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Tracing the sources and fate of nitrogen oxides in a polar urban environment: an isotopic survey during the ALPACA campaign

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Urban polar areas can be subject to severe air pollution in winter, exacerbated by sharp temperature inversions very effective at trapping pollutants near to the surface. However, the formation of secondary aerosols in these cold and dark conditions and the role of the arctic boundary layer are still poorly understood. To address this issue, a major international measurement campaign was conducted in January/February 2022 in and around Fairbanks, Alaska, as part of the CASPA (Climate-relevant Aerosol Sources and Processes in the Arctic) and ALPACA (ALaskan Pollution And Chemical Analysis) projects. Among the various atmospheric chemical and physical measurements, gaseous and particulate samples were collected and subjected to isotopic analyses in order to identify the sources of nitrogen oxides (NO_x) emissions and understand how they are oxidized to atmospheric nitrate (NO_3^-).

The use of stable isotopes has demonstrated its ability to provide information relevant for tracing emission sources, individual chemical processes and budgets of atmospheric trace gases. Of particular interest is the propagation of the ozone oxygen-17 anomaly ($\Delta^{17}O$) into the reactive nitrogen cycle which has led to a better understanding of NO_3^- formation pathways in various environments. However, there remain some difficulties to interpret NO_3^- isotopic composition, mainly due to the lack of clearly established understanding about the link between the oxygen and nitrogen isotopic composition of the NO_x and the chemical state of the atmosphere.

Here, we present the results of the multi-isotope composition of atmospheric NO_2 collected on denuder tubes (n = 33) in downtown Fairbanks. We find significant oscillations in $\Delta^{17}O$ and $\delta^{15}N$ values. The maximum $\Delta^{17}O$ value is recorded during the day (39.7 ‰), and the minimum value at night (3.6 ‰). $\Delta^{17}O$ values are significantly different during the day and night, with a mass-weighted mean of 30.8 and 11.5 ‰, respectively. Conversely, $\delta^{15}N$ values show no diurnal trend and range from -10.2 to 24.1 ‰ (mass-weighted mean of (12.3 ± 11.1). By collating atmospheric observations (e.g., NO, NO_2, O_3,) and $\Delta^{17}O/\delta^{15}N$ data, we investigate the sources and fate of NO_x during winter in Fairbanks. Implications for the isotopic composition of nitrate are also discussed.