2018a, b). We speculate that, similar to WSL river  $\text{CO}_2$  emission (Serikova et al., 2018),  
341 the longer travel time of RSM in large rivers may decrease the PHg concentration by biotic or  
342 abiotic PHg reduction and evasion of gaseous  $\text{Hg}^0$  loss (Gårdfeldt et al., 2001; Poissant et al.,  
343 2004; Osterwalder et al., 2017). Overall, the WSL river transport of PHg may be strongly  
344 controlled by POM quality and origin, which emphasizes the need to provide molecular-level  
345 characterization of POM across permafrost and river-size gradients (McClelland et al., 2016;  
346 Kutscher et al., 2017).

347 The overwhelming role of POM from peat soils as the main factor supplying PHg to  
348 rivers is further confirmed by geochemical analyses of RSM and the links of Hg with other  
349 elements in the suspended load. Observed negative correlations of  $\text{PHg}_{\text{RSM}}$  with inorganic  
350 components of RSM reflect that Hg is bound to organic matter (N, C,  $R_{\text{Spearman}} = 0.79$  and  $0.76$ ,)  
351 and not to the mineral fraction of RSM bearing insoluble (Ti, Al, Zr, REE) silicates, nor to  
352 partially soluble carbonate particles containing B, Mg, Ca, Sr, B and U. Therefore, during  
353 spring, the mineral fraction of RSM decreases  $\text{PHg}_{\text{RSM}}$  by mere dilution with solid mineral  
354 matter. This was especially visible in large rivers, having a higher proportion of silicate minerals  
355 in their RSM compared to OC-dominated RSM of small rivers. In autumn,  $\text{PHg}_{\text{RSM}}$  was

356 positively correlated with Pb, Zn, Cu, Sb, and Mo which could reflect co-deposition from the  
357 atmospheric dust to vegetation and soils (Shevchenko et al., 2017).

358

359

360

#### 361 *4.2. Impact of permafrost coverage on PHg*

362 Climate and permafrost coverage are considered as main drivers of dissolved and  
363 particulate river load in the WSL (Frey et al., 2007; Frey and Smith, 2005; Pokrovsky et al.,  
364 2015). Due to very flat topography of this region, the permafrost distribution in western Siberia  
365 roughly follows the latitude and MAAT so that there is a progressive change of the permafrost  
366 zones (“absent → isolated → sporadic → discontinuous → continuous”) in the south → north  
367 direction. Both  $PHg_{water}$  and  $PHg_{RSM}$  demonstrated a maximum around 60-62°N, in the sporadic  
368 and discontinuous permafrost zones (**Fig. 3**). The May-October PHg export fluxes were also  
369 maximal in this part of the WSL (**Fig. 5 B, C**). Note that spatial patterns of PHg fluxes in  
370 western Siberia cannot be explained by latitudinal variation of precipitation or runoff, because  
371 the water runoff remains constant or gradually increases northward (Pokrovsky et al., 2015 and  
372 **Fig. 5 E**).

373 The discontinuous permafrost zone can be considered as a large-scale thawing front of  
374 the frozen peat, and corresponds to the southern boundary of permafrost persistence. Here, an  
375 enhanced mobilization of C- and N-rich particles from soils to rivers (Krickov et al., 2018) and  
376 maximal CO<sub>2</sub> emission from rivers (Serikova et al., 2018) occur. Further, a maximum of  
377  $PHg_{RSM}$  and export fluxes occurred in isolated and sporadic permafrost in spring. This  
378 maximum shifted to discontinuous permafrost zone in summer and moved northward, to the  
379 discontinuous permafrost zone in autumn (**Fig. 3 B and 5 A**, respectively).



380 The active (unfrozen) layer thickness increases from spring to autumn and more  
381 importantly it moves northward during the open –water period from May to October (Trofimova  
382 and Balybina, 2014; Raudina et al., 2017). This increase controls the degree of peat and mineral  
383 particles eroding from the soil profile to the rivers (e.g., Krickov et al., 2018). Thus, enhanced  
384 mobilization and subsequent river inputs of C, nutrients and metals at the “hot spot” of the  
385 permafrost thawing front was reported at the local scale of frozen peat landscapes of western  
386 Siberia (Loiko et al., 2017).

387 The 2 to 3 times higher Hg:C ratio in spring compared to summer and autumn likely reflects  
388 the run-off of snow Hg that accumulated by atmospheric deposition over the previous 6 months  
389 cold season. The Hg:C ratio in RSM during summer and autumn was 2 to 3 times higher in  
390 northern rivers of discontinuous and continuous permafrost zone compared to southern rivers (**Fig.**  
391 **4 B**). Note that during this period, the water travel times in the WSL rivers and from the soil profile  
392 to the river are much shorter in the north than in the south (Ala-aho et al., 2018b).

