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Photochemical Isotope Effects in Snowpack Nitrate

C. Meusinger (1), T.A. Berhanu (2), J. Erbland (2), F. Domine (2), J. Savarino (2), and M.S. Johnson (1)

(1) Copenhagen Center of Atmospheric Research, Department of Chemistry, University of Copenhagen, Denmark, (2) Laboratoire de Glaciologie et Géophysique de l'Environnement, St Martin d'Heres, France

Sunlight is known to change snowpack nitrate concentrations, but the specific mechanisms are not well characterized preventing the recovery of historical nitrate concentrations. Studies of isotope distributions can distinguish specific processes, potentially giving a powerful tool for interpreting the records. In particular, the reformation of nitrate from its photolysis products is in need of further study.

We present measurements of nitrate concentration changes and the accompanying N and O isotopic fractionations resulting from experiments performed using Antarctic snow illuminated by a Xe lamp. The snow was typically irradiated for several days at -30 °C and flushed at 1 l/min with N_2 to remove gas-phase photolysis products. The N_2 stream was at 100 % relative humidity to prevent physical changes of the snow, which was confirmed by measurements of the snow specific surface area. After illumination the snow column was treated like an ice core and cut into slices that were melted and analyzed using Liquid Chromatography and Isotope Ratio Mass Spectrometry employing the bacteria method.

The δ^{15}N data shows no typical Rayleigh behavior but levels off, showing no stronger enrichment than $\sim$7 ‰ (on top of the initial 34 ‰). These profiles are best explained by the mixing of reactant and product reservoirs of N. The triple O isotope data shows trends similar to those observed in field campaigns indicating a loss of the initial mass independent signal.

Altering the flow direction, and therefore presumably the location of reformed nitrate does not change the profiles of nitrate concentration and isotopes.

A TUV-snow model of the system under question is presented with a discussion of other processes that could possibly also give rise to the observed data.