Geophysical Research Abstracts,
Vol. 10, EGU2008-A-04821, 2008
SRef-ID: 1607-7962/gra/EGU2008-A-04821
EGU General Assembly 2008

Isotopes of nitrate in Greenland ice: a record of recent changes in the global nitrogen cycle

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The global nitrogen cycle has been significantly altered by human activities over the last 200 years. Nitrogen deposition has greatly increased worldwide as a direct result of the increase in ammonia and nitrogen oxide (NOx) emissions, contributing to worsening air and water quality and fundamentally altering the biogeochemical cycling of reduced forms of nitrogen. Indeed, measurements of nitrate in Greenland ice cores show at least a doubling by 1960 compared to background concentrations between 1760 and 1900. It remains difficult, however, to quantify the impact of anthropogenic changes on natural variability in the global N cycle.

The connection between atmospheric concentrations of NOx and nitrate in ice core records is not straightforward, primarily because post-depositional processing can alter nitrate concentrations in snow (see also Jarvis et al. presentation, this meeting). Recent advances in analytical methods have allowed for exploration of the isotopes of nitrate (δ^{15}N, δ^{18}O and δ^{17}O) in ice cores at high resolution. The isotopes of nitrate contain more information than concentration alone, reflecting both the sources and chemistry of NOx. We have generated a record of δ^{15}N and δ^{18}O of nitrate spanning the last 300 years from a 100-meter ice core drilled at Summit, Greenland. Recent work has shown that the oxygen isotopes of nitrate (δ^{18}O, δ^{17}O) reflect the chemical reactions that produce nitrate in the atmosphere, namely the interaction of NOx and oxidants such as ozone and OH. The high δ^{18}O of nitrate in the Greenland ice core ($\sim$40 to 90 per mil vs. VSMOW) reflects the influence of ozone, which typically exhibits a δ^{18}O of $\sim$90-120 per mil vs. VSMOW. Although the δ^{15}N of nitrate may also

be influenced by chemical reactions in the atmosphere, the δ^{15}N found in the Greenland core is clearly impacted by the relatively recent increase in anthropogenic NOx emissions. For example, the mean δ^{15}N of nitrate prior to 1900 is 11 per mil (vs. N_2) in comparison to a mean of 3 per mil in the last 100 years. The ability to distinguish nitrate sources using δ^{15}N has implications for evaluating changes in the nitrogen cycle over time, including diagnosing changes in the biosphere and atmosphere and its connection with changes in climate.