393 We hypothesize that the breakdown of river POM to CO<sub>2</sub> (Serikova et al., 2018; Krickov et  
394 al., 2018) leads to concomitant reduction and volatilization of PHg. This process is then more  
395 pronounced in the southern rivers with longer water residence time, while in the north, more  
396 refractory organic matter of RSM (Gladyshev et al., 2015) and lower biological activity  
397 (Kawahigashi et al., 2004) lead to better preservation of POM-bound Hg in riverine particles. This  
398 may produce the observed Hg to C enrichment in the RSM of northern rivers during the summer  
399 and fall base flow period.

400 Overall, PHg generation in WSL rivers across the permafrost gradient may depend on  
401 both the PHg supply and losses in the catchments. Atmospheric Hg retention in continental  
402 ecosystems is thought to be dominated by the plant Hg pump during the vegetation growth  
403 season, where vegetation directly sequesters gaseous elemental Hg<sup>0</sup> from the atmosphere (Obrist  
404 et al., 2017; Jiskra et al., 2018). During the winter season Hg deposition is likely dominated by

405 wet deposition in the form of snowfall (Nelson et al., 2010). The main sources of suspended  
406 matter in WSL rivers are 1) vegetation litter such as plant leaves in the south and mosses, lichens  
407 and dwarf shrub leafs in the permafrost-bearing zone; these organic debris are easily mobilized  
408 by surface run-off to the river, notably in spring during snow melt; 2) surface and deep peat and  
409 mineral horizons, providing the particles via bank abrasion in spring and via supra-permafrost  
410 flow in summer and autumn, and 3) lake and bog open water sediments as well as flood plain  
411 sediments. In the north, soil erosion is limited by the low thickness (30-40 cm) of the active  
412 (unfrozen) layer and the low net primary productivity, providing POM, whereas in the south, the  
413 soil is stabilized by abundant forest vegetation. We suggest that in the north, shallow unfrozen  
414 peat depth and lower biomass accumulation cannot supply sufficiently high PHg and the fluxes  
415 remain relatively low in spring. In contrast, the PHg concentration and fluxes greatly increase in  
416 summer, when the thickness of active (unfrozen) peat layer increases in the north. As such,  
417 physical erosion of peat soils in the WSL drives maximum RSM levels at the sporadic  
418 permafrost zone due to maximal thickness of unfrozen histosols, notably in the beginning of the  
419 snow-free season.

420

#### 421 *4.3. PHg flux and climate warming in western Siberia*

422 The Taz and Pur Rivers ( $S_{\text{watershed}} = 150,000$  and  $112,000$  km<sup>2</sup>) integrate a significant part  
423 of permafrost-affected WSL territory, and export ca. 0.036 and 0.0067 Mg PHg y<sup>-1</sup> (Table 1).  
424 The PHg flux of these two rivers represents less than 0.4 % of the overall Eurasian River ( $S_{\text{total}} =$   
425  $10,962,000$  km<sup>2</sup>) flux (11.3 Mg PHg y<sup>-1</sup>, Sonke et al., 2018) which is disproportionally (6.5  
426 times) low compared to their total watershed area and respective discharge. The most likely  
427 cause for this is low concentration and low annual yield of RSM in WSL rivers due to low water  
428 runoff (100-200 mm y<sup>-1</sup>) and limited river slopes. Measured open-water period PHg export

429 yields of Ob, Pur and Taz (0.20, 0.06, and 0.24 g km<sup>-2</sup> y<sup>-1</sup>, respectively, **Table S2**) are lower than  
430 the modeled yields by Sonke et al. (2018) (0.63, 1.5 and 1.7 g km<sup>-2</sup> y<sup>-1</sup>, respectively).

431 It is possible that the individual PHg fluxes of WSL rivers, assessed in this study, are  
432 biased by small number of sampling days, and poor constraining of water discharges for these  
433 essentially ungauged rivers. However, the Hg:C ratio, which is based on a large number of  
434 measurements, may become an efficient proxy for reconstructing overall Hg export based on  
435 known POC export. Indeed, the measured PHg and POC fluxes of Arctic Rivers (Leitch et al.,  
436 2007; Kirk et al., 2009; Carrie et al., 2012; Sonke et al., 2018; McClelland et al., 2016) yield  
437 ranges of Hg:C (Mg:Tg) ratios equal to 2.0-4.8 (MacKenzie), 1.3-2.8 (Sev. Dvina), and 2.6-7.8  
438 (Yenisey). This is consistent with the discharge-weighted Hg:C ratio of 2.7±0.5 recommended  
439 for WSL rivers in this study. Further, using the mean ratio Hg:C of open water time period in  
440 rivers of WSL, and overall POC flux of the pan-Arctic rivers (5.77 Tg y<sup>-1</sup>, McClelland et al.,  
441 2016), the pan-Arctic flux of PHg is equal to 15.6±3.0 Mg y<sup>-1</sup> which is comparable to the value  
442 21.9 Mg y<sup>-1</sup>, assessed by Sonke et al (2018). For the Ob River with its 0.57 Tg POC y<sup>-1</sup> flux  
443 (McClelland et al., 2016), the observation-based PHg flux is 1.5±0.3 Mg PHg y<sup>-1</sup> in this study, in  
444 agreement with 1.4 Mg PHg y<sup>-1</sup> modeled by Sonke et al. (2018) based on Hg-DOC and Hg-  
445 discharge relationships.

