



Seasonal patterns of atmospheric N₂O isotopic composition observed at the alpine station Jungfraujoch, Switzerland

Longfei Yu (1), Eliza Harris (1,2), Sarah Eggleston (1), Erkan Ibraim (1), Stephan Henne (1), Martin Steinbacher (1), Lukas Emmenegger (1), Christoph Zellweger (1), and Joachim Mohn (1)

(1) Swiss Federal Laboratories for Materials Science and Technology (EMPA), Laboratory for Air Pollution / Environmental Technology, Duebendorf, Switzerland (longfei.yu@empa.ch), (2) Institute of Ecology, University of Innsbruck, Austria

Nitrous oxide (N₂O) is a strong greenhouse gas and the strongest ozone-depleting substance emitted in the 21st century. The substantial increase in atmospheric N₂O mixing ratio since the preindustrial era has raised worldwide concern. This has been largely attributed to enhanced anthropogenic N₂O emissions (e.g. agriculture sources). However, due to the long life time of N₂O (~ 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 N₂O mixing ratio and growth rate remain poorly understood. Recently, isotope measurements have provided additional constraints on the global N₂O cycle, based on distinct isotopic signatures of N₂O produced from various sources. However, long-term isotopic measurement with high precision is still limited.

In this study, we measured N₂O mixing ratio and isotopic composition in tropospheric air sampled at the high-altitude Jungfraujoch (JFJ) research station, from 2014 to 2017. N₂O 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 N₂O mixing ratios at JFJ is comparable to the global average (0.73 ppb y⁻¹). $\delta^{15}\text{N}^{bulk}$ of N₂O decreased over the study period, likely due to ¹⁵N-depleted anthropogenic sources. However, we observed no clear interannual tendency for $\delta^{15}\text{N}$ site preference (SP) and $\delta^{18}\text{O}$ of N₂O. Surprisingly, our dataset demonstrated significant seasonal patterns for $\delta^{15}\text{N}^{bulk}$ and SP, which might reflect seasonal variation in N₂O 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 N₂O 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 N₂O isotopic measurements in the Southern Hemisphere (Cape Grim Air Archive), to investigate hemispheric difference and therefore to enhance our knowledge of the global N₂O cycle.