Poster #21-69

Phosphorus Speciation in Atmospherically Deposited Particulate Matter and Potential Impact on Terrestrial Soil Nutrient Cycling and Ecosystem Productivity

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BER Program: SBR
Project: University Award

An accepted paradigm of terrestrial ecosystems in temperate climates is that nitrogen (N) rather than phosphorus (P) is the dominant limiting nutrient to plant growth. Recent studies, however, suggest that the anthropogenic release of large quantities of N-oxides into the atmosphere through fossil fuel combustion has greatly enhanced N inputs to ecosystems, such that the bioavailability of P rather than N may now control terrestrial productivity. To investigate P speciation, sources, seasonal changes, and potential bioavailability of atmospherically deposited P to soil in mountain ecosystems, particulate matter (PM) was collected by passive sampling at high elevation sites in the East River Watershed (CO) study area (in collaboration with the LBNL Watershed Function SFA), and at high and low elevation sites in the Southern Sierra Critical Zone Observatory (SSCZO, CA) during two seasonal periods (Sept.-Oct. 2016 and April-Sept. 2017). Sized-fractioned PM samples were analyzed by bulk and microfocused XANES at the P K-edge. Aqueous extractions of PM were studied by 31-P NMR. Analysis of lead isotopes provided information about PM sources. Results from linear combination fits of XANES spectra indicated a mixture of organic and inorganic P species, with organic-P dominating samples from high elevation sites at both CA and CO (~80-95% of total P). Fit results showed that organic P was a mixture of either bulk or adsorbed inositol hexakisphosphate (IHP, also known as phytic acid or phytate), commonly derived from plants, and DNA-P, mostly as an adsorbed species. Results from NMR confirmed the presence of DNA-P in the soluble fraction in addition to monoester-P, pyrophosphate, and inorganic orthophosphate. The lower elevation SSCZO site had a higher fraction of inorganic P (~30-45%), mostly as hydroxyapatite or Ca-associated P. For comparison, soil samples collected at the SSCZO sites had concentrations of total P up to three times lower than PM samples, primarily as inorganic P species (Al-phosphate and clay-associated phosphate). Lead isotope ratios indicated strong seasonal differences in long-range PM input from Asian sources to the CO sites, varying from 0 to almost 60% Asian lead between fall and spring, respectively. These results indicate that organic P dominates in high elevation PM samples, and that seasonally variable long-range transport is an important PM source to mountain ecosystems. Identification of DNA-P in PM samples suggests a more reactive and bioavailable form of P than IHP that has not been previously identified as a significant P component of PM.