446 Our results allow a first order assessment of the consequences of climate warming and  
447 permafrost thaw in western Siberia on riverine transport of PHg from the land to the Arctic  
448 Ocean. For this, we can employ a “substituting space for time” approach which was successfully  
449 used in western Siberia for the dissolved (< 0.45 μm) fraction of inorganic river loads (Frey et  
450 al., 2007), DOC (Frey and Smith, 2005; Pokrovsky et al., 2015), trace metals (Pokrovsky et al.,  
451 2016), nutrients (Vorobyev et al., 2017), as well as CO<sub>2</sub> emission scenarios (Serikova et al.,  
452 2018). In a broader context, the “space-for-time” substitution approach postulates that

453 contemporary spatial phenomena can be used to understand and model temporal processes that  
454 are otherwise unobservable, such as past and future events (Blois et al., 2013).

455 In the northern part of the WSL, soil temperature may rise by 0.15 to 0.3 degree per 10  
456 years over next 50 to 100 years (Pavlov et al., 2009; Anisimov et al., 2012). On a short-term  
457 prospective (10-50 years), this will transform the discontinuous and continuous permafrost zones  
458 into sporadic and isolated permafrost (Anisimov and Reneva, 2006). Given the current  
459 maximum of PHg at the sporadic PF zone, this may enhance both concentrations and fluxes of  
460 PHg in the northern rivers, roughly by a factor of 2 (see **Figs 3 and 5B**), which will also enhance  
461 the export of PHg to the Arctic Ocean. On a longer prospective (50-150 years), the  
462 discontinuous and continuous permafrost may disappear (Romanovsky et al., 2008; Nadyozhina  
463 et al., 2008) and the border of isolated to sporadic permafrost development will follow the Arctic  
464 circle (Bedritsky et al., 2018). Moreover, if the current rate of soil warming persists, by 2100,  
465 the active layer thickness may reach 4 m in the tundra of Yamal Peninsula which is currently  
466 continuous permafrost zone (Sudakov et al., 2011). These changes will certainly produce much  
467 deeper water paths in organic and mineral soils. Progressive moving of the permafrost thawing  
468 front to the north will therefore locally and temporarily increase PHg concentration and fluxes at  
469 the sites of maximal ALT. However, complete disappearance of permafrost in the north of WSL  
470 may bring about a decrease in PHg export by northern rivers to the values which are currently  
471 observed in rivers south of 60°N. Furthermore, if the permafrost-free zone expands northward  
472 into the current sporadic zone, this could produce a concomitant reduction in PHg fluxes thus  
473 compensating for the shift northwards of the sporadic zone.

474

## 475 **Conclusions**

476 The concentration of particulate Hg (PHg) in the WSL rivers is controlled by the organic  
477 nature of abundant peat soils that dominate river watersheds. The overwhelming role of deep

478 and intermediate peat horizons rather than mineral layers, surface peat and plant litter in  
479 providing Hg to riverine suspended load is confirmed by *i*) strong positive correlation between  
480 C, N and Hg in RSM, and *ii*) much higher  $\text{PHg}_{\text{RSM}}$  and  $\text{PHg}_{\text{water}}$  in discontinuous and sporadic  
481 permafrost rivers compared to southern, permafrost-free or northern, continuous permafrost-  
482 bearing rivers.

483 The concentration of Hg in RSM decreased with the increase in river watershed size,  
484 illustrating the importance of organic particles in small rivers which drain peatlands and the  
485 abundance of mineral matter from bank abrasion in larger rivers. This was most visible in  
486 spring, when the bank abrasion and sediment transport and deposition within the floodplains  
487 were pronounced. Overall, the spring flood provided the largest contribution to overall PHg  
488 export by WSL rivers to the Arctic Ocean, despite its relatively short duration (2 of 6 months of  
489 open-water period).

490 At the sporadic permafrost zone, we observed maximum concentration and export fluxes  
491 of PHg, which corresponded to peak of particulate C and N concentration and fluxes across the  
492 latitudinal profile. This suggests the enhanced generation of Hg, C-rich RSM at the thawing  
493 front of permafrost, where the thickness of the active layer is maximal.

494 To assess a northward permafrost boundary shifts with increases in air and soil  
495 temperature we used a substituting space for time scenario of climate warming in the WSL, well  
496 developed for the dissolved fraction of C and metals. From a short-term (10-50 years) climate  
497 warming perspective, the effect of a simple northward shift of permafrost boundary may produce  
498 a two-fold increase in PHg concentration and fluxes in rivers of the discontinuous and  
499 continuous permafrost zone. On longer time scales (50-100 years), full disappearance of  
500 permafrost in the northern part of WSL may decrease the concentrations of PHg in the river  
501 water and its export fluxes back to their contemporary level.

