Long-term data reveals the importance of hydraulic load and inflow water quality for Sb removal in boreal treatment peatlands

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Abstract

Antimony (Sb) is a common contaminant in natural peatlands used as treatment wetlands for water influenced by metal mining in cold-climate regions. However, while other metalloids such as arsenic have been well studied, little is known about removal and retention of Sb in northern wetlands under challenging environmental conditions. In this study we assessed short-term, long-term and seasonal variations in mobility, removal, and retention of Sb from mining-influenced water in two peat based natural wetlands with different loading and physical conditions. Analyses based on 10 years of water quality data and data on contaminant accumulation in the peat soil revealed that the wetland with significantly lower hydraulic load and Sb areal load achieved adequate Sb removal, but with a slight decline in recent years. Antimony concentrations at the wetland outlet were slightly lower in summers than in winters. Dilution due to high rainfall during summer may be the likely reason for low outlet concentrations towards the end of summer. Outlet Sb concentrations were on the rise after inlet water quality was significantly improved through enhanced pre-treatment indicating mobilization of accumulated Sb. In comparison, the smaller wetland with higher hydraulic and Sb loading had very low Sb removal and a stronger decrease in Sb concentration through dilution due to snowmelt. The results highlight the challenges in Sb retention which should get more attention when treatment wetlands are designed for Sb rich waters such as mine waters and there are changes in water treatment arrangement in specific cases.

Keywords: *Antimony; Mine water; Peatland; Long-term data; Arctic region*

1. Introduction

Antimony (Sb) in the environment can be traced back to natural processes and anthropogenic activities. Natural sources of Sb include rock weathering and runoff, while anthropogenic sources include smelting plants, municipal waste incineration, abrasion of brake linings and tires, and fuel combustion in vehicles and power plants (Ainsworth et al., 1990; Bosco et al., 2005; Iijima et al., 2009; Watanabe et al., 1999). However, concentrations in soil, sediments and waters resulting from natural processes are usually far lower than those ensuing from anthropogenic activities (Filella et al., 2002). Antimony can be one of the most common contaminants in waters influenced by metal mining activities, particularly extraction and processing of sulfide ores such as pyrite and galena. It is also a contaminant of concern due to its severe health effects and eco-toxicity in aquatic and terrestrial environments (Borgmann et al., 2005; Cooper and Harrison, 2009; Dovick et al., 2016). As early as the 1970's, it was already included in the list of priority pollutants and dangerous substances by the US Environmental Protection Agency and the Council of European Communities (CEC, 1976; USEPA, 1979). Antimony typically accumulates in plants without showing signs of toxicity although the amounts accumulated may be toxic for humans and animals consuming the plants (Tschan et al., 2009). The toxicity of Sb depends on its oxidation state (-III, 0, III, V) and the type of compounds in which it is present i.e. organic (such as methylated compounds) or inorganic (such as $Sb(OH)_3$ and $Sb(OH)_6^-$), all of which exist in the environment (Guo et al., 2018; Herath et al., 2017; Koch et al., 2000). Inorganic Sb species are generally regarded as more toxic than organic species, and Sb(III) compounds are considered more harmful and mobile than Sb(V) compounds (Gebel, 1997; He and Yang, 1999; Wilson et al., 2010).

Interest in the geochemical behavior of Sb in the environment has developed fairly recently, whereas much research has been performed on other similar elements e.g. arsenic (As), another

Group 15 element, which usually coexists with Sb in nature and has analogous chemical and toxic properties (Guo et al., 2018; Herath et al., 2017; Lehr et al., 2007; Ungureanu et al., 2015). Despite the similarities, the biogeochemical behavior of Sb and As differ e.g. in terms of mobility, reaction rates, and bioavailability (Fu et al., 2016; Leuz et al., 2006; Tschan et al., 2009). Available information on mobilization of Sb in the environment and its transformation through the geosphere and hydrosphere is still limited (Herath et al., 2017). Various active treatment methods have been reported for Sb removal from water and wastewater including ion-exchange (Dzul Erosa and Höll, 2006; Parker et al., 1979), adsorption (Shan et al., 2014; Wang et al., 2019; Xu et al., 2011), coagulation-flocculation-sedimentation/filtration (Du et al., 2014; Guo et al., 2009; Kang et al., 2003), co-precipitation (Gannon and Wilson, 1986; Wang et al., 2013), membrane filtration (Kang et al., 2000; Pawlak et al., 2006), electrochemical methods (Koparal et al., 2004; Zhu et al., 2011), etc. At the same time, passive treatment methods e.g. natural (Palmer et al., 2015; Warnken et al., 2017) and constructed wetlands (Dombeck et al., 1998; Kröpfelová et al., 2009; Sheoran and Sheoran, 2006) are widely used as a main treatment unit or a polishing step for Sb contaminated waters, due to their negligible operational cost and maintenance requirement. In the arctic region, which contains some of the largest metal mines and mineral reserves in the world (Boyd et al., 2016), together with abundant mires and peatlands, peat-based natural treatment wetlands or treatment peatlands are particularly important for polishing mining influenced waters (Eger and Lapakko, 1988; Palmer et al., 2015; Parviainen et al., 2014) and lower environmental impacts of mine activities on recipient water bodies.

Removal of metals and metalloids from wastewater using constructed wetlands has been previously studied (Kröpfelová et al., 2009; Sheoran and Sheoran, 2006; Sobolewski, 1996). However, research on the processes responsible for Sb retention and mobility in treatment wetlands is still in its early stages although it is essential from a peatland sustainability

perspective. Even less research has been done on peat-based treatment wetlands in cold climates periodically subjected to seasonal freezing-thawing cycles. A good understanding of the function of peatlands under these conditions is especially important since winters in artic regions are typically long and surface layers of peatlands remain frozen for much of the year. There is also evidence of a long-term risk of Sb leaching under certain conditions in laboratory tests (Khan et al., 2019) which needs to be further confirmed under field conditions.

Antimony removed by co-precipitation and adsorption with iron (Fe) and manganese (Mn) oxides can be released under reducing conditions through Fe and Mn (oxyhydr)oxide reduction and dissolution (Gambrell, 1994; Hockmann et al., 2014; Nakamaru and Altansuvd, 2014). However, under anoxic conditions Sb can also be immobilized by microbial Fe reduction (Burton et al., 2019) and by sulfide precipitation in peatlands with constant sulfate (SO_4^{2-}) input (Gadd and White, 1993; Palmer et al., 2015; Webb et al., 1998; White and Gadd, 1996). Importance of reduced sulfur species has also been highlighted for retention of Sb (e.g., in the form of Sb sulfide or sequestration through carboxyl/phenol/thiol groups of natural organic matter (NOM)) in wetland soils (Bennett et al., 2017; Besold et al., 2019a). However, sulfide precipitates can release Sb under oxidizing conditions, when sulfide oxidizes to SO_4^{2-} (Casiot et al., 2007; Reddy and DeLaune, 2008). Arsic et al. (2018) demonstrated that Sb mobilization from wetland soil is decoupled from the Fe cycle and possibly linked to oxidative dissolution of antimony sulfide or microbial breakdown of Sb-containing organic matter. Any of these processes, or a combination, can be important for mobilization of Sb from mining influenced water treatment peatlands in cold climate regions, but more studies are needed to determine the risk of Sb mobilization under various conditions. Knowledge of Sb mobilization is also important for assessment of Sb pollution sources on catchment scale (Chon et al., 2010) for compliance with European “Water Framework Directive (WFD)” (European Commission,

2000), since management of long-term pollution from mines will be a major challenge in achieving the chemical quality targets of the WFD (Wolkersdorfer, 2005).

There is a particular dearth of long-term field studies based on real mine water treatment wetlands in cold climate, focused on Sb and its reliable removal throughout the year. This study examined the long-term Sb removal trends in two peatlands polishing pre-treated mining-influenced water in an Arctic region since the start of their use as treatment peatlands. Data were evaluated with particular regard to the long-term effectiveness of Sb removal and changes in removal efficiency upon changes of mine water input composition (e.g. intensification of mine water purification before discharging waters to peatlands) and seasonal changes such as summer rain, snowmelt, and soil frost period. The main hypothesis is that peat-based treatment wetlands can provide stable Sb removal in cold climate under optimum conditions. In this context, the following research questions were addressed:

a) How do hydraulic load and contaminant areal load affect removal of Sb from natural treatment peatlands?
b) Since treatment peatlands accumulate pollutants over time, is there a decline in their effectiveness in retaining Sb and, subsequently, their capacity for absorbing sudden peaks of Sb?
c) How do seasonal changes such as freezing of upper layers of peatlands in winter and thawing in spring affect Sb cycling through treatment peatlands? Does the removal efficiency decrease in winter due to slowed down microbial and chemical activity?
d) Can melting of accumulated snow over treatment peatlands in spring and major rain events leach out Sb less strongly bound to peat?

The insights from the study presented here regarding functioning (removal of pollutants from wastewater), viability (performance under changing conditions) and sustainability

143 (implications of long-term use) of boreal peatlands (fen type of mires) used for water treatment
144 in cold climate are applicable and transferable to other cases of Sb pollution and similar types
145 of constructed wetlands (peat or other organic soils used as a medium of passive treatment
146 systems) all over the world, also including southern latitudes where tropical peatlands are
147 abundant (Kyambadde et al., 2004; Leng et al., 2019; Mitsch et al.,2008).

148 **2. Materials and Methods**

149 **2.1. Site description**

150 The study site comprises two natural peatlands polishing mining influenced water from a gold
151 mine in Finnish Lapland (67º54'N; 25º22'E) (Fig. 1). Treatment peatland A (TP-A, 44 ha) has
152 been used for purification of pre-treated excess process water since 2010. Treatment peatland
153 B (TP-B, 17 ha) is subjected to water from mine de-watering after pre-treatment and has been
154 in operation since 2006, although regular operation of the mine did not start until 2008. Both
155 treatment peatlands work more or less as a buffer zone between the mine and the recipient river
156 meaning that all water quality criteria for mine effluent waters must be met already in the
157 inflow waters to the peatlands. Based on the current environment permit, the mine effluent
158 waters cannot exceed Sb concentration of 0.3 mg L^{-1} as monthly flow weighted average or to
159 be over 0.8 mg L^{-1} in individual water sample (Court order 2201/2016). The concentration
160 limit for SO_4^{2-} is 2000 mg L^{-1} in the valid environmental permit of the mine company.

