Linkage between Hg(II) pore water speciation and MeHg production in contaminated sediments

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Introduction

The formation of toxic and biomagnifying mono methyl mercury (MeHg) in sediments has been proposed to be largely controlled by a passive uptake of dissolved, neutral Hg-sulfides in sulphate reducing bacteria. We have studied relationships between the concentration of dissolved neutral Hg-sulfides, modeled by equilibrium speciation calculations, and specific methylation rate constants (K_m, day^{-1}), as well as concentrations of MeHg (µg kg^{-1}), in sediments contaminated by Hg from the pulp and paper industry.

Materials and Methods

Sediments were sampled at seven sites in Sweden, covering a range of environments differing in salinity, total Hg concentration, annual air temperature sum, and organic matter content and quality. The potential methylation rate was determined by addition of isotopically enriched ^{201}Hg(II) followed by incubation at 23° C under N_2(g) for 48 h. Pore water Hg(II) speciation was modeled using measured concentrations of pore water Hg(II), pH, inorganic sulphides, halides and dissolved organic matter. Formation of HgS(s) and mixed Hg/FeS(s) was considered in models and determined independently by Hg EXAFS and S XANES.

Results and Discussion

The sum of dissolved neutral Hg-sulfide species $[Hg(SH)_2^0(aq)] + [HgS^0(aq)]$ was significantly, positively ($p<0.001$, $n=20$) correlated to K_m at depths of 5-100 cm in brackish water sediments. In contrast, total Hg, total Hg-sulfides or $Hg(SR)_2$ (aq) in pore water gave no significant relationships with K_m. In two sets of freshwater sediments, both neutral Hg-sulfides and total Hg (because of an autocorrelation) gave significant relationships with K_m. Furthermore, the sum of neutral Hg-sulfides in pore water was significantly, positively correlated to total sediment MeHg (µg kg^{-1}) in brackish waters ($p<0.001$, $n=23$), in southern, high-productivity freshwaters ($p<0.001$, $n=20$) as well as in northern, low-productivity freshwater ($p=0.048$, $n=6$). Our results confirm findings from laboratory experiments that concentrations of dissolved neutral inorganic Hg-sulfide species largely control Hg methylation rates, as well as accumulated MeHg concentrations, in contaminated sediments.

References

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Archean granites from the Rum Jungle Complex, Australia

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The 2.55 Ga Rum Jungle Complex (RJC), N Australia, comprises two Archean basement cores, the Rum Jungle and the Waterhouse domes that are surrounded by ortho- and paragneisses of the Palaeoproterozoic Pine Creek Orogen (PCO). The RJC predominantly comprises granite, syenite, quartz-monzonite, and quartz-monzodiorite intruding earlier schists and BIFs. A large number of uranium and base metal deposits have been discovered in black pelites of the PCO neighbouring the granitoid bodies.

Perthitic K-feldspar and quartz are the main minerals present in the granitoids, accompanied by variable amounts of plagioclase (An_{0-23}), magnetite, relict biotite, and accessories such as zircon, monazite, titanite, allanite, apatite, and ilmenite. Most of the samples underwent major post-emplacement mineralogical changes, e.g., formation of white mica at the expense of feldspar, replacement of titanite by rutile, and formation of biotite rims (X_{Mg}: 0.45-0.53) around magnetite. A pervasive alteration is characterised by sulphide mineralisation, chloritization of biotite and white mica, martitization of magnetite, formation of tourmaline, muscovite, and thoro-silicates along micro-shear zones.

The major element data of the granites show well-defined fractionation trends, involving a negative correlation of Si with Ti, Fe, Mg, Ca and P, whereas K increases in the same direction. The granitoids are metaluminous to peraluminous and alkalic to alkali-calcic in composition, the X_{Mg} is generally high (0.14–0.42). The chondrite-normalized trace element and REE plots of all granitoid samples are similar (troughs for Nb, Ta, Sr, P, Ti; $(La/Yb)_{CN}$: 7-115; Eu/Eu*: 0.2-0.8), suggesting that they were derived from the same parental magma. Remarkably, they have conspiciously high Th (8.6-123.3 ppm) and U (2.9-39.9 ppm) compared to the average Upper Crust. All samples, except three highly altered granitoids, can be defined as I-type granites (ASI 0.9-1.1).

Post-emplacement deformation and alteration are associated with secondary, low-salinity (0-14 wt.% NaCl equiv) to high-salinity fluid inclusions (15-38 wt.% NaCl equiv.) in magmatic quartz that display generally moderate homogenisation temperatures (107-384°C and 81-392°C, respectively). High-salinity inclusions also comprise minor gas-rich mixed H_2O-CO_2 inclusions. All fluid inclusions contain a variable number of presumably accidentally trapped solid inclusions. The fluids are suggested to have been derived by de-watering of metasediments of the PCO and to be modified by fluid-rock interactions during transport and metamorphism of the RJC and PCO at c. 1.8 Ga. This process might also be responsible for the formation of U and ore deposits in the vicinity of the RJC.