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Mercury stable isotope fractionation in a tropical ecosystem including human hair: New insights for an isotope balance

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Mercury contamination is an environmental problem in the Amazon basin still relevant today as impacts on human health are poorly studied. In Bolivia, indigenous people have elevated methylmercury concentrations (between 2719 and 23701 ng.g-1) in their hair. This highly toxic molecule is formed after methylation of inorganic Hg released by chemical and physical weathering and from human activities. The aim of our study is to propose a first isotope balance in a Bolivian Amazon ecosystem, through variations in Hg isotopic compositions. The discovery of mass-independent fracionation (MIF) of odd-isotopes in our organic samples (fish and human hair) opened a new way of research in tracing the sources and the processes involved in the cycle of Hg.
Four types of samples are studied: liquid Hg0 from gold mining, sediment samples, fish coming from the Beni River basin (from the main channel and an associated floodplain lake) and hair from gold miners and fish-eating native populations. Hg isotopic compositions were analyzed on a Thermo-Finnigan Neptune MC-ICP-MS at the LMTG after sample digestion by HCl/HNO3 or by H2O2/HNO3 for fish samples, at 120°C.
The 202Hg values (relative to NIST 3133) are signicantly different with respect to the external precision on UM-Almaden#2 of 0.18 %■ (2, n = 42): -0.34 ± 0.02 %■ for liquid mercury, between -1.33 and -0.81 %■ for bottom and floodplain sediments (n=18), between -0.87 and 2.22 %■ for miners hair (n=26), +1.29 ± 0.41 %■ for native hair (n=13) and between -0.91 and -0.21 %■ for fish samples (n=53). A large mass-independent isotope fractionation (MIF) was observed for odd isotope ratios in all hair samples and fish samples whereas weak anomalies were measured for sediment samples:
- Δ199Hg anomaly: -0.12 to -0.04 %■ for sediment, -0.22 to +0.63 %■ for fish samples and +0.13 to +1.63 %■ for hair
- Δ201Hg anomaly: -0.12 to -0.02 %■ for sediment, -0.21 to +0.43 %■ for fish samples and +0.06 to +1.25 %■ for hair.

Both anomalies Δ201Hg vs. Δ199Hg are linearly correlated with a slope of +1.12%■ for native hair and a mass dependant fractionation (MDF) of ∼+2%■ has been evidenced between native communities hair and fish species constituting their diet. For the same fish species, MIF anomalies differ with floodplain lakes and drainage basins suggesting that the methylmercury (MMHg) analysed in fish caught in floodplain lake reflects the photodemethylation process while in the Beni R. mainstream the negative anomalies confirm that this photochemical reactions are limited. Within the aquatic food chain of an hydrosystem, it appears that MIF anomalies can trace the fish ecology and the aquatic photochemical processes, while mass dependent fractionation can trace the trophic level as a result of excretion and metabolic processes involving mercury in the body.