

3.050 Long-term trend of tropospheric N₂O isotopocule ratios in the Northern Hemisphere .

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Abstract:

Nitrous oxide (N₂O) is one of the increasing greenhouse gases and is the most important stratospheric ozone-depleting gas emitted in the present century. Isotopocule ratios of N₂O, which include not only elemental ¹⁵N/¹⁴N and ¹⁸O/¹⁶O ratios but also site-specific ¹⁵N/¹⁴N ratio in asymmetric NNO molecule, are regarded as useful parameters to infer the origin and production-consumption mechanisms of N₂O, and to estimate its global budget. Previous studies on the firn air in polar ice sheet revealed the secular trend of isotopocule ratios, but there have been only a few reports on long-term monitoring of atmospheric N₂O isotopocule ratios in the Northern Hemisphere.

We present up to 19-year record of monthly or biweekly mixing ratio and isotopocule ratios of N₂O obtained at three sites in the Northern Hemisphere: Hateruma, a southwestern island of Japan (24°N, 124°E) (since 1999), Novosibirsk in the western Siberia, Russia (55°N, 83°E) (since 2005), and Churchill, northern Canada (59°N, 94°W) (since 2011). Results show that the bulk nitrogen isotope ratio (d¹⁵N^{bulk}) are decreasing at the similar rate (about -0.04‰ yr⁻¹) as reported by firn-air analysis while the N₂O mixing ratio are increasing (about 0.8 ppbv yr⁻¹) at the three sites. This suggests isotopically light N₂O sources such as agriculture are still contributing to the increase in the atmospheric N₂O. Detailed analysis of the time series reveals that year-to-year variation of the mixing ratio and d¹⁵N^{bulk} has been enhanced since around 2010 at all the three sites, and that vertical gradient of the mixing ratio and d¹⁵N^{bulk} over Novosibirsk has been also variable recently. Cause of these findings will be discussed with respect to temporal change in N₂O flux and isotopic signature of surface sources and change in atmospheric circulation including troposphere-stratosphere exchange, with the aid of atmospheric model simulation.