502

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510

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512

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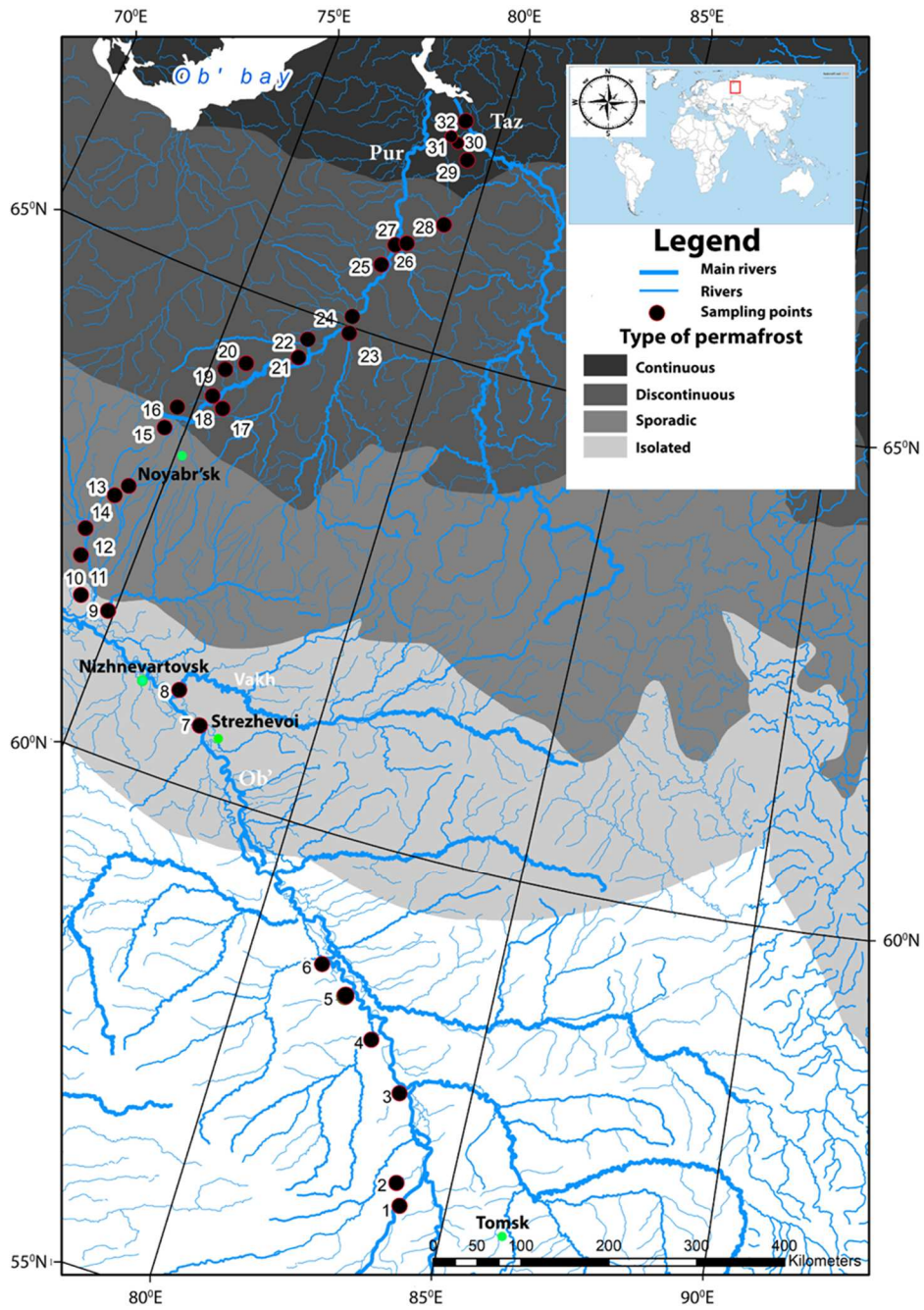
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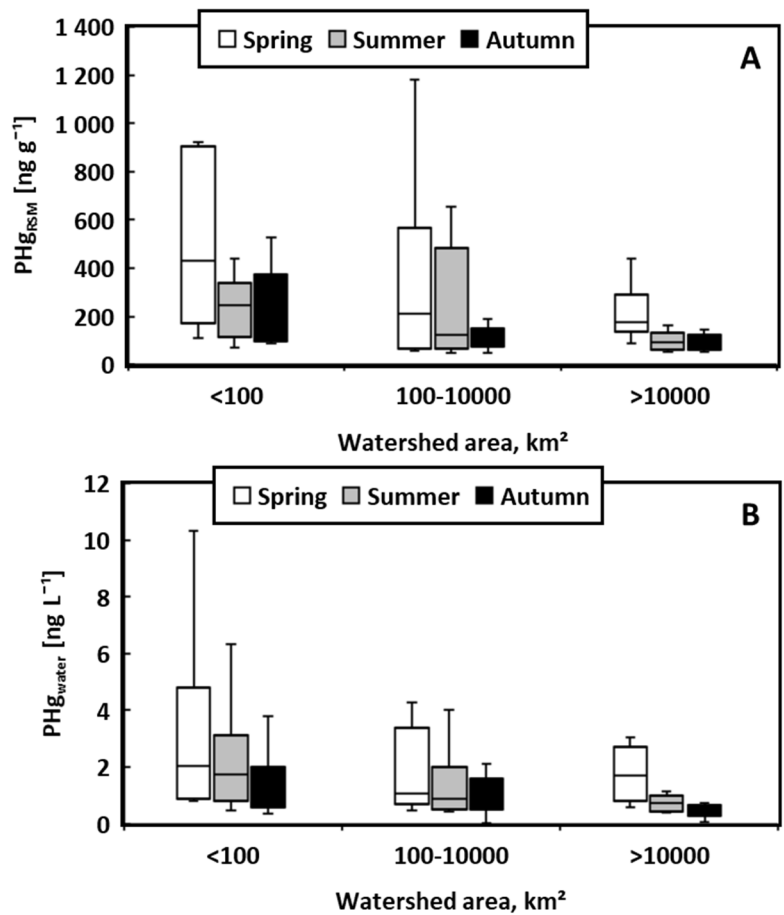
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 796 **Fig. 1.** Sampling sites and physico-geographical context of WSL territory investigated in this  
 797 work.



