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Back-Trajectory Analysis and Source-Receptor Relationships: Particulate Matter and Nitrogen Isotopic Composition in Rainwater

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ABSTRACT

The southeastern portion of North Carolina features a dense crop and animal agricultural region; previous research suggests that this agricultural presence emits a significant portion of the state's nitrogen (i.e., oxides of nitrogen and ammonia) emissions. These findings indicate that transporting air over this region can affect nitrogen concentrations in precipitation at sites as far as 50 mi away. The study combined nitrate nitrogen isotope data with back-trajectory analysis to examine the relationship between regional nitrogen emission estimates independent of pollutant concentration information. In 2004, the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model was used to determine potential sources of nitrogen in rainwater collected at an urban receptor site in Raleigh, NC. The $\delta^{15}N$ isotope ratio signatures of each sample were used to further differentiate between sources of the rainwater nitrate. This study examined the importance of pollution sources, including animal agricultural activity, and meteorology on rainfall chemistry as well as the implications in fine particulate matter ($PM_{2.5}$) formation. Samples that transited the dense crop and animal (swine) agricultural region of east-southeastern North Carolina (i.e., the source region) had lower $\delta^{15}N$ isotope ratios in the nitrate ion (average = -2.1 ± 1.7‰) than those from a counterpart nonagricultural region (average = 0.1 ± 3‰.) An increase in $PM_{2.5}$ concentrations in the urban receptor site (yearly average = 15.1 ± 5.8 $\mu g/m^3$) was also found to correspond to air transport over the dense agricultural region relative to air that was not subjected to such transport (yearly average = 11.7 ± 5.8 $\mu g/m^3$).

IMPLICATIONS

Understanding the source (emission)-receptor (deposition) relationships will provide information that is important for regulators and policy-makers. The potential of the stable isotopes of nitrogen and sulfur to investigate the source-receptor relationships for atmospheric sulfur and nitrogen has long been recognized. By combining the isotopic composition of the major acidic components of atmospheric deposition (nitrogen and sulfur) with information on the air mass source region, it is possible to determine the geographic origin of the nitrogen and sulfur compounds. This paper addresses this important issue involving analyses of regional meteorology, origins of air masses, and rainfall isotopic measurements.

INTRODUCTION

Wet and dry acid deposition is a major concern in the eastern United States. The chemical components of these acids, including nitrogen oxides (NO_x = nitric acid [NO] + nitrogen dioxide [NO_2]), nitrate ions (NO_3^-), ammonia (NH_3), and ammonium ions (NH_4^+), have important effects on rainfall chemistry and fine particulate matter ($PM_{2.5}$) formation. NH_3 may be scavenged from the air by rain, absorbed by plants, or rapidly converted to NH_4^+ aerosol. NO_x is a precursor to nitric acid and can react with NH_3 to produce ammonium nitrate, $NH_4NO_3(s)$.[1] Increases in emissions from anthropogenic sources of NO_x correspond to increases in nitric acid deposition.[2]

Specifically, North Carolina is home to approximately 10 million hogs, which are almost exclusively confined to the southeastern portion of the state. Walker et al.[3] suggest that swine agriculture represents approximately 21% of the state's nitrogen emissions. The study concludes that NH_4^+ concentrations in precipitation at downwind sites were 50% higher on weeks when at least 25% of the precipitation came from the swine agricultural source region. The atmospheric emissions of the source region were substantial enough to influence the nitrogen content of wet deposition at a receptor site over 50 mi away.

A more accurate measurement of animal operations' influence on rainfall chemistry, independent of concentration analysis, was thus pursued because it would prove helpful in further verification of the hypothesis. To this end, we examined the isotopic composition of the nitrogen compounds in rainwater, taking advantage of the severe isotopic depletion associated with agriculturally related biological activities, including nitrification and volatilization. Specific isotopic signatures may allow for differentiation between natural and anthropogenic source contributions.[4]

BACKGROUND

The distribution of light stable isotopes in the environment is controlled by chemical, physical, and biological processes that can be viewed as reversible equilibrium reactions or irreversible unidirectional kinetic reactions. The heavier isotope has a higher disassociation energy,