161 Each peatland receives influent through an inlet channel and discharges effluent to a nearby
162 river through outlet ditches. The treatment peatlands have been designed and located in a way
163 that minimizes the flow of natural runoff waters (from outside of the treatment areas) to the
164 treatment areas meaning that the mine waters are the major component of the inflow to the
165 treatment peatlands as has been studied in Palmer et al. (2015) (Fig. 1). Average volume of
166 inflow to TP-A is 3100 m^3 day^{-1} while TP-B receives 7000 m^3 day^{-1} (Table 1 and the year-wise

167 breakup of conditions in the Supplementary Material Table S1). The inlet water to TP-B is
168 lower in temperature and pH as compared to TP-A (Table 1). Another distinguishing factor is
169 that hydraulic load to TP-A has generally remained comparable over the years, while it has
170 gradually increased for TP-B in recent years (Supplementary Material Table S1). The mine
171 extracts and processes ore mainly comprising sulfide minerals such as arsenopyrite, arsenic-
172 rich pyrite, and pyrrhotite (Doucet et al., 2010). In November 2012, open-pit excavation was
173 discontinued, and the operations were moved entirely underground (Pöyry, 2015). Inflow to the
174 peatlands contains high loads of SO_4^{2-}, nitrogen (N), and various metals and metalloids
175 including, As, Sb, and nickel (Ni) (Palmer et al., 2015. More detailed inflow water quality data
176 can be seen in Supplementary Material Table S2). To meet the requirements concerning SO_4^{2-}
177 for a new environmental permit for the mine, a new mine process water treatment unit based on
178 gypsum precipitation was introduced in the beginning of 2017. This considerably reduced the
179 SO_4^2 concentrations in the influent to TP-A. Due to this issue and to highlight any possible
180 changes brought about by the new mine process water treatment plant, the data for TP-A has
181 been studied according to these two separated periods: the years 2010-2016 and the year 2017.

182 The studied mine is located well inside the Arctic Circle (Fig. 1a) in a region which typically
183 experiences permanent snow cover from October to May. During this period the precipitation
184 is accumulated as snow in the peatland areas and most of the melting takes place during the
185 rather short period of one month (usually April-May). Average amount of precipitation in the
186 form of snow for the snow cover period has been 197 mm (data from years 2008-2018).
187 Wetlands around the mine comprise 1-2 m of peat underlain by 3-6 m of moraine deposits
188 (Larkins et al., 2018) (Fig. 1c). Prior to their use as treatment unit for mining-affected waters,
189 the TPs have been fen-type peatlands which are typically fed by local groundwaters. However,
190 upon the startup of mining activity, the water table in the area has been lowered due to
191 intensive dewatering of the large mine area. Thus, the mine waters are the major component

keeping the mean water table within ±20 cm of the surface and maintaining water-saturated mire condition in the TPs. The surface vegetation cover in the treatment peatlands includes mosses, *Eriophorum angustifolium*, *Carex sp.*, and *Trichophorum cespitosum* (Palmer et al., 2015). More detailed information on the study sites can be found in previous publications (Khan et al., 2019; Larkins et al., 2018; Palmer et al., 2015).

Table 1. Brief description of conditions (mean ± standard deviation) in treatment peatland A (TP-A) and B (TP-B).

	TP-A (44 ha)		TP-B (17 ha)
	2010-2016	2017	2008-2017
Inflow (m^3 day^{-1})	3100±2522	3100±2383	7000±3390
Hydraulic load (mm day^{-1})	7±5.8	7±5.4	41±19.9
Inlet water temperature (°C)	7±5.6	6±5.4	5±5.2
Inlet water pH[a]	8.0±0.4	7.9±0.2	7.5±0.15
pH in peat porewater[a,b]	6.9±0.7		7.4±0.47
Peat hydraulic conductivity[b] (cm s^{-1})	0.017±0.02		0.036±0.03

[a] median ± standard deviation

[b]data is based on the field measurements done in 2013-2015

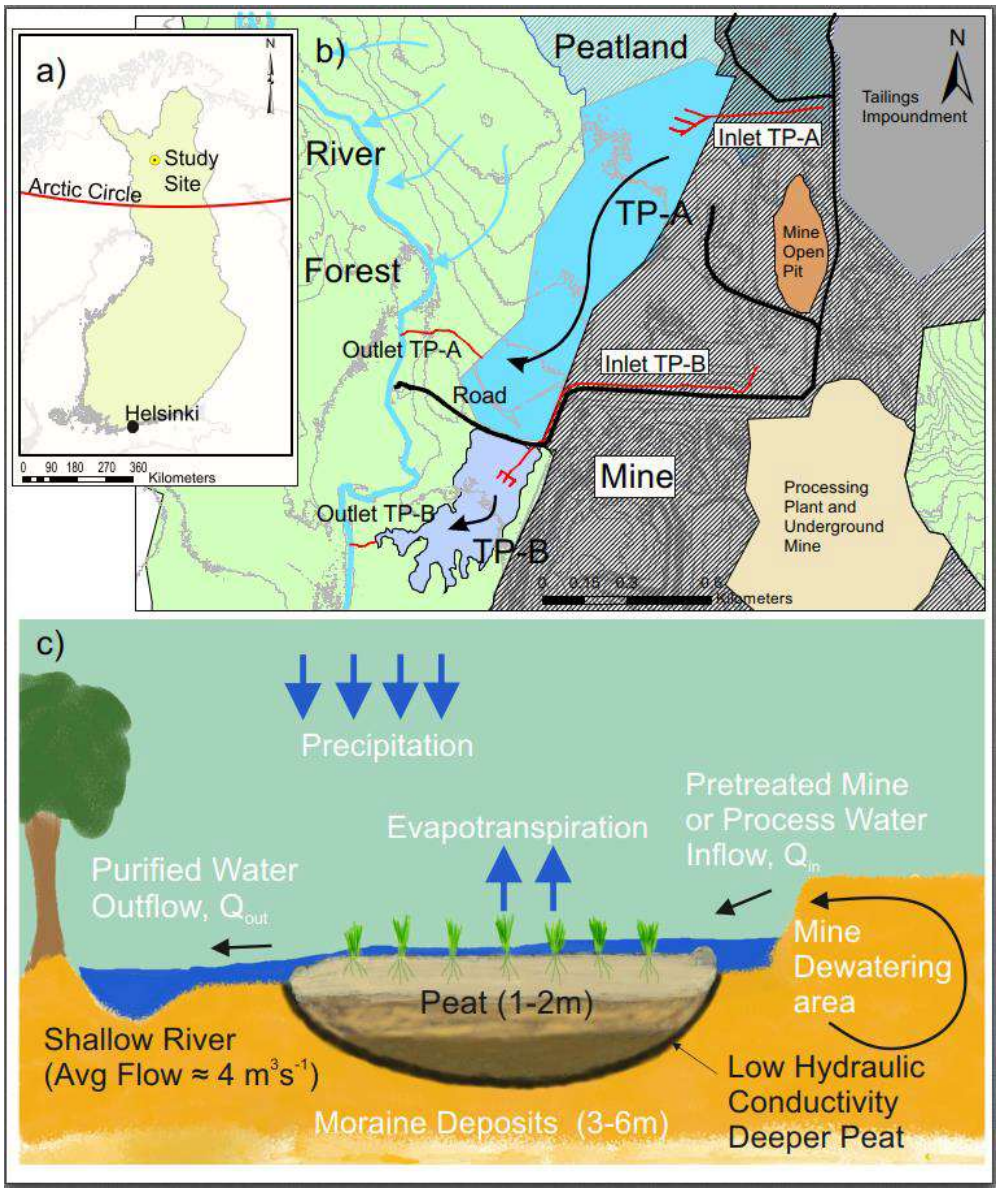


Figure 1. a) Location of the study site in Finnish Lapland. b) Topography of the area and layout of the peatlands. Treatment peatland A (TP-A) receives treated excess of mine process water and TP-B receives pre-treated mine dewatering water. Treated effluent from both peatlands is discharged into a nearby river. c) Conceptual cross section of the study site and water inputs and outputs to/from the treatment peatlands.

2.2. Data on water, peat and porewater characteristics

Data on quality of inflow and outflow water since the start of use of the treatment peatlands were collected specifically for this study or provided by the mining company (as a part of its monitoring program) or by the environmental authorities (mainly Geological Survey of Finland). Water samples were collected at different frequencies over the study

211 period and generally well represented the seasonal variability in a year. All water samples
212 were analyzed in accredited laboratories (AHMA Ympäristö Oy., Ramboll Analytics Oy.,
213 EUROFINS environment testing Finland Oy., and Lounais-Soumen vesi-ja
214 ympäristötutkimus Oy.), using standard methods specified by the Finnish Standards
215 Association (SFS) and certified by the Finnish Accreditation Service (FINAS). Data for
216 the period March 2010-December 2017 were compiled and analyzed for TP-A, and data
217 for the period July 2008-January 2018 for TP-B. Measurements below detection limits
218 (DL) were replaced by DL, which may have resulted in an under estimation of Sb removal
219 efficiency in TP-A in some cases, since most of the measurements below DL for Sb (2-15
220 $\mu g\ L^{-1}$) were from its outlet. In addition, the results so obtained have been compared,
221 where necessary, with results obtained by DL/2 substitution (USEPA, 2009). In addition
222 to inflow and outflow water analysis, various peat samples were collected in the period
223 2011-2018 to assess changes in concentrations of elements retained in the peat. Peat
224 samples from all sources were analyzed by HNO_3/HCl extraction (EPA 3051) at
225 accredited laboratories (Labtium Oy., AHMA Ympäristö Oy., and EUROFINS
226 environment testing Finland Oy.) following analysis methods in line with standards as
227 described for the water samples before.

