Distinguishing natural from anthropogenic sources of arsenic and interpreting post-depositional change in soils and sediments

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Fifty years of roasting gold ore near Yellowknife, NWT, Canada has converted arsenopyrite into arsenic trioxide, a more toxic, soluble and bioaccessible form of arsenic (As). A second roaster product is spongy As-bearing iron oxide pseudomorphs of the original arsenopyrite. Limited emission controls during mining resulted in widespread aerial distribution of 20,000 tonnes of As to soils and sediments in the region. Core sampling in areas previously considered undisturbed by mining activities revealed high concentrations of As. For soils, the highest concentrations (maximum ~8000 mg/kg) were in the top 10 cm, and for sediments the highest (maximum ~3000 mg/kg) were a few cm below the sediment-water interface. Synchrotron-based microanalysis and scanning electron microscopy were used to distinguish natural from anthropogenic As, and identify post-depositional transformations. Arsenic trioxide has persisted in near-surface peaty soils, exhibiting limited dissolution resulting in As-rich pore waters and resorption on organic material and Fe-Mn oxyhydroxides. There is little evidence of natural As in the upper 20 cm of soil. Arsenic trioxide also persists in near-surface stream and lake sediments although the abundance of arsenic sulfide suggests post-depositional conversion of arsenic trioxide to arsenic sulfide, a less bioaccessible form likely to be more stable in oxygen-poor, organic sediments.