Importance of inherited Fe oxides on toxic (As, Cd, Pb) and potentially toxic metals (Cu, Ni, Zn) from grey shale-derived soils on geologic timescales

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Shale is an important sedimentary rock in the eastern United States and globally due to its spatial abundance and potentially elevated trace element concentrations under shallow, low energy coastal depositional environments. Shale-derived soils inherit organic matter and oxides (Fe and Mn) which can promote accumulation and retention of both geogenic and exogeneous trace metals. This is particularly important as many shale environments are exposed to elevated deposition of trace element pollution from combustion of fuels, local smelting and metal works, and, industrial activities for energy extraction and manufacturing. Shale-derived soils have developed on the geologic time scale of recent glaciation (10 to 20 kya) to earlier Pleistocene peri-glacial action (100 to 200 kya). Pedogenesis promotes the formation of neoformed clay and secondary oxides that can promote sorption of trace elements in shale-derived soils. Here, we utilized a chronosequence of grey-shale derived soils to determine if the trace elements present in these soils are inherited and if pedogenesis has enhanced the trace element sorption capacity of these soils. We focused in on trace elements associated with anthropogenic activities: arsenic (As), cadmium (Cd), copper (Cu), nickel (Ni), lead (Pb), and zinc (Zn) due to their potential impacts on water resources and aquatic ecosystems. Titanium-based tau values show that shale-derived soils are net accumulators of Cd, Cu, Ni, and Pb for most sites across the chronosequence, despite the variation in proximity to human pollution sources. Considering sorption of the metals, $< 5\%$ of the metals were in oxidizable forms (e.g. organic matter bound) but 20 to 60% of Cu and Cd and 5 to 20% of Ni and Pb were within Fe oxides in both soils and shale bedrock samples. This suggests that a combination of inherited and neoformed oxides are retaining an important fraction of toxic and potentially toxic metals. These results imply that soils derived from grey-shale are net accumulators of metals from exogeneous sources and retain their geogenic trace elements.