228 **2.3. Meteorological Data**

229 In order to examine seasonality in wetland purification performance, available
230 meteorological data were used. Daily temperature and precipitation data from three
231 meteorological observation stations near the study site (Kittilä kirkonkylä, Kittilä Pokka
232 and Kittilä airport located about 30-35 km from the site) were obtained from the Finnish
233 Meteorological Institute. The values obtained from the three stations were combined and
234 the overall mean was calculated using data for the period 2008-2018. Precipitation was
235 separated between phases (snow/rain) using air temperature as the indicator and the

differentiating temperature between snow and rainfall was set at 1.0°C (Feiccabrino and Lundberg, 2009). Duration of the four seasons was as described for Northern Finland by the Finnish Meteorological Institute (2019), but with some modifications based on the meteorological data, and to fit calendar months into seasons. The seasons were defined as: spring (April and May); summer (June, July and August); autumn (September and October); winter (November, December, January, February and March).

2.4. Calculations and Statistical Analysis

Removal of Sb was determined from the concentrations in inlet and outlet (Eq. 1) as it is also used in many previous studies (e.g. Goulet et al., 2001; Kujala et al., 2019).

$$\text{Removal (\%)} = (C_{inlet} - C_{outlet}) \times 100 / C_{inlet} \quad (1)$$

where C_{inlet} and C_{outlet} is the Sb concentration ($\mu g\ L^{-1}$) in inlet water and outlet water of the peatlands, respectively. Mean of measurements in a calendar month was used for both C_{inlet} and C_{outlet}.

Since most of the data were not normally distributed as determined with the commonly used Shapiro-Wilk test of normality (Royston, 1995; Shapiro and Wilk, 1965), non-parametric Mann-Kendall test was used to detect significant trends in the time-series data (Kendall, 1975; Mann, 1945) and Sen's slope estimator (Sen, 1968) was used to calculate the magnitude of trends. Mann-Kendall test analyzes the sign (+ or -) of difference between successive values in a time series data and is a commonly used test to assess whether data has a monotonic trend over time. The significance level was set to $p \leq 0.05$, at which the null hypothesis of no trend is rejected if Mann-Kendall $Z > 1.96$. Trend analysis was carried out using "trend" package in R (Pohlert, 2018) and Microsoft Excel. Correlation analysis between Sb outlet concentration/removal and loading parameters was

assessed by non-parametric Spearman's rank correlation coefficient (r_s) using OriginPro 2018.

In order to understand the large dataset, exploratory analysis of the multivariate water composition data was performed through ordination by principal component analysis (PCA). The package "vegan" for R was used for this purpose (Oksanen et al., 2017). The first two principal components (PC) accounted for about 50% of the total variance in the data for both TP-A and TP-B.

Dilution due to snowmelt water and rainfalls were estimated in monthly scale using hydrological data. The snow was assumed to accumulate during the winter months and the Sb concentration of precipitation was assumed to be 0 µg Sb L^{-1} as it has been measured to be in the level of nanograms (Tripathi and Patel, 1998). The time scale of one month was selected for dilution effect calculations as this is also the typical water residence time estimated in TP-A. Both two weeks and one-month duration for melting of snow accumulated over the winter was used to get a better understanding about the role of this water fraction in concentration changes.

3. Results

3.1. Effect of Sudden Changes in Inlet Water Composition

Typically, the treated excess of mine process water received by TP-A contained 57±33.7 µg Sb L^{-1} (excluding the year 2017 measurements). This is lower than Sb in the mine dewatering water (228±106.14 µg L^{-1}) treated by TP-B. Generally, data for TP-A indicated stable performance of the wetland up to 2016, with the outlet Sb concentrations remaining relatively low and constant even though the inlet concentrations greatly varied (Fig. 2a). Year on year comparison of in- and outlet concentrations indicated the same pattern (Fig. 3a). The two-dimensional score plots of water samples obtained by PCA also showed clear

differences between overall composition of inlet and outlet water samples for TP-A (Fig. 4a) pointing the effect of treatment peatland (in this case lowering the outlet concentrations) on water quality. Furthermore, the PCA analysis proved that overall the inlet water composition changed after the year 2016 (samples taken in 2017 formed a distinct group of points in Fig. 4b). This can be seen especially in concentrations of Sb, As and SO_4^{2-} (Fig. 5).

A sharp drop in the inlet Sb concentration of TP-A after the year 2016 led to remarkable decline in relative Sb removal in 2017 (Fig. 3c) along with a gradual increase in outlet Sb concentrations (Fig 2f; Mann-Kendall Z = 2.19, p=0.028). Magnitude of Sen's slope for this period was 0.036 and slope of linear regression was 0.041 (R^2=0.41). Man-Kendall Z value here indicate that a positive monotonic trend is present in the data at 5% significance level (as Z >1.96) while the magnitude of Sen's slope quantifies the magnitude of this trend, which may be interpreted as mild in this case. This increase in outlet Sb concentrations with declining inlet Sb concentrations indicates leaching of retained Sb from the peatland. For comparison, no such trend was seen in the data for 2016 (Z = 0.36, p=0.72) or 2015 (Z = 0.86, p=0.38).

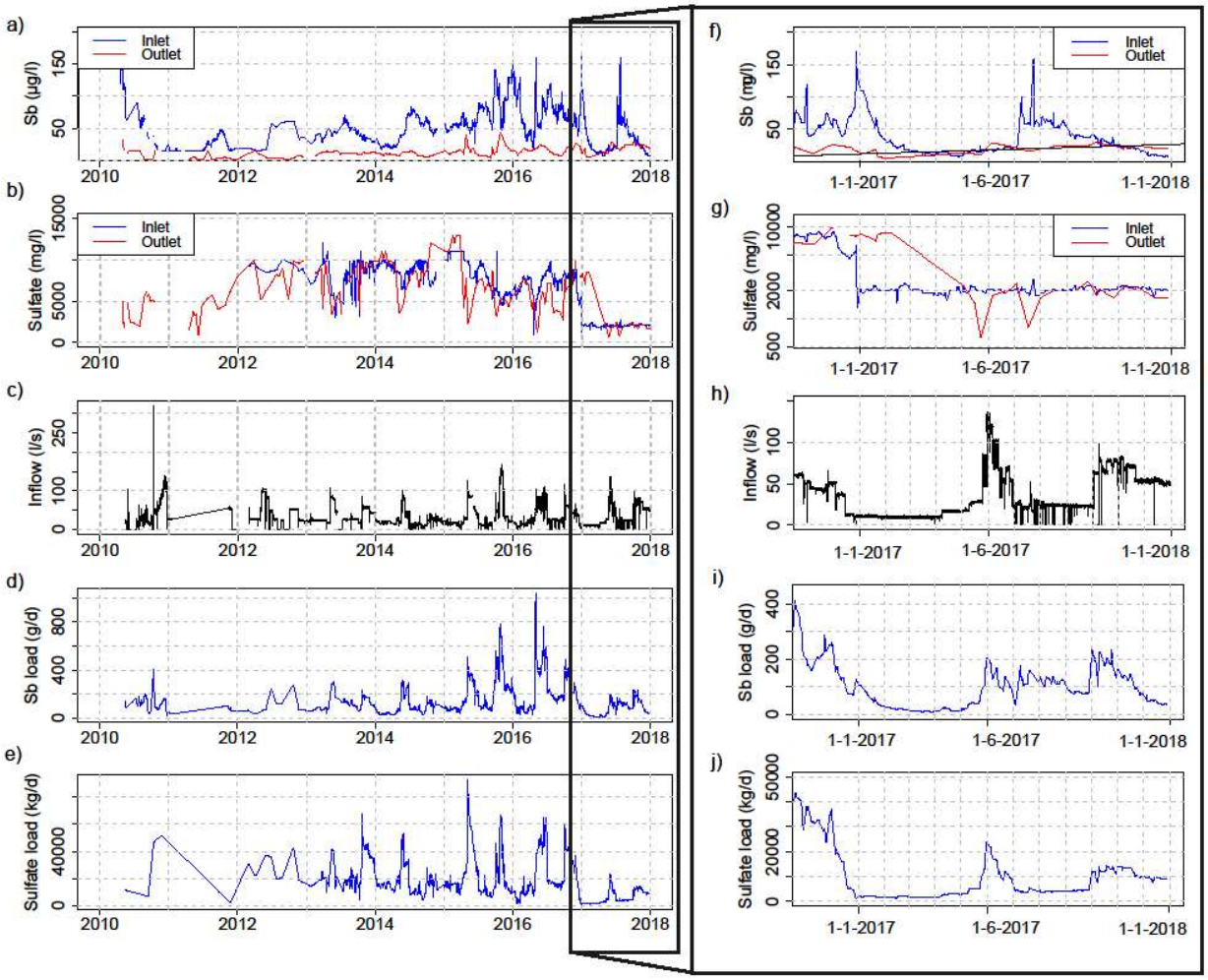


Figure 2. Time series of measured concentrations of a) antimony (Sb) and b) sulfate (SO_4^{2-}) in inlet and outlet water of treatment peatland A (TP-A), c) its inlet water flow rate, and inlet loads of d) Sb and e) SO_4^{2-}. The last year of the time series in a, b, c, d and e is enlarged in f, g, h, i and j, respectively, in order to highlight the period after the new process water treatment plant preceding TP-A became operational. Daily loads calculated on the assumption that the measured concentration at any time represented that of the entire day. Data is based on 1192 and 176 measurements of Sb concentration from inlet and outlet of TP-A, respectively.

