

Publication

Assessing the origin of sulfate deposition at the Hubbard Brook Experimental Forest

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The geographical and chemical origin of SO42- deposition has become a concern, because anthropogenic S emissions have influenced the biogeochemistry of forested ecosystems and surface waters. Our aim was to evaluate the origin of SO42- in bulk precipitation at the Hubbard Brook Experimental Forest (HBEF), New Hampshire. We analyzed 26 years of archived bulk precipitation samples for sulfur stable isotopes. We compared the delta(34)S values with anthropogenic SO2 emissions, the relative contribution of sea salt aerosols (as the SO42-/Na+ ratio in precipitation), and temperature and solar radiation effects on the long-term patterns of delta(34)S values. The long-term pattern of delta(34)S values in bulk precipitation could be explained partly by the relative contribution of marine SO42- or solar radiation but not by temperature variation or anthropogenic SO2 emissions. The high variability of the delta(34)S values of various fossil fuels makes it difficult to use stable S isotopes for identifying whether changing fossil fuel use is affecting the delta(34)S values in bulk precipitation. The seasonal pattern of delta(34)S values in bulk precipitation (significantly higher values in the winter than the summer) may be explained by the temperature dependence of the isotopic shift during SO42- formation in the atmosphere. A greater relative contribution of marine SO42- during the winter also may have contributed to the higher delta(34)S values in the winter. Previous investigations may have overestimated the role of biogenic emissions in affecting the delta(34)S pattern.

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