Lead transport and speciation in organic horizons of forest soils

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Anthropogenic emissions during the 20th century resulted in global lead (Pb) contamination of soils. Remote forests in New England received 1 to 4 g Pb m-2, with higher amounts deposited at montane sites. We present isotopic and soil time-series data demonstrating that forest floor horizons (e.g., the upper 5 to 9 cm of organic material) retain atmospherically-delivered Pb in excess of 50 years. Despite their role in retaining lead, the precise nature of Pb partitioning within this organic-rich matrix remains elusive. Selective chemical extractions and x-ray absorption spectroscopy suggest that Pb is frequently bound to organics; however, a significant proportion of the Pb appears to be associated with inorganic phases. Extractions with sodium hydroxide and $Na_2P_4O_7$, two commonly used treatments for targeting organically-bound metals released 50 to 100% of the Pb, but the specificity of these reagents to organic matter are suspect. Selective chemical extractions with the reducing agent hydroxylamine hydrochloride released up to 40% of the Pb, but liberated negligible organic matter. Lead released during chemical extractions coincided with iron release. We conclude that despite very high organic matter contents (>80%) a significant proportion of the lead in forest floor horizons may associated with inorganic and possibly “reducible” mineral phases.

A potentiometric and ^{113}Cd NMR study of cadmium complexation by natural organic matter at two different magnetic field strengths

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The binding of cadmium(II) to Suwannee River natural organic matter (NOM) has been investigated across a broad range of molar Cd/C ratios (0.00056 – 0.0056) and pH values (3.5 – 11) by ^{113}Cd NMR spectroscopy at two magnetic field strengths (B_0 = 9.4 and 11.2 T). As a result of the very peculiar and highly complex characteristics of the Cd-NOM exchanging system, these ^{113}Cd NMR spectra are characterized by a pH- and concentration-dependent superposition of slow, intermediate and fast chemical exchange. Detailed insight into the complex interplay of solution chemistry and chemical exchange is provided by a thorough mapping of this Cd-NOM chemically exchanging system through acquisition of more than 100 ^{113}Cd NMR spectra at two magnetic field strengths and with systematic variation of Cd/C ratios and pH values. The interpretation of ^{113}Cd NMR spectra is greatly facilitated and constrained by simultaneous measurements of pH and pCd, which allows a model-independent calculation of organically bound Cd(II) under all experimental conditions. Under the conditions applied in this study, Cd(II) has been found to bind to oxygen, nitrogen, and, to a much lesser extent, sulfur ligands of NOM.