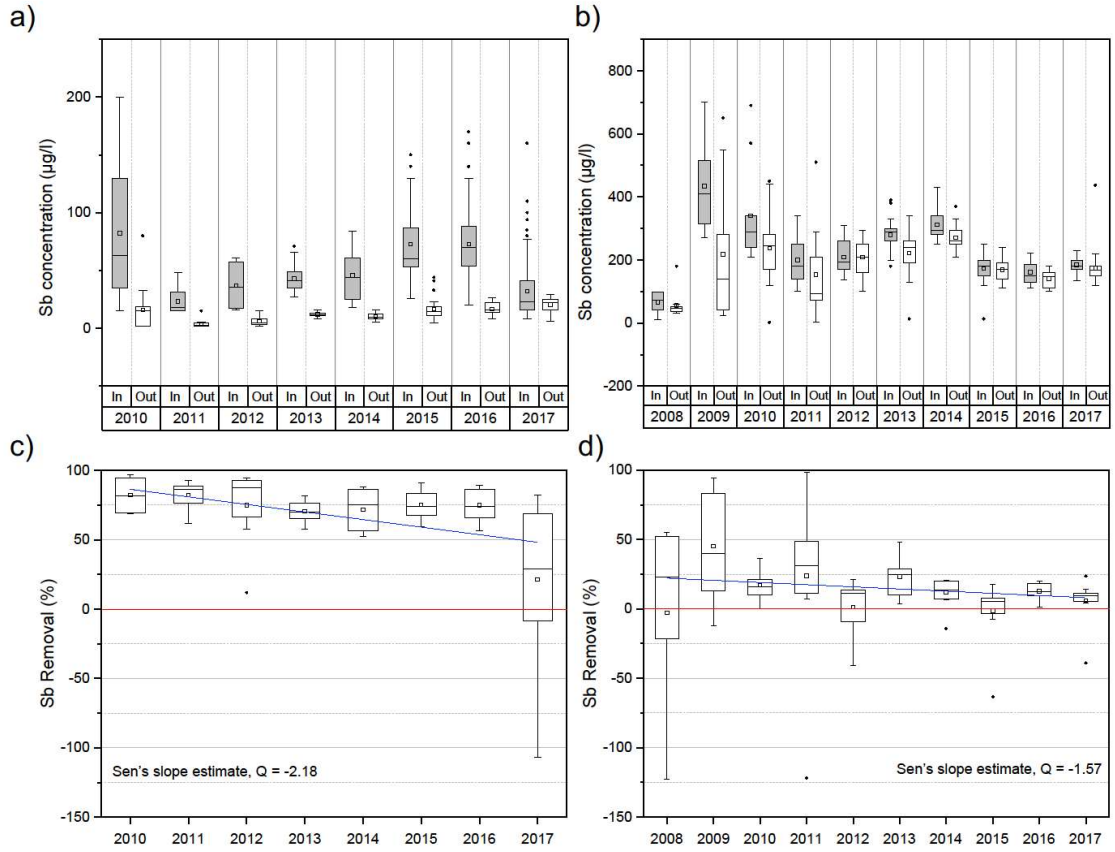


Figure 3. Year-wise antimony (Sb) concentrations in inlet and outlet water of a) treatment peatland A (TP-A, 1192 inlet and 176 outlet samples) and b) TP-B (180 inlet and 182 outlet samples), and calculated removal of Sb from c) treated excess of mine process water in TP-A and d) pre-treated dewatering water in TP-B. Removal was calculated for a calendar month from mean inlet and outlet concentrations during the month. Negative values indicate leaching/mobilization. Blue lines present trend of the yearly mean values. Box shows median, 25th percentile and 75th percentile of the data. Small squares represent mean while whiskers show outermost data point within 1.5×IQR.

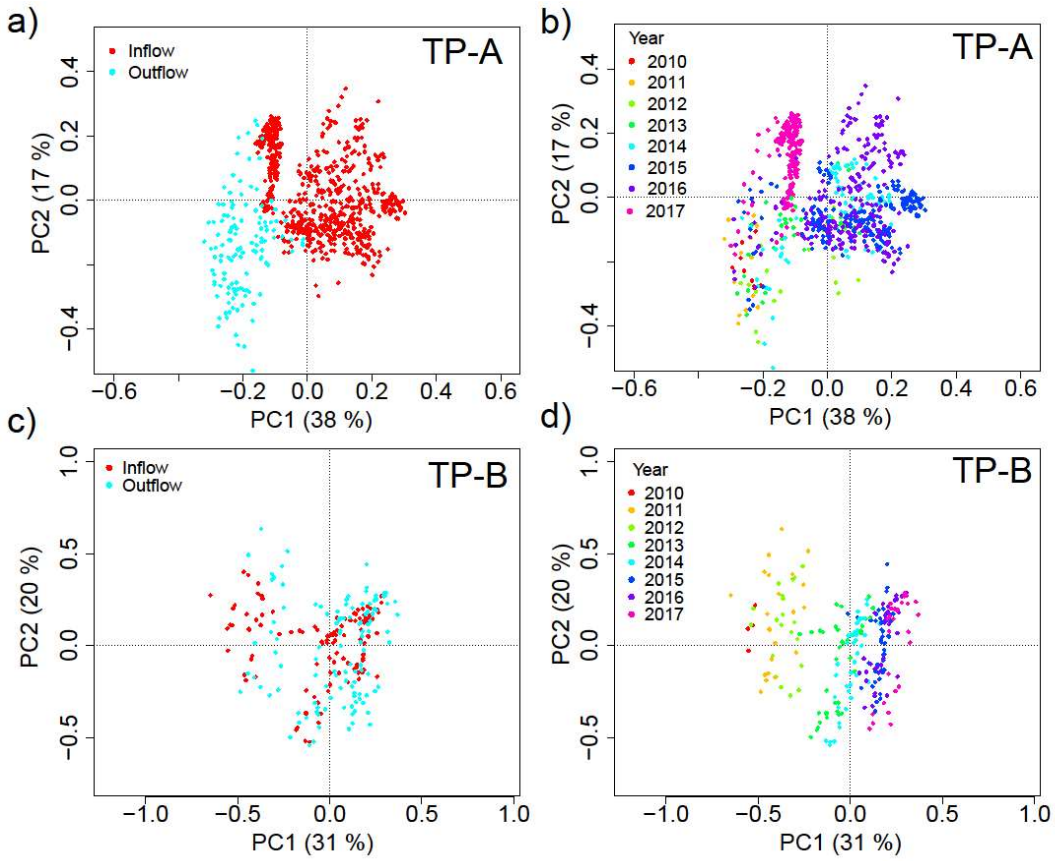
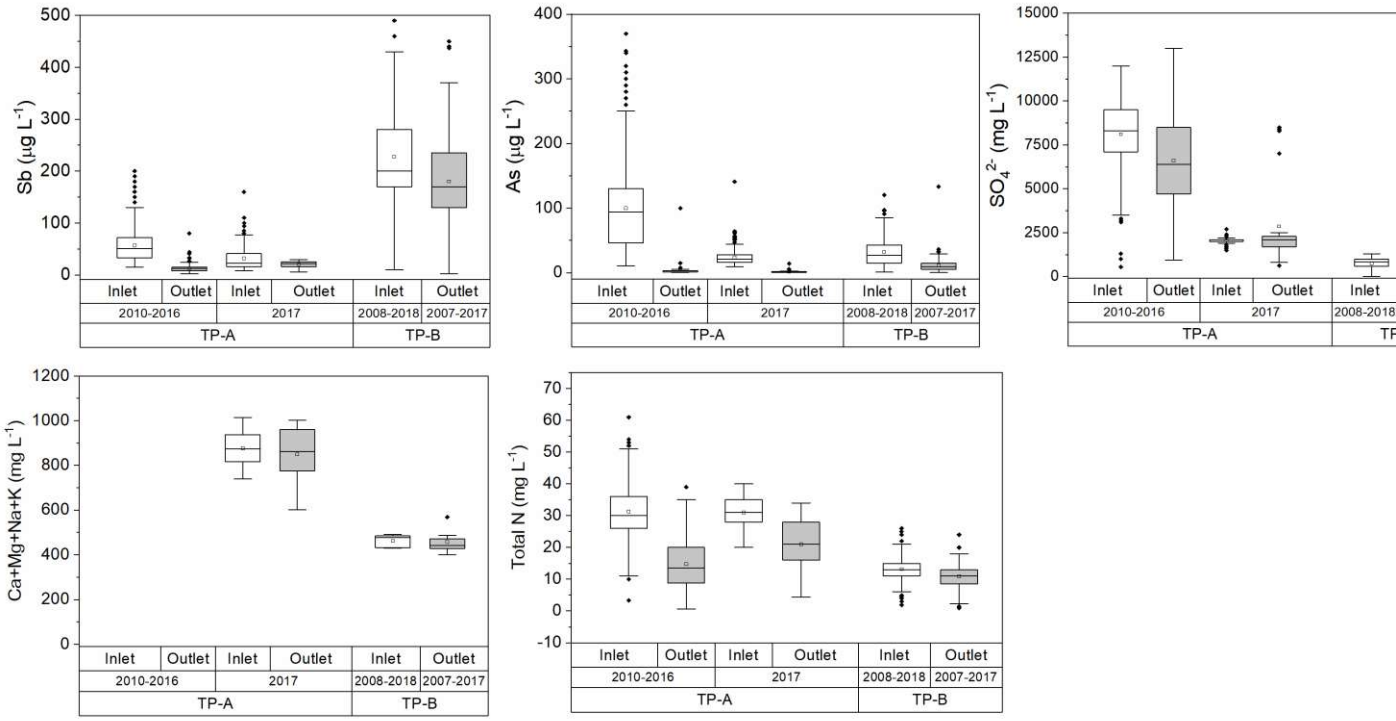


Figure 4. Ordination plots of multivariate water composition data for treatment peatland A (TP-A) and treatment peatland B (TP-B) in principal component analysis. Each point represents a single water sample plotted along the two principal components (PC) explaining the highest variance in data, clustered by sample source (a and c) and sampling year (b and d). Percentages in axis labels indicate the proportion of total variance explained by the respective principal component.



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329 Figure 5. Concentration of antimony (Sb) and various contaminants in inlet and outlet water for treatment peatl
330 The data of TP-A are split into two periods, 2010-2016 and 2017, to highlight changes in water quality due to t
331 water treatment plant in 2017. Calcium (Ca) and potassium (K) measurements were not available for TP-A in t
332 Box shows median, 25th percentile and 75th percentile of the data. Small squares represent mean while whiskers
333 point within 1.5×IQR (Inter-Quartile Range).

