Recent trend of atmospheric nitrous oxide isotopocules in the Northern Hemisphere

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Nitrous oxide (N₂O) is one of the increasing greenhouse gases in the troposphere and is the most important stratospheric ozone-depleting gas emitted in the 21st century. In the Northern Hemisphere, sources of atmospheric N₂O include human activity in Europe, Russia, Asia, and North America (e.g., agriculture), biomass burning (forest fires), oceans, and future climate change might cause substantial change in such sources. 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 N₂O trapped in the firn 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 have been measuring mixing ratio and isotopocule ratios of N₂O at one site in the low latitude and two sites in the high latitude of the Northern Hemisphere. At Hateruma, a southwestern island of Japan (24°N, 124°E), monthly air samples are collected into glass flasks at 46 m above sea level, and N₂O isotopocule analysis has been conducted since 1999. At Novosibirsk in the western Siberia, Russia (55°N, 83°E), monthly samples are collected at altitudes of 500 m and 7000 m by aircraft, and N₂O isotopocule data have been obtained since 2005. At Churchill, northern Canada (59°N, 94°W), surface air samples are collected biweekly, and N₂O isotopocule analysis has been conducted bimonthly since 2011.

Results show that the bulk nitrogen isotope ratio (d¹⁵Nbulk) 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 d¹⁵Nbulk has been enhanced since around 2010 at all the three sites, and that vertical gradient of the mixing ratio and d¹⁵Nbulk 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.

Keywords: Greenhouse gas, Troposphere, Stable isotope analysis