

XAFS determination of the chemical form of lead in smelter-contaminated soils and mine tailings: Importance of adsorption processes

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ABSTRACT

We investigated smelter-contaminated soils from Evin-Malmaison, Nord-Pas-de-Calais, France, and mine tailings from Leadville, Colorado, U.S.A. Bulk Pb concentrations range from 460 to 1900 ppm in the topsoils at Evin-Malmaison site and from 6000 to 10000 ppm in the tailings samples from the Leadville site. These concentrations necessarily raise human health and environmental concerns, but bioavailability and chemical lability of Pb in these materials vary dramatically and show little correlation with bulk concentrations. This study provides detailed information on the speciation of Pb in these materials. Emphasis is on the identification and characterization of poorly crystalline and/or fine-grained species, such as sorption complexes and poorly crystalline (co)precipitates, which are likely to control Pb bioavailability and mobility in these natural systems.

Because these samples are heterogeneous, multi-phase mixtures, a variety of bulk analytical methods were used including powder X-ray diffraction (XRD) and Rietveld refinement, scanning electron microscopy, electron probe microanalysis, synchrotron-based X-ray absorption, and micro-fluorescence spectroscopies. The synchrotron-based techniques enable identification of amorphous or nanocrystalline Pb-containing phases and the spatial distribution of Pb at the 25 μm scale.

These techniques, in conjunction with physical and chemical separation techniques, allowed identification and characterization of several species not amenable to detection by conventional microanalytical techniques. In the Evin-Malmaison samples, direct spectroscopic evidence for Pb sorbed to humic acids was found, as well as to both manganese and iron (oxyhydr)oxides. In the Leadville samples, variations in Pb speciation with pH are consistent with predictions based on simplified model system studies of adsorption processes; specifically, the carbonate-buffered tailings with near-neutral pH contain up to 50% of total Pb as adsorption complexes on iron (oxyhydr)oxides, whereas Pb speciation in sulfide-rich low pH samples is dominated by Pb-bearing jarosites with no evidence for adsorbed Pb in these latter samples.