3.2. Long Term trends in Wetland Performance

At the time of this study, the treatment peatlands TP-A and TP-B had been in use for 8 and 10 year, respectively. During these years, Sb removal efficiency in TP-A has been 67±32% whereas it has been only 14±32% in TP-B which has received clearly higher Sb load (0.009±0.003 g m^{-2} d^{-1}, in the period 2010-2017) than TP-A (0.0003±0.0002 g m^{-2} d^{-1}). For the first 5 years of operation (up to the year 2014), the TP-A outlet Sb concentrations were steadily low although the inlet Sb concentrations highly fluctuated (Fig. 2a). After that, there were more peaks of high concentrations visible in the Sb outlet concentration time series, which followed peaks in Sb inlet loads with a certain lag. This may suggest a decline over time in the ability of peatlands to absorb peak loads. Generally, lag time between the peaks was around 20-30 days which was in the range of water residence times (7-42 days depending on the hydraulic load) determined in the year 2013 (Palmer et al. 2015), but due to sparse sampling of outlet water, the accurate lag time between inlet and outlet peaks was difficult to determine.

Sb removal efficiency were not high and stable in TP-B. The outlet Sb concentrations were only slightly lower than in the inlet water, particularly in recent years (Fig. 6a). The two-dimensional score plots of water samples obtained by PCA also show that the TP-B inlet and outlet samples were very similar in composition as they overlapped in the plots, indicating overall minimal removal performance in TP-B (Fig. 4c). Peaks in outlet concentrations of Sb followed peaks in inlet concentrations (Fig. 6a), indicating the inability of the peatland to absorb sudden peaks in Sb loads, unlike TP-A in its early years. In TP-B, inlet Sb concentrations declined starting in 2015 and the outlet concentrations followed a similar trend, although mean inlet loads remained comparable to previous years (1.8±0.4 kg day^{-1} in 2014 and 1.8±0.5 kg day^{-1} in 2017) since the dewatering water amount has increased (due to expansion of the mine activities). The opposite observation was

made for SO_4^{2-} concentrations and SO_4^{2-} loads as those to TP-B both showed a continuous rise (Fig. 6b and Fig 6e).

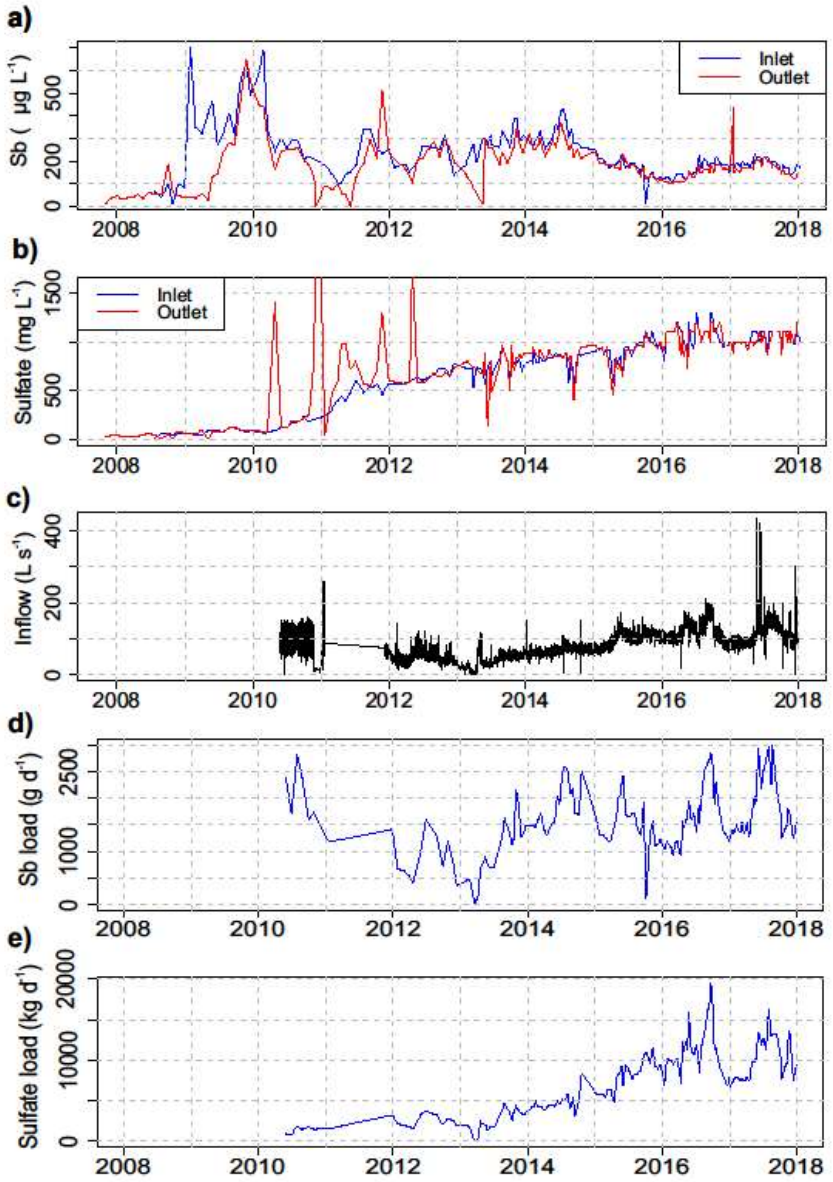


Figure 6. Time series of measured concentrations of a) antimony (Sb) and b) sulfate (SO_4^{2-}) in inlet and outlet water of TP-B as well as inlet flow rate (c), and inlet loads of Sb (d) and SO_4^{2-} (e) of TP-B. Daily loads have been calculated on the assumption that the measured concentration at any time represented that of the entire day. Data is based on 181 and 184 measurements of Sb concentration from inlet and outlet of TP-B, respectively.

371 In both of the studied treatment peatlands, Sb removal efficiency changed during their
372 operation time. Median removal of Sb from treated excess of mine process water for TP-A
373 (Fig. 3c) exceeded 80% in the first three years (2010, 2011 and 2012; n=22), but declined
374 slightly, ($p<0.05$, Mann-Whitney U=239), to around 75%, in later years (2014, 2015 and
375 2016; n=34). In 2017, the median removal efficiency dropped to 29% with even negative
376 values (mobilization) obtained for some months. The decline in removal efficiencies in
377 later years still persisted when different calculation methods for values below the detection
378 level (DL/2 substitution) was used, although it was slightly smaller in magnitude
379 (Supplemental Fig. S1) and not significant ($p=0.06$, Mann-Whitney U=282). In contrast to
380 TP-A, Sb removal from dewatering water in TP-B was much lower and more variable (Fig
381 3c and Fig 3d). Leaching was observed mainly in 2008, 2012, and 2015. Inlet
382 concentrations to TP-B declined in later years and relatively lower Sb removal efficiency
383 was observed in these years (Fig. 3d). Meteorological data from these years
384 (Supplementary material, Fig. S2) did not reflect any major variations that may indicate
385 that these changes in treatment peatland performance were due to climate. However, mean
386 yearly volume of inflows to TP-B correspond to annual precipitation, as higher
387 precipitation requires more dewatering, i.e. yearly inflow volumes were higher in wetter
388 years (Supplementary material, Table S1 and Fig. S2).

389 Concentrations of Sb in the surface peat layer (0-10 cm) of TP-A continued to increase
390 over the years, but the most recent samples, collected in 2018 when the process water had
391 been discharged to the peatland for 8 years, showed a sharp decline (Fig. 7a). The decline
392 was confirmed with an additional sample taken three months after the previous sample. By
393 this time, the new mine water treatment plant had been in use for more than one year
394 meaning that the inlet concentration to TP-A had been low for the last one year.

Accumulation of Sb in the surface peat of TP-B reached its peak after five years of use (in 2013) and varied only slightly over the years since then (Fig. 7b).

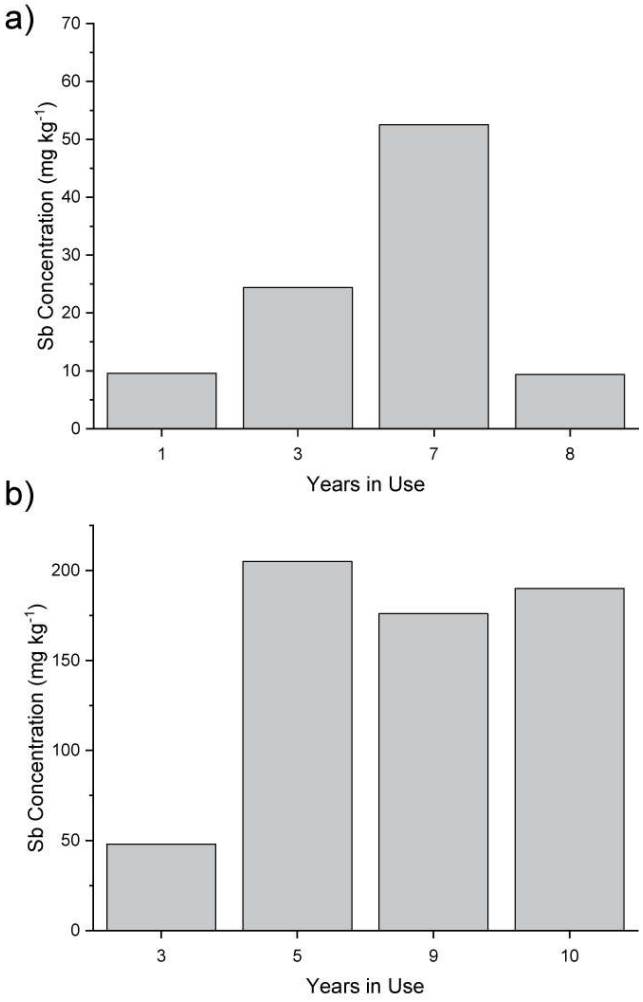


Figure 7. Accumulation of antimony (Sb) in surface peat (0-10cm) as a function of years after establishment of treatment peatland TP-A (a) and TP-B (b). The year in which mining operations started (2008) was taken as the start of use for TP-B, while the start of use for TP-A was in 2010. Values shown are for samples collected from a point close to the inlet in each treatment peatland. Decline in Sb concentration in TP-A (a) was confirmed through an additional sample which was taken 3 months after the previous sample (samples taken in July 2018 and October 2018).