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800 **Fig. 2.** Particulate (> 0.45 μm) Hg concentration (median ± IQR) in the river suspended matter  
 801 (PHg<sub>RSM</sub>, A) and riverwater (PHg<sub>water</sub>, B) as a function of river watershed size.

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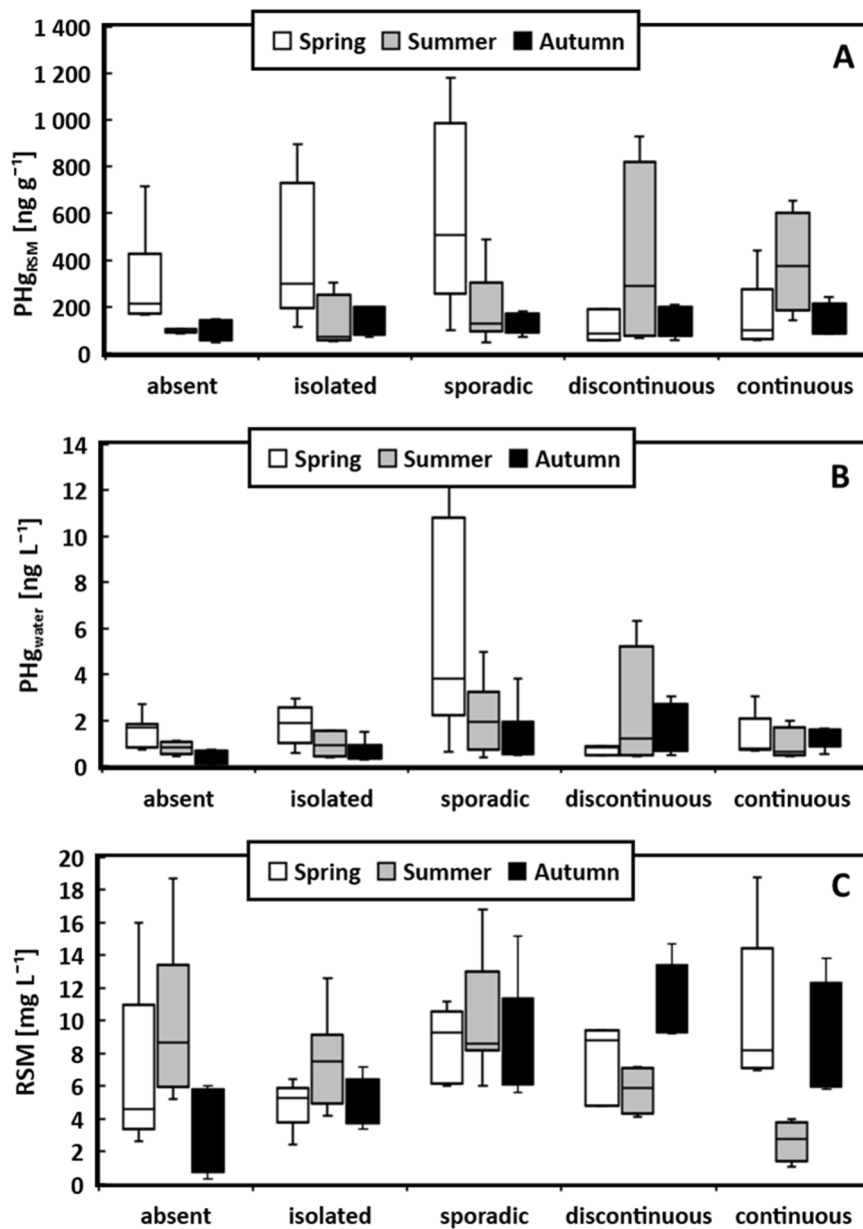
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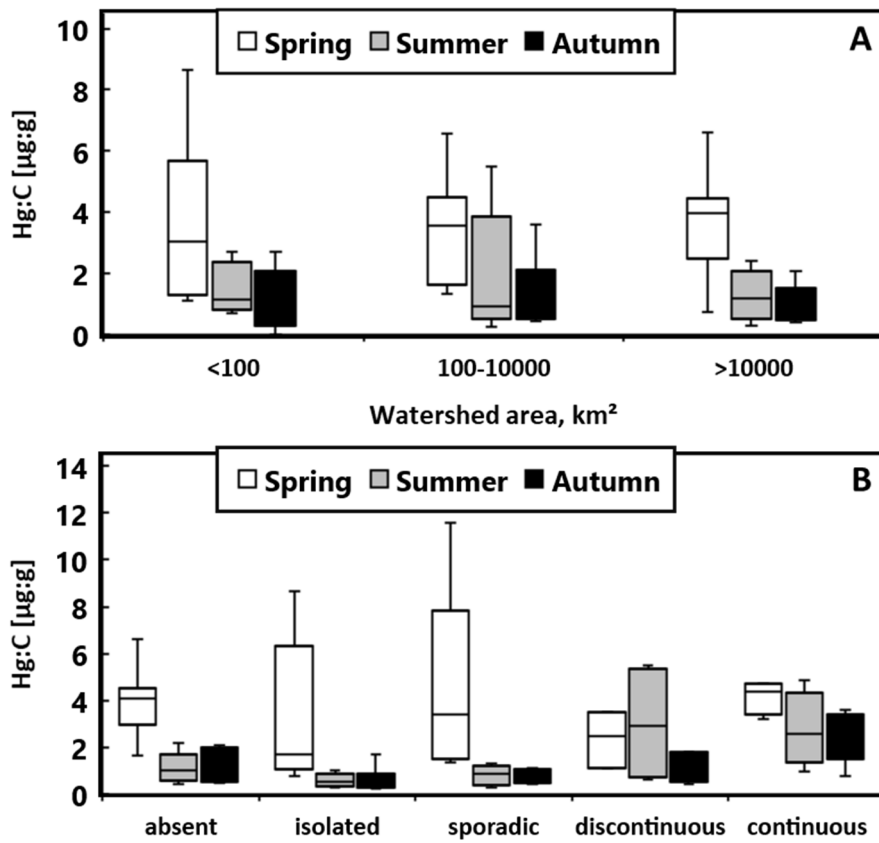
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 817 **Fig. 3.** Box plot of median and first and third quartiles (25 and 75%) of PHg<sub>RSM</sub> (A) and  
 818 PHg<sub>water</sub> (B) in five permafrost zones over three seasons. A disconnect between PHg<sub>RSM</sub> and  
 819 PHg<sub>water</sub> in discontinuous and continuous permafrost regions in summer is due to lower bulk  
 820 RSM concentration in northern rivers during this period of the year (C).

