Recent trend of atmospheric nitrous oxide isotopocules in the Northern Hemisphere

*Sakae Toyoda[1], Yuma Watanabe[2], Toshinobu Machida[3], Yasunori Tohjima[3], Motoki Sasakawa[3], Shinji Morimoto[4], Doug Worthy[5], Kentaro Ishijima[6], Naohiro Yoshida[1,7]

1. School of Materials and Chemical Technology, Tokyo Institute of Technology, 2. Interdisciplinary Graduate School of Science and Technology, Tokyo Institute of Technology, 3. National Institute of Environmental Studies, 4. Center for Atmospheric and Oceanic Studies, Graduate School of Science, Tohoku University, 5. Environment Canada, 6. Japan Agency for Marine-Earth Science and Technology, 7. Earth-Life Science Instute, Tokyo Institute of Technology

Nitrous oxide (N_2O) 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 N_2O 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 N_2O, which include not only elemental $^{15}N/^{14}N$ and $^{18}O/^{16}O$ ratios but also site-specific $^{15}N/^{14}N$ ratio in asymmetric NNO molecule, are regarded as useful parameters to infer the origin and production–consumption mechanisms of N_2O, and to estimate its global budget. Previous studies on N_2O 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 N_2O isotopocule ratios in the Northern Hemisphere.

We have been measuring mixing ratio and isotopocule ratios of N_2O at one site in the low lattitude and two sites in the high lattitude 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 N_2O 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 N_2O isotopocule data have been obtained since 2005. At Churchill, northern Canada (59˚N, 94˚W), surface air samples are collected biweekly, and N_2O isotopocule analysis has been conducted bimonthly since 2011.

Results show that the bulk nitrogen isotope ratio ($d^{15}N^{bulk}$) are decreasing at the similar rate (about -0.04‰ yr^{-1}) as reported by firn-air analysis while the N_2O mixing ratio are increasing (about 0.8 ppbv yr^{-1}) at the three sites. This suggests isotopically light N_2O sources such as agriculture are still contributing to the increase in the atmospheric N_2O. Detailed analysis of the time series reveals that year-to-year variation of the mixing ratio and $d^{15}N^{bulk}$ has been enhanced since around 2010 at all the three sites, and that vertical gradient of the mixing ratio and $d^{15}N^{bulk}$ over Novosibirsk has been also variable recently. Cause of these findings will be discussed with respect to temporal change in N_2O 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