3.3. Effect of Inlet Sb Concentration and Loading

The two studied treatment peatlands differed highly in their purification performance but also in their inlet water quality. Based on correlation and regression analysis for both datasets together, there were slightly positive correlation between Sb removal efficiency

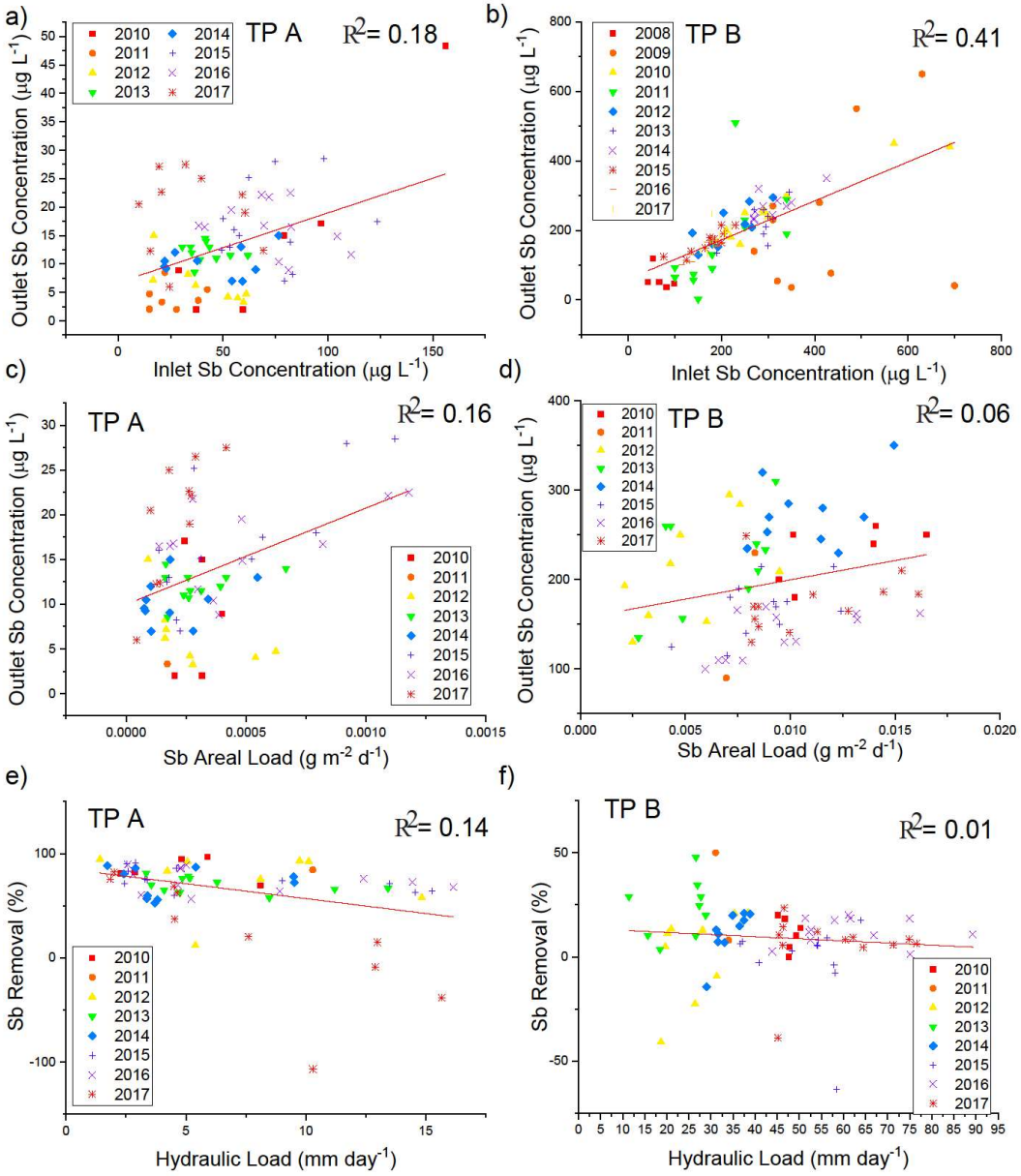
and inflow pH (r_s=0.55, $p<0.05$)/electrical conductivity (EC,r_s=0.70, $p<0.05$). Since EC describes total amount of cations and anions in the water (Visconti et al., 2010), this result indicates the general understanding that higher loads can lead to higher removal efficiency. However, when both peatlands were studied separately, inlet and outlet Sb concentrations appeared to show a positive linear correlation (Fig. 8a and Fig. 8b). The strength of the positive correlation waned as the hydraulic loading to the peatlands decreased, i.e., it was weaker for TP-A (r_s=0.31, $p<0.05$) than TP-B (r_s=0.7, $p<0.05$). Mean monthly hydraulic load to the two peatlands range between 1.5-16 mm d^{-1} and 11-89 mm d^{-1} for TP-A and TP-B respectively, assuming the entire area (between inlet and outlet) of the two peatlands contribute to the treatment under all conditions. The hydraulic load itself displayed a negative linear correlation with relative Sb removal when the hydraulic load was sufficiently low, as was the case for TP-A (r_s=-0.34, $p<0.05$) (Fig. 8e). With higher hydraulic load, as was the case for TP-B, no correlation between hydraulic load and Sb removal was observed (r_s=-0.16, $p>0.05$) (Fig. 8f). The outlet Sb concentrations also increased with increasing Sb areal loading to the peatlands. The correlation between the two was stronger (r_s=0.29, $p<0.05$) when Sb areal load was low (<0.002 $gm^{-2}d^{-1}$ for TP-A) (Fig. 8c). In comparison, Sb areal load to TP-B was higher (>0.002 $gm^{-2}d^{-1}$) and the correlation between areal load and outlet Sb concentration was weaker (r_s = 0.26, $p<0.05$) (Fig. 8d).

3.4. Influence of season on Sb retention

Based on the two studied treatment peatlands, there were no significant differences in Sb removal efficiencies between summer and winter seasons. However, slight differences in processes depending on seasons were observed. Generally, Sb concentrations in outlet of TP-A, grouped by calendar months, were more or less stable (Fig. 9a), although visibly lower in the months at the end of summer season with high rainfall (Fig. 9c) (i.e., August

435 and September, 11±7.2 µg L^{-1}, n=42, Fig. 9a) than in other months (15±9.5 µg L^{-1},
436 n=134). Excluding the measurements from 2017, since the new mine process water
437 treatment unit became operational, outlet Sb concentrations for TP-A in the period
438 December-March (13±4.7 µg L^{-1}, n=32) tended to be slightly higher than concentrations in
439 summer months June-August (11±5.6 µg L^{-1}, n=39) ($p<0.05$, Mann-Whitney U= 466).
440 However, the difference became insignificant (p=0.06, Mann-Whitney U=488) when
441 DL/2 substitution was used for measurements below the detection limit.

442 For TP-B, the inlet and outlet concentrations of Sb were lowest in April (Sb_{in} = 180 µg L^{-1}
443 and Sb_{out} = 130 µg L^{-1}) the month when snowmelt typically starts (Fig. 9b). Since TP-B
444 treated wastewaters from de-watering of the mine, it is obvious that snow melting in the
445 whole mine area subjected to de-watering can lead to lower inlet concentrations. If snow
446 accumulated over the winter is considered to melt in a duration of two weeks, it generates
447 the inflow of 8642 m^3 day^{-1} to TP-B which is 36% more than typical inflow during April
448 (6450 m^3 day^{-1}). The dilution effect from this alone can explain the observed low median
449 outlet concentration of 130 µg L^{-1} since estimated outlet Sb concentration after direct
450 dilution of the snow melting water would be nearly equal (132 µg L^{-1}). If the effect of
451 dilution was estimated considering one-month duration of snowmelt, the outflow
452 concentration would be 152 µg L^{-1} that is slightly higher than the measured concentrations
453 of the outflow.



454

455 Figure 8. Linear regression analysis comparing outlet antimony (Sb) concentration and
456 inlet Sb concentration (a, and b), outlet Sb concentration and Sb areal load (c and d), and
457 Sb removal and hydraulic load (e and f). Each point represents mean of measurements in a
458 calendar month.

459

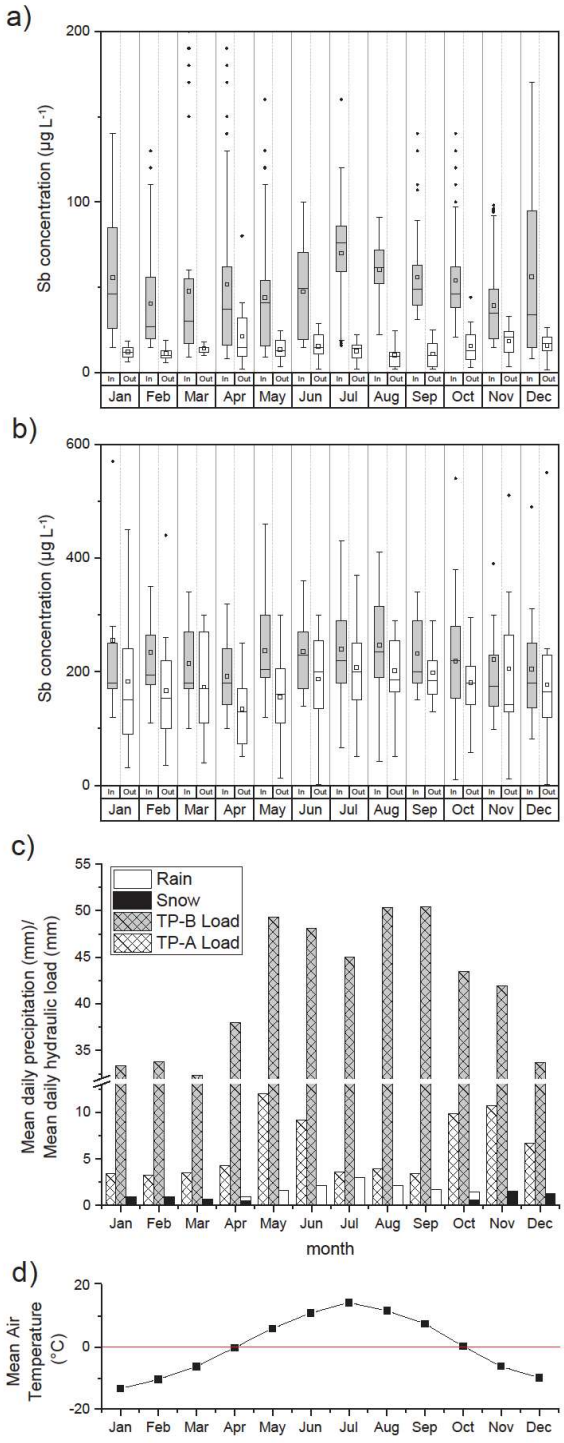


Figure 9. Antimony (Sb) concentrations, grouped by calendar months, in inlet and outlet water of a) treatment peatland A (TP-A) and b) TP-B. The data include all measurements from November 2007 to January 2018 for TP-A and from March 2010 to December 2017 for TP-B. Meteorological data from three observation stations near the study site were used to calculate c) mean daily precipitation (mean daily hydraulic load to each peatland is also presented for comparison) and d) mean monthly air temperature. Box shows median, 25th percentile and 75th percentile of the data. Small squares represent mean while whiskers show outermost data point within 1.5×IQR.

