Seasonal patterns of atmospheric N$_2$O isotopic composition observed at the alpine station Jungfraujoch, Switzerland

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Nitrous oxide (N$_2$O) is a strong greenhouse gas and the strongest ozone-depleting substance emitted in the 21$^{st}$ century. The substantial increase in atmospheric N$_2$O mixing ratio since the preindustrial era has raised worldwide concern. This has been largely attributed to enhanced anthropogenic N$_2$O emissions (e.g. agriculture sources). However, due to the long life time of N$_2$O (~ 120 years) in the atmosphere, spatial and temporal gradients are small, which makes it difficult to distinguish sources and develop mitigation strategies. Also, the factors governing seasonal and interannual variabilities in N$_2$O mixing ratio and growth rate remain poorly understood. Recently, isotope measurements have provided additional constraints on the global N$_2$O cycle, based on distinct isotopic signatures of N$_2$O produced from various sources. However, long-term isotopic measurement with high precision is still limited.

In this study, we measured N$_2$O mixing ratio and isotopic composition in tropospheric air sampled at the high-altitude Jungfraujoch (JFJ) research station, from 2014 to 2017. N$_2$O mixing ratios were determined on-line by OA-ICOS, while discrete air samples for isotopic analysis were collected weekly/bi-weekly and analysed subsequently using quantum cascade laser absorption spectroscopy at Empa with overall repeatability below 0.1‰. Samples were classified according to their associated air mass origin (free tropospheric versus boundary layer). The observed rise in N$_2$O mixing ratios at JFJ is comparable to the global average (0.73 ppb y$^{-1}$). $\delta^{15}$N$^{bulk}$ of N$_2$O decreased over the study period, likely due to $^{15}$N-depleted anthropogenic sources. However, we observed no clear interannual tendency for $\delta^{15}$N site preference (SP) and $\delta^{18}$O of N$_2$O. Surprisingly, our dataset demonstrated significant seasonal patterns for $\delta^{15}$N$^{bulk}$ and SP, which might reflect seasonal variation in N$_2$O production processes in the Northern Hemisphere. For example SP was generally lowest in summer (July to September) and highest in winter (January). This can be explained by a larger contribution of N$_2$O produced by biological denitrification, with low SP in cold, dry winters, as compared to nitrification, which dominates during warm, humid summers. Stratospheric intrusion of isotopically enriched air could be another contributor to the observed seasonal pattern. Our dataset is complemented with N$_2$O isotopic measurements in the Southern Hemisphere (Cape Grim Air Archive), to investigate hemispheric difference and therefore to enhance our knowledge of the global N$_2$O cycle.