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The role of permafrost soils in Arctic mercury cycling: source tracing with Hg stable isotopes and revised soil pool estimate

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Mercury (Hg) is a pollutant of great concern for indigenous populations in the Arctic, which are exposed to high dietary Hg from fish and marine mammal consumption. Hg in marine biota can be derived from direct atmospheric deposition to the Arctic Ocean or from terrestrial sources by river runoff. Permafrost soils thereby play a pivotal role in the Arctic Hg cycle by storing atmospheric Hg deposition and providing a reservoir for later mobilization to the Arctic Ocean. The stability of Hg in permafrost soils depends on the pathway of atmospheric Hg deposition and Hg release processes, i.e. reduction and re-emission to the atmosphere and transfer to river runoff. We combined Hg stable isotope with Hg flux measurements in a field study on the Arctic Coastal Plain in northern Alaska. We could show that gaseous elemental Hg uptake by vegetation represents 70% of total atmospheric Hg deposition. Atmospheric Hg uptake by vegetation results in a characteristic Hg isotope fingerprint. This fingerprint dominates Hg signatures in permafrost soils measured across the Arctic coastal plain and is also imprinted in marine mammals and Ocean sediments, suggesting that Hg from Arctic permafrost soils represent a major source to the Arctic Ocean. Knowing the pool and spatial distribution of Hg in permafrost soils is therefore essential to assess current Hg mobilization to aquatic ecosystems and potential future changes due to permafrost thaw and climate change. Two recent studies have used Hg to carbon (C) ratios, R_{HgC} , measured in Alaskan permafrost mineral and peat soils, together with a northern soil carbon inventory, to estimate that these soils contain large amounts, 184 to 755 Gg of Hg in the upper 1 m. In a second part, we present new Hg and C data for six peat cores, down to mineral horizons, across a latitudinal permafrost gradient in the Western Siberian lowlands. Hg concentrations increase from south to north in all soil horizons, reflecting enhanced net accumulation of atmospheric gaseous elemental Hg by the vegetation Hg pump. We reviewed and estimate pan-arctic organic and mineral soil R_{HgC} to be 0.19 and 0.77 Gg Pg⁻¹, and use a soil C budget to revise the northern soil Hg pool to be 67 Gg (37-88 Gg, interquartile range (IQR)) in the upper 30 cm and

225 Gg (102-320 Gg, IQR) in the upper 1 m. Finally, we discuss how climate change may affect the mobilization of Hg from permafrost soils to the atmosphere and the Arctic Ocean.