469

470 **4. Discussion**

471 The two treatment peatlands studied here were similar in terms of climate and location but
472 different in terms of composition of incoming water, hydraulic loading, areal loading of
473 contaminants, vegetation type, and flow conditions. These differences resulted in
474 differences in Sb purification performance. Based on the results of this study, in natural
475 treatment peatlands, even in cold Arctic regions, high Sb removal efficiencies (>50%) and
476 low stable outlet concentrations (13±9.2 μg L^{-1}) can be achieved, if the loading is
477 sufficiently low, as was the case for TP-A (<16 mm d^{-1} and <0.002 g Sb m^{-2} d^{-1},
478 respectively, before 2017). Large areas and small input loads are particularly recommended
479 for long-term metal removal in constructed wetlands, where the main metal removal
480 mechanisms depend on availability of retaining volume and removal sites (Sheoran and
481 Sheoran, 2006). Natural organic matter in the treatment peatlands provide such volume for
482 removal of Sb (Bennet et al., 2017; Besold et al., 2019a; Besold et al., 2019b, Dousova et
483 al., 2015). The importance of hydraulic and areal loading, observed in this study for Sb
484 removal, was also seen previously for nitrogen and phosphorous removal by treatment
485 peatlands (Heikkinen et al., 2018). Hydraulic loading affects the peatland hydrology, which
486 in turn controls their biogeochemistry (Reddy and DeLaune, 2008). When the hydraulic
487 loading is high, peatlands remain submerged and the peat remains saturated, which was
488 evident as free water surface over the entire area of TP-B in this study. High hydraulic
489 loading also results in shorter residence time for contaminants in peatlands. Presence of
490 preferential flow paths in wetlands (Ronkanen and Kløve, 2009) can further reduce the
491 effective residence time (P.Lane and Associates Limited, 1990). Since there is then less
492 contact between contaminants in water and peat and less time for various physical,
493 chemical and biological processes to take place, the outlet concentrations readily reflect the

494 inlet concentrations, as in TP-B. Shorter residence times also hinder Sb removal in
495 peatlands because of insufficient time for reduction of antimonate, which shows less
496 tendency for sorption to peat, to antimonite which is efficiently removed (Besold et al.,
497 2019b). Interestingly, for TP-B in 2009 (its second year in use for treating mine water) inlet
498 and outlet concentrations deviated strongly from the linear regression line. It is possible
499 that in the beginning there was an initial stabilization phase for TP-B leading to
500 fluctuations in its treatment efficiency.

501 Concentrations of Sb in peat in TP-B stabilized quite early on (Fig. 7) which may be
502 interpreted as the peatland reaching saturation through contaminant accumulation,
503 contributing to a further decline in relative removal of Sb in later years. The sharp decline
504 in relative Sb removal at TP-A in 2017 can be linked to a drop in the inlet Sb
505 concentrations possibly due to co-precipitation of Sb during SO_4^{2-} removal in the new mine
506 process water treatment unit installed in the beginning of 2017. The treatment unit
507 decreased inflow SO_4^{2-} concentrations to the peatland by 75%, to a level of 2060±144.9 mg
508 L^{-1} (Fig. 2g). Besides Sb and SO_4^{2-} removal, the water treatment unit also decreased As
509 concentration and other metals in the mine process water. The decline in inlet Sb was
510 accompanied by a gradual rise in outlet Sb concentrations during 2017, which can be
511 attributed to leaching and/or low retention of Sb in peat when concentrations are low in the
512 incoming water. As shown in a previous study based on batch sorption experiments using
513 peat samples from the same peatland (Khan et al., 2019), the leaching of Sb increases when
514 inflow concentrations decrease remarkedly. The results of this study confirm that there is
515 indeed a risk of mobilization which needs to be considered from a sustainability point of
516 view when making the decision to use natural peatlands for Sb removal.

517 The possible leaching of Sb is also revealed by the measured sudden decrease in the
518 concentration of Sb in surface peat samples collected in 2018 (Fig. 7). This surface layer,
519 however, may not be indicative of Sb removed by metal sulfide precipitation since that
520 process is considered to occur predominantly in the deeper peat layers which are typically
521 anoxic/reducing in treatment peatlands (Heikkinen et al., 2018). Comparing the trend in Sb
522 concentrations in surface peat layer, it can be seen that the peak concentrations, although
523 they may not be representative of concentrations in the entire peatland and Sb may have
524 travelled from surface to deeper layers, stabilized around 200 mg kg^{-1} in TP-B, while the
525 Sb peak reached in TP-A was around 50 mg kg^{-1} before it decreased substantially in the
526 2018 samples. This may also indicate that mobilization of Sb in TP-A occurred due to
527 concentration gradient between porewater and peat, and not because the removal capacity
528 of peat for Sb was exhausted. Palmer et al. (2015) proposed the possibility of contaminant
529 leaching from the same peatlands following changes in concentration gradients under
530 scenarios such as this. During later years (2014-2016), the relative Sb removal from TP-A
531 decreased which also points to accumulation of Sb in peat contributing to the changes in
532 concentration gradient. This, along with the decline in the capacity of peatland to absorb Sb
533 concentration peaks, suggests that treatment peatlands should not be used indefinitely for
534 treatment of Sb contaminated waters e.g. as a long-term treatment system after mine
535 closure. In addition, the peak Sb concentrations in peat reported in this study are higher
536 than the soil toxicity guidelines of Sb for many countries including Finland (Bagherifam et
537 al., 2019). This highlight the importance of careful planning so that peatlands receiving Sb
538 contaminated waters can be sustainably maintained and risk in Sb leaching prevented.

539 An important pathway for retention of Sb in contaminated peatlands is through sorption on
540 (oxyhydr)oxides of metals, e.g., Fe and Mn. Changes in redox conditions in the peatlands
541 brought about by the improved inlet water quality and associated changes in peatland

environment may have led to reduction of Fe and Mn oxides and mobilization of Sb bound to them (Nakamaru and Altansuvd, 2014). Because Mn concentrations in outlet water of TP-A during 2017 have remained comparable to the concentrations in earlier years even when its inlet concentrations declined substantially (supplemental Table S2), we can surmise mobilization of Sb bound to Mn. However, recent evidence suggests that removal of Sb through Fe phases is limited only to the surface layers (Besold et al., 2019b). The results of this study also showed SO_4^{2-} mobilization, when the 2017 measurements of inlet and outlet SO_4^{2-} concentrations were considered. The sudden decline in SO_4^{2-} input in 2017 may have resulted in mobilization of retained SO_4^{2-} from the peatland, possibly with associated mobilization of some of the retained Sb since there were no remarkable changes in other water quality parameters such as dissolved oxygen or pH.

Many of the biological and chemical processes in treatment wetlands are affected by temperature. In addition to the effect of temperature, the studied peatlands remain covered with snow for a large part of the year with a frozen surface layer changing oxygen transportation from air to the water and peat. Therefore, seasonal variations in conditions were expected to culminate in variation in relative Sb removal in the artic peatlands. However, the results of this study indicated that temperature and snow cover did not influence the processes of Sb removal in TP-B, most likely because of the high hydraulic loading and small wetland area, which have been observed to keep the preferential surface flow paths unfrozen and active even in winter. In addition, sulfate reducing bacteria (SRB) have been shown to adapt well to low temperature conditions in some studies (Fortin et al., 2000; Sagemann et al., 1998) suggesting stable Sb removal through sulfide precipitation. How winter conditions affect direct binding of Sb to NOM in peatlands under reducing and sulfur rich conditions is not completely clear and needs further exploration.

In contrast to the situation in TP-B, Sb removal in TP-A was mildly responsive to seasonal variations, with e.g., slightly higher outlet concentrations in winter. This suggests that the processes responsible for the fate of Sb in TP-A may be affected by: i) changes in temperature, ii) reduced microbial activity (Stein et al., 2007), iii) a reduction in available volume and sites due to a frozen top layer, iv) changes in redox conditions due to variations in oxygen concentrations, and v) increased metal flux due to evapotranspiration in summer (Sobolewski, 1996), or a combination of these factors. Concentration of dissolved oxygen in inlet waters suggest that antimonate should be the dominant Sb species, although both antimonite and antimonate could be present in aerated waters (Filella et al., 2002). Large size and longer retentions times in TP-A due to smaller hydraulic loading can provide more time for reduction of antimonate to antimonite, which has more affinity for peat (Besold et al., 2019b). This could be one of the reasons why the two peatlands behave slightly differently under the same seasonal variation.

Seasons, especially snow melting, seemed to play a part as far as decline in outlet concentrations through direct dilution is concerned. In TP-B, dilution due to snowmelt was estimated to decrease the outlet Sb concentrations in April but already in May dilution was observed to play only a minor role to lower contaminant concentrations in the outlet. Generally, dilution during snowmelt has been cited as the primary metal attenuation process in wetlands with channelized flow (Höglund et al., 2004). On the other hand, a high snowmelt flow can also emphasize leaching of Sb from the peatland because of its role in hydrological processes. Precipitation in the form of snow keeps accumulating over the entire winter and causes dilutions mainly during the months of snowmelt. The scale of dilution during the snowmelt season can be high enough to cause leaching by inversion of the concentration gradient between the peat and the porewater. This process can partly

590 explain the high outlet Sb concentration and low removal efficiency in TP-A during
591 snowmelt (Fig. 9a).