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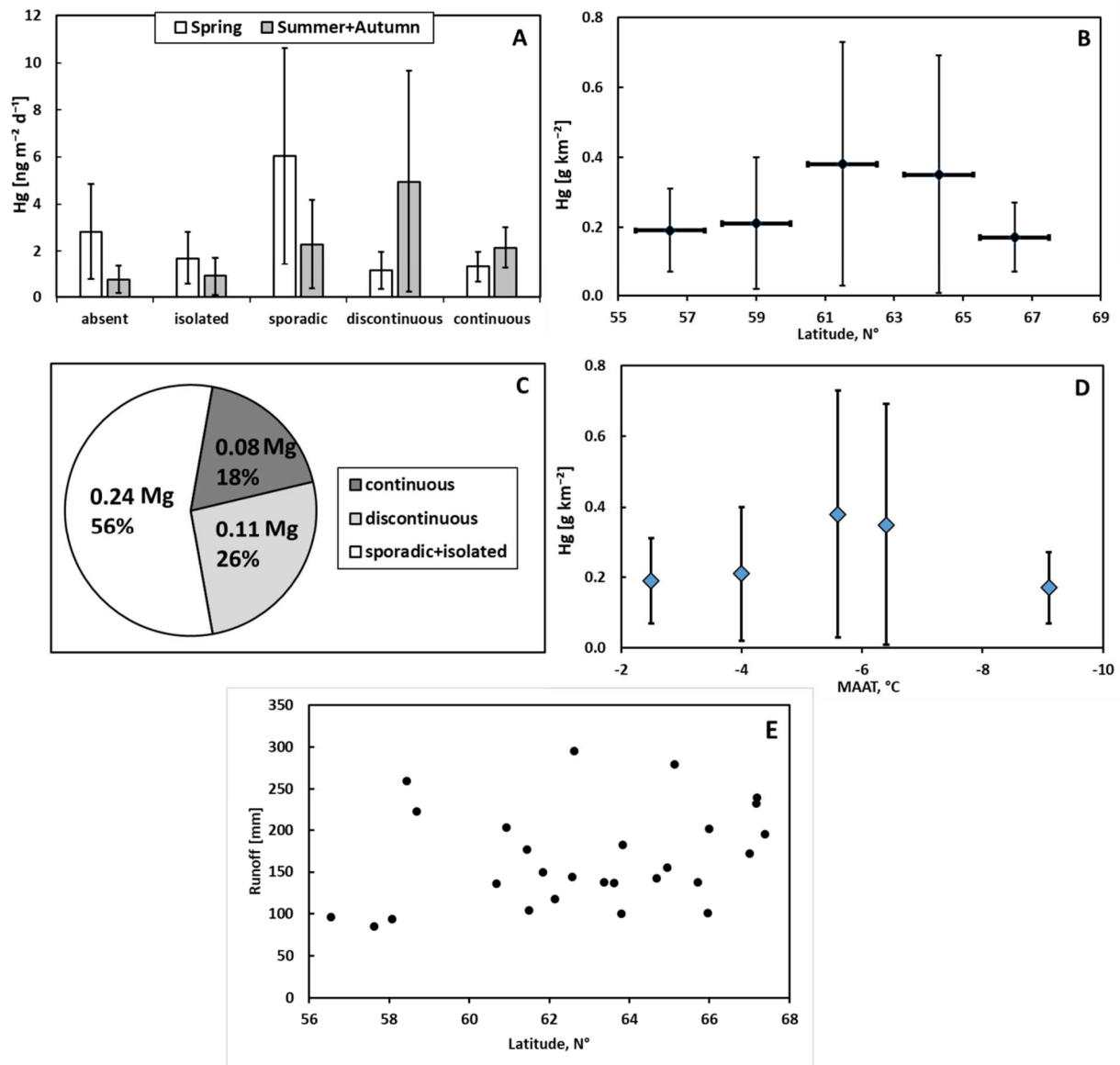
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830 **Fig. 4.** The mass ratio Hg:C ( $\mu\text{g g}^{-1}$ ) for rivers and streams of different size (A) and type of

831 permafrost distribution (B).

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 835 **Fig. 5.** Watershed surface-normalized  $\text{PHg}_{\text{water}}$  export yields during high flow (spring, May and  
 836 isolated, sporadic, discontinuous, continuous) and low flow (4 months, July to October) in 5 permafrost zones (A) the total open-water  
 837 period  $\text{PHg}$  flux (6 months, May to October) across a 2-degree latitude grid (B). C, The partial  
 838 contribution and  $\text{PHg}$  export in Mg of three main permafrost zones of the WSL during 6 months  
 839 of open water period. D, Relationship between the yield of  $\text{PHg}$  by WSL rivers and the MAAT  
 840 (mean annual air temperature) of the watershed (binned to 5 permafrost zones). The error bars in  
 841 A, B and D represent  $\pm 1$  S.D. Note that during open water period (May to October), the river  
 842 runoff from the WSL territory is maximal in spring and independent on latitude (E).

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846 **Table 1.** Median ( $\pm$  IQR) and mean ( $\pm$  SD) values of PHg concentration in water (PHg<sub>water</sub>, ng  
847 L<sup>-1</sup>) and in RSM (PHg<sub>RSM</sub>, ng g<sup>-1</sup>) as well as the ratio of Hg:C in RSM ( $\mu$ g g<sup>-1</sup>) for 5 permafrost  
848 zones and 3 seasons across the WSL transect. For the permafrost-affected zone of WSL, we  
849 recommend annual mean Hg concentration equals to 1.8 (median 1.2) ng Hg L<sup>-1</sup> in water and  
850 246 (median 139) ng Hg g<sup>-1</sup> in RSM. The discharge-weighted Hg:C ratio is estimated as 2.7 $\pm$ 0.5  
851  $\mu$ g g<sup>-1</sup>.

