MERCURY SPECIATION IN SOILS AND VEGETATION AT ABANDONED MERCURY MINES IN SOUTHWEST ALASKA

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To characterize the distribution of mercury species in soils and vegetation and the geochemical controls on the behavior of Hg in the terrestrial environment, we measured total Hg and methylHg concentrations in vegetation and total Hg, methylHg, Hg^{2+}, and Hg^0 in soils near three abandoned Hg mines (Red Devil, Cinnabar Creek, and Red Top) in southwest Alaska. Total Hg and methylHg in all samples collected near the mines are elevated over those in regional background samples. Vegetation contains as much as 970 ppb total Hg, whereas background samples contain no more than 190 ppb. MethylHg levels are as high as 37 ppb in mine site vegetation samples but background samples contain no more than 1.5 ppb. A subset of the vegetation samples was separated into leaf tissue, stem tissue, and flowering/fruiting body tissue and analyzed individually for total Hg. Leaf tissues consistently exhibited higher concentrations of Hg than either the stem or fruiting body tissues. Soil samples collected at the mine sites contain as much as 5,326 ppm total Hg and 133 ppb methylHg, whereas background samples contain no more than 3.7 ppm total Hg and 9.2 ppb methylHg. Divalent and monovalent Hg were measured in a subset of the soils samples collected and contain Hg^{2+} and Hg^0 elevated over background samples. Soil samples collected from the mines contain as much as 484 Hg^{2+}, background samples contain no more than 0.37 ppm Hg^{2+}. Mine site soils exhibit Hg^0 emission rates up to 8.80 ng/g/hr whereas fluxes from background soil samples are generally less than the detection limit of 0.02 ng/g/hr. Although samples from the mines have significantly elevated levels of all Hg species measured when compared with regional background sites, our data show that the ratio of methylHg to total Hg is higher in the background sites. That is, Hg contaminated soils may not accumulate significantly higher levels of methylHg. This is consistent with Hg data from aquatic environments but little is understood about the biogeochemical cycling of methylHg in terrestrial ecosystems. The elevated levels of Hg^{2+} found in the mine soils may play a role in controlling methylHg concentrations by way of an enzyme catalyzed (organomercurial lyase) microbial demethylation reaction that produces Hg^{2+} thus preventing high concentrations of methylHg from accumulating in these soils. The Hg^{2+} produced by demethylation may be further reduced to Hg^0 by another enzyme catalyzed (mercuric reductase) microbial reaction that is also controlled by the presence of Hg^{2+}. This microbial demethylation and reduction pathway is dependent on the presence of Hg^{2+} to trigger synthesis of the enzymes used in the reactions. When Hg^{2+} is present in low concentrations, such as in the regional background sites, these enzymes are not synthesized and methylHg may accumulate to proportionally higher levels than at sites where Hg2+ is elevated. It is assumed that most Hg found in plants comes from foliar absorption of Hg0 from the atmosphere. The elevated concentrations of Hg0 emission from mine soil samples probably explain the elevated levels of total Hg measured in mine vegetation samples.