592 Also, high rainfalls can dilute the concentrations during summer and autumn seasons in
593 cold climate region. This was seen in TP-A where hydraulic load to the peatland area was
594 close to the water input contributed by total precipitation during July-September (months
595 with high rainfall, Fig. 9c) and concurrently outflow Sb concentrations were slightly lower
596 than in the other months (Fig. 9a). When only dilution was estimated to lower outflow
597 concentration it would produce the outflow with around 40 µg Sb L^{-1} due to direct dilution
598 of the rainfall. The measured median Sb concentrations, however, were around 10 µg L^{-1}
599 indicating also high Sb removal efficiencies in TP-A. This emphasizes that summer
600 rainfalls, although total precipitation is typically higher during summer than winter (Fig.
601 9c), does not cause dilution or leaching in a similar way that snowmelt flow does since it
602 has a different effect on treatment peatland hydrology. During summer months,
603 precipitations can immediately contribute to dilution but the distributed nature of the
604 precipitation, large size of treatment areas, role of evapotranspiration, and changes in
605 retention times should be taken into account as these can lower the role of rain in dilution.
606 The results highlight the significance of climatic factors to be considered for
607 comprehensive understanding of removal efficiencies of treatment wetlands.

608 **5. Conclusions**

609 Peat-based natural wetlands in the Arctic region can efficiently remove Sb from mining
610 influenced water if the area of the wetland is adequate to provide sufficient contact between
611 water and peat soil. In such wetlands, outlet Sb concentrations increase with increasing Sb
612 areal load and hydraulic load. When a wetland has been used for some years, a sudden
613 improvement in inflow water quality can lead to deterioration of treatment efficiency, even

in large wetlands. This is possibly because of changes in concentration gradients and subsequent leaching of Sb accumulated in the peat. Therefore, the perception that natural wetlands can keep providing long-term treatment for mine water, even after mine closure, needs to be revisited and more attention should be given to restoration of treatment wetlands to their natural state.

Season has only a minor influence on Sb removal processes even in cold climatic region, resulting in slightly lower outlet concentrations in summer. Inundation of wetlands due to melting of accumulated snow and rainfall can affect Sb retention in treatment peatlands. Summer rainfall can potentially cause dilution but snow has a more pronounced cumulative effect at the end of winter. However, the effect of more evenly contributing summer rainfalls can be diminished due to good Sb removal processes in treatment peatlands which can maintain good purification efficiency.

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Conflict of interests statement

637 The authors confirm that there are no known conflicts of interest associated with this publication
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639 outcome.

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911

912 **Supplementary Material**

913 Table S1. Year-wise breakup of conditions in treatment peatland A (TP-A) and B (TP-B). The

914 values represent annual mean ± standard deviation.

Year	TP-A				TP-B			
	Inflow (m3/day)	Hydraulic load (mm day^{-1})	Inlet water temperature (°C)	Inlet water pH*	Inflow (m3/day)	Hydraulic load (mm day^{-1})	Inlet water temperature (°C)	Inlet water pH*
2008	-	-	-	-	-	-	4.1±4.1	7.4±0.2
2009	-	-	-	-	-	-	4.8±5.4	7.6±0.1
2010	4500±3192	10.2±7.2	7.3±5.4	7.8±0.4	6597±3474	38.8±20.4	4.9±4.1	7.7±0.1

2011	2543±1734	5.8±3.9	5.4±4.0	8.4±0.3	5428±3469	31.9±20.4	5.1±5.2	7.5±0.1
2012	3071±2121	7.0±4.8	6.4±5.0	7.8±0.5	4540±1560	26.7±9.2	3.4±3.4	7.4±0.15
2013	2845±1693	6.5±3.8	8.2±6.3	8.1±0.2	3482±1802	20.5±10.6	5.2±6.0	7.5±0.2
2014	2063±1311	4.7±2.9	7.1±6.1	8.3±0.2	5868±937	34.5±5.5	6.5±6.5	7.5±0.1
2015	3438±3309	7.8±7.5	6.9±4.7	7.9±0.4	8792±1628	51.7±9.6	4.7±4.4	7.4±0.1
2016	3140±2179	7.1±4.9	7.1±5.4	7.8±0.4	10480±2366	61.6±13.9	4.7±4.7	7.5±0.1
2017	3081±2383	7.0±5.4	6.0±5.4	7.9±0.2	9835±3107	57.9±18.3	5.1±5.4	7.6±0.1

*Median ± standard deviation

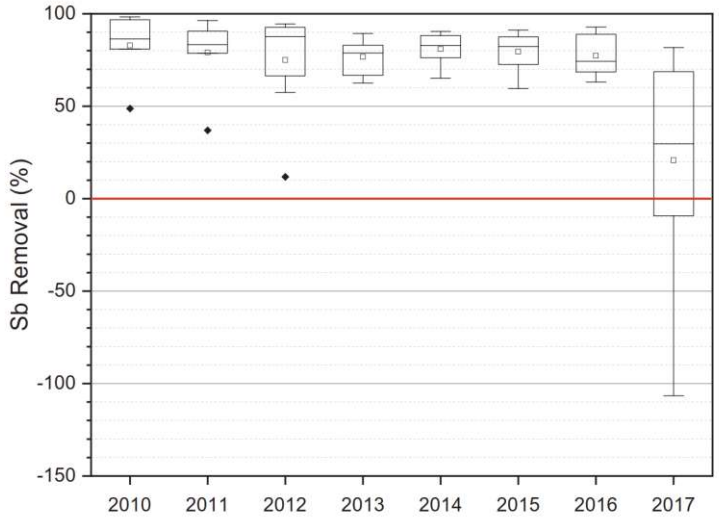
915

916

917 Table S2. Summary of inlet and outlet water quality for treatment peatland A (TP-A) and B
918 (TP-B). The data of TP-A are split into two periods, 2010-2016 and 2017, to highlight changes
919 in water quality due to the new mine process water treatment plant in 2017. The values
920 represent mean ± standard deviation.

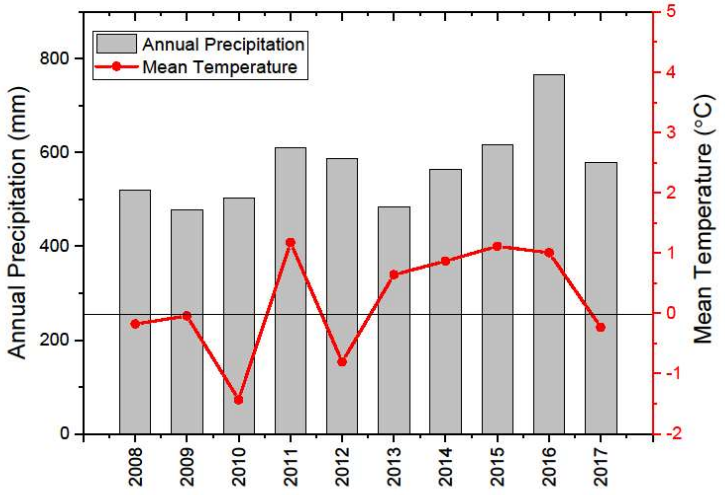
Parameter		TP-A				TP-B	
		Inlet		Outlet		Inlet	Outlet
		2010-2016	2017	2010-2016	2017	2008-2018	2007-2017
pH*		8.0 ±0.4	7.9 ±0.2	6.8 ±0.3	7.3 ±0.4	7.5 ±0.2	7.4 ±0.3
EC	(mS m^{-1})	820±142.5	342±17.7	755±237.1	416±227.8	162±59.7	164±78.6
Alkalinity	(mmol L^{-1})	-	2.93†	-	0.82†	2.73†	1.84 †
O_2	(mg L^{-1})	7.5 ±2.1	9.8 ±1.2	8.3 ±2.7	8.5 ±2.9††	9.5 ±6.1	7.0 ±2.8
Cl^-	(mg L^{-1})	27±5.9	29±4.2	24±11.2	26 ±7.7††	52±41.1	51±41.7
COD	(mgO_2 L^{-1})	31±10.1	24±7.6	33±21.0	18 ±4.66††	28±7.7	28±8.0
DOC	(mg L^{-1})		3.7†		3.1†	2.3†	2.9†
SO_4^{2-}	(mg L^{-1})	8110±1735.8	2060±144.9	6613±2629.2	2862±2362.4	754±343.5	764±453.8
Total N	(mg L^{-1})	31±7.7	31±4.4	15±8.1	21±8.8	13±3.8	11±3.9
Sb	(µg L^{-1})	57±33.7	32±23.8	13±9.2	20±6.6	228±106.14	180±96.8
Mn	(µg L^{-1})	2014±1739.2	212±198.1†† †	1202±1233.3	1047±2070.5	302±225.7	174.4±474.8†† †
As	(µg L^{-1})	100±62.9	24±13.2	5±9.7	2±2.8	32±21.6	12±11.7
Ni	(µg L^{-1})	39±37.1	21±8.3	9±6.1	7±7.6	64±41.7	36±58.8
Mg	(mg L^{-1})	1709±1670.3	47±42.7	1571 ±250.0††	114 ±43.2††	91 ±7.1††	79 ±26.5††
Na	(mg L^{-1})	232 ±97.1	158 ±32.7	218 ±32.8††	159 ±44.9††	70 ±8.1††	68 ±9.9††
Ca	(mg L^{-1})	-	560 ±59.9	-	474 ±93.3††	290 ±14.2††	299 ±42.2††
K	(mg L^{-1})	-	116 ±16.3		102 ±24.4††	12 ±1.9††	11 ±1.4††

*Median ± standard deviation
† One measurement
†† Calculated from less than 20 measurements
†††One measurement omitted from calculations as outlier



921

922 Figure S1. Year-wise removal of antimony (Sb) from pre-treated process water in treatment
923 peatland TP-A when measurements below the detection limit (DL) were replaced by DL/2.
924 Removal calculated for each calendar month from mean inlet and outlet concentrations during
925 the month. Negative values indicate leaching/mobilization. Box shows median, 25th percentile
926 and 75th percentile of the data. Small squares represent mean while whiskers show outermost
927 data point within 1.5×IQR.



928

929 Figure S2. Annual precipitation and mean temperatures at the study site during the study
930 period.