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Quantification of Labile Soil Mercury by Stable Isotope Dilution Techniques

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Mercury (Hg) is a toxic element that can cause severe health problems to humans. Mercury is emitted to the atmosphere from both natural and anthropogenic sources and can be transported over long distances before it is deposited to aquatic and terrestrial environments. Aside from accumulation in soil solid phases, Hg deposited in soils may migrate to surface- and ground-water or enter the food chain, depending on its lability. There are many operationally-defined extraction methods proposed to quantify soil labile metals. However, these methods are by definition prone to inaccuracies such as non-selectivity, underestimation or overestimation of the labile metal pool. The isotopic dilution technique (ID) is currently the most promising method for discrimination between labile and non-labile metal fractions in soil with a minimum disturbance to soil-solid phases. ID assesses the reactive metal pool in soil by defining the fraction of metal both in solid and solution phases that is isotopically-exchangeable known as the 'E-value'. The 'E-value' represents the metal fraction in a dynamic equilibrium with the solution phase and is potentially accessible to plants. This is carried out by addition of an enriched metal isotope to soil suspensions and quantifying the fraction of metal that is able to freely exchange with the added isotope by measuring the equilibrium isotopic ratio by ICP-MS. E-value (mg kg^{-1}) is then calculated as follows:

$$\text{E-Value} = (M_{soil}/\ W)\ (C_{spike}V_{spike}/\ M_{spike})\ ({}^{Iso1}IA_{spike} - {}^{Iso2}IA_{spike}Rss\ /\ {}^{Iso2}IA_{soil}\ Rss\ -\ {}^{Iso1}IA_{soil})$$

where M is the average atomic mass of the metal in the soil or the spike, W is the mass of soil (kg), C_{spike} is the concentration of the metal in the spike (mg L^{-1}), V_{spike} is the volume of spike (L), IA is isotopic abundance, and Rss is the equilibrium ratio of isotopic abundances (Iso1:Iso2).

Isotopic dilution has been successfully applied to determine E-values for several elements. However, to our knowledge, this method has not yet been applied to estimate the labile pool of mercury in contaminated soils. We performed a series of soil incubations spiked with ${}^{196}Hg^{2+}$aiming at measuring and modelling the progressive assimilation of Hg ions into less labile forms. Soils with a wide range of characteristics are taken for our research purpose, inclusive of Hg concentrations ranging from 0.1 to 390 mg kg^{-1}, pH between 3.5 - 7.5 and total organic carbon (TOC) between 2.5 – 8 %. In parallel, the labile pool of Hg estimated using ID will be compared with that determined using conventional extraction methods, e.g. sequential extraction procedures. These altogether allows us to answer (1) how the E-value of Hg in soils is comparable to those estimated based on selective extraction methods, (2) how the labile Hg correlates with the total soil Hg, soil pH and TOC, and (3) how the solubility of added Hg (e.g. via rainfall) decreased in soils of different properties during aging. The obtained results fills the knowledge gap concerning Hg biogeochemistry in the terrestrial environment and serves as a basis for estimating (and predicting) the risk of soil Hg diffusion from a point source to the adjacent environments.