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Season	Variable	Permafrost free	Permafrost			
		Absent	Isolated	Sporadic	Discontinuous	Continuous
Spring	PHg <sub>water</sub> , ng L <sup>-1</sup>	<u>1.7<math>\pm</math>1.0</u>	<u>1.9<math>\pm</math>0.7</u>	<u>3.8<math>\pm</math>7.6</u>	<u>0.8<math>\pm</math>0.4</u>	<u>0.8<math>\pm</math>0.3</u>
		1.6 $\pm$ 0.7	1.8 $\pm$ 0.9	5.6 $\pm$ 4.6	0.7 $\pm$ 0.2	1.3 $\pm$ 1.0
Spring	PHg <sub>RSM</sub> , ng g <sup>-1</sup>	<u>211<math>\pm</math>257</u>	<u>299<math>\pm</math>290</u>	<u>509<math>\pm</math>617</u>	<u>86<math>\pm</math>134</u>	<u>97<math>\pm</math>42</u>
		304 $\pm$ 204	430 $\pm$ 308	588 $\pm$ 400	110 $\pm$ 70	155 $\pm$ 161
Spring	Hg:C, $\mu$ g g <sup>-1</sup>	<u>4.1<math>\pm</math>1.5</u>	<u>2.7<math>\pm</math>5.2</u>	<u>2.5<math>\pm</math>5.0</u>	<u>2.5<math>\pm</math>2.4</u>	<u>4.4<math>\pm</math>1.1</u>
		4.0 $\pm$ 1.5	3.3 $\pm$ 3.2	4.6 $\pm$ 3.9	2.4 $\pm$ 1.2	4.1 $\pm$ 0.7
Summer	PHg <sub>water</sub> , ng L <sup>-1</sup>	<u>0.8<math>\pm</math>0.4</u>	<u>0.7<math>\pm</math>0.8</u>	<u>1.7<math>\pm</math>1.4</u>	<u>1.2<math>\pm</math>3.7</u>	<u>0.7<math>\pm</math>0.8</u>
		1.0 $\pm$ 0.8	1.0 $\pm$ 0.6	2.0 $\pm$ 1.6	2.3 $\pm$ 2.8	0.9 $\pm$ 0.7
Summer	PHg <sub>RSM</sub> , ng g <sup>-1</sup>	<u>94<math>\pm</math>11</u>	<u>65<math>\pm</math>79</u>	<u>159<math>\pm</math>178</u>	<u>288<math>\pm</math>626</u>	<u>372<math>\pm</math>323</u>
		99 $\pm$ 28	137 $\pm$ 110	197 $\pm$ 142	394 $\pm$ 405	385 $\pm$ 216
Summer	Hg:C, $\mu$ g g <sup>-1</sup>	<u>1.1<math>\pm</math>1.0</u>	<u>0.6<math>\pm</math>0.2</u>	<u>0.9<math>\pm</math>0.7</u>	<u>2.9<math>\pm</math>4.3</u>	<u>2.6<math>\pm</math>2.0</u>
		1.1 $\pm$ 0.6	0.6 $\pm$ 0.3	1.1 $\pm$ 1.0	3.0 $\pm$ 2.5	2.7 $\pm$ 1.6
Autumn	PHg <sub>water</sub> , ng L <sup>-1</sup>	<u>0.3<math>\pm</math>0.5</u>	<u>0.6<math>\pm</math>0.4</u>	<u>0.8<math>\pm</math>0.9</u>	<u>1.4<math>\pm</math>1.4</u>	<u>1.4<math>\pm</math>0.3</u>
		0.5 $\pm$ 1.3	0.7 $\pm$ 0.4	1.4 $\pm$ 1.1	1.6 $\pm$ 1.1	0.2 $\pm$ 0.4
Autumn	PHg <sub>RSM</sub> , ng g <sup>-1</sup>	<u>151<math>\pm</math>118</u>	<u>93<math>\pm</math>33</u>	<u>142<math>\pm</math>124</u>	<u>151<math>\pm</math>91</u>	<u>154<math>\pm</math>101</u>
		151 $\pm$ 157	152 $\pm$ 144	148 $\pm$ 89	141 $\pm$ 65	151 $\pm$ 67
Autumn	Hg:C, $\mu$ g g <sup>-1</sup>	<u>1.3<math>\pm</math>1.3</u>	<u>0.5<math>\pm</math>0.2</u>	<u>0.5<math>\pm</math>0.4</u>	<u>1.3<math>\pm</math>1.0</u>	<u>2.4<math>\pm</math>0.9</u>
		1.3 $\pm$ 0.7	0.6 $\pm$ 0.5	0.9 $\pm$ 0.7	1.2 $\pm$ 0.6	2.4 $\pm$ 1.1

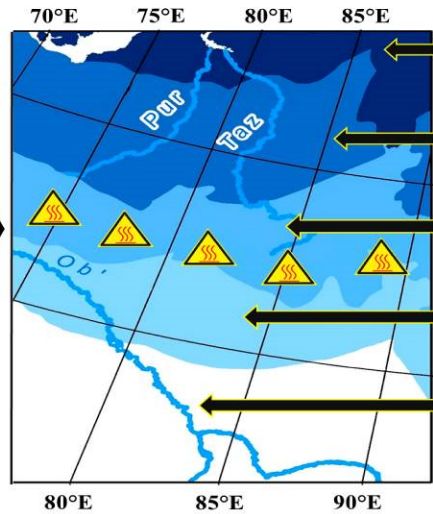
853 Footnote : the median values are given in the numerator and the mean values are given in the denominator.

854

855

# Particulate Hg in Siberian rivers

Permafrost  
thawing  
front



## Seasonal flux

■ Spring  
■ Summer + Autumn

