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Isotopic constraints on the budget of atmospheric nitrate in the Arctic

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Nitrate is an ubiquitous species of the Arctic environment. In the atmosphere, it is the ultimate end-product of nitrogen oxides (NOx) oxidation, and contributes to aerosol acidification and Arctic haze. Stable isotopes provide informations on its budget. While the $^{15}N/^{14}N$ ratio indicates the origin and fate of its precursors, oxygen isotope ratios ($^{18}O/^{16}O$ and $^{17}O/^{16}O$) reveal the influence of ozone, a major atmospheric oxidant, on nitrate formation : ozone possesses a unique oxygen isotope signature (the isotope anomaly, $\Delta^{17}O$, which originates from mass-independent fractionation during the formation of ozone) that is transferred to other species during oxidation reactions. We present results gathered from three field campaigns in the high Arctic (Alert, Nunavut, 2004, Barrow, Alaska, 2005 and Ny Ålesund, Svalbard, 2006), focusing on the isotopic composition of atmospheric nitrate and its link with the seasonal snowpack. Additional insights from a year-round survey of the oxygen and nitrogen isotopic composition of atmospheric nitrate at Alert, Nunavut, are presented.

The correlation between the oxygen isotope anomaly of atmospheric nitrate and the ozone levels during ozone depletion events (ODEs), occurring in the Arctic atmosphere in spring, indicates that local processes, namely the halogen chemistry responsible for ODEs, have a major influence on the budget of NOx and nitrate. Indeed, due to its high reactivity with NO, BrO competes with ozone and peroxy radicals in terms of NO oxidation, thus playing a substantial role in transferring its isotope anomaly (also inherited from ozone). In addition, isotope measurements point towards a signif-

icant (and even dominant) influence of $BrONO_2$ and N_2O_5 hydrolysis, in comparison with the 'classical' $OH+NO_2$ reaction pathway, to form atmospheric nitrate from NOx in these conditions.

At Ny Ålesund in 2006, virtually no ODEs occurred and weather conditions were unusually mild with no sea-ice formation in the vicinity of the sampling site. The isotope anomaly of atmospheric nitrate was remarkably steady, a result fully consistent with the correlation presented above. Simultaneous measurements carried out on surface snow nitrate reveal similar isotope ratios in both media, for both nitrogen and oxygen isotopes. This suggests that, in these conditions, snowpack nitrate originated from the atmosphere and did not undergo strong post-depositional recycling and reemission until snowmelt, which would have induced isotopic fractionation and altered the isotopic composition of the snowpack left-over nitrate. This has to do with the high sea-salt burden (hence, alkalinity) of the snow surface, that seems to have prevented significant NOx and HONO emissions for most days.