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## Three-phased growth of atmospheric N<sub>2</sub>O concentration over the last century

### Content

Nitrous oxide (N<sub>2</sub>O) is the third important anthropogenic greenhouse gas after carbon dioxide and methane. However, its temporal changes in magnitude and geographic distribution of different sources are not well constrained. To better understand the dynamics of atmospheric N<sub>2</sub>O over the last century, we reconstructed the mixing ratio and stable isotope compositions of N<sub>2</sub>O ( $\delta^{15}\text{N}_{\text{bulk}}$ ,  $\delta^{18}\text{O}$  and  $\delta^{15}\text{N}_{\text{SP}}$ ) from ice cores, firn air, and modern atmospheric air. Especially, we took advantage of a new firn air records from the Styx glacier, Antarctica. We also updated firn air transport modeling and improve the ages and chemical compositions. The composite reconstruction shows a three-phased growth of atmospheric N<sub>2</sub>O during the last century, where the growth rate during 1966–2021 CE (P3) was ~ 3 and 5 times higher than during 1850–1930 CE (P1) and 1931–1965 CE (P2), respectively. The  $\delta^{15}\text{N}_{\text{bulk}}$  and  $\delta^{18}\text{O}$  values of the N<sub>2</sub>O show a decreasing trend during the last century. Overall, a significant trend has not been observed in  $\delta^{15}\text{N}_{\text{SP}}$  data. However, the increasing trend in  $\delta^{15}\text{N}_{\text{SP}}$  values since 2000 CE suggests microbial N<sub>2</sub>O emission was dominated by nitrification over denitrification processes. A two-box model calculation suggests that the total N<sub>2</sub>O flux (FT) at 2015 CE was  $17.6 \pm 1.1 \text{ TgN yr}^{-1}$ , where flux from the natural (FN) and anthropogenic (FA) sources were ~ 60% and 40% of FT, respectively. The estimation also indicates that the FA was ~30% of the FT at 1900 CE and suggests rising contribution from FA to the FT over last century. The anthropogenic emission from the continental region were 14%, 23% and 77% of FA during P1, P2 and P3 phases, respectively and indicates that the continental region has been the hot spot of anthropogenic emission for the last four decades. The estimated  $\delta^{15}\text{N}_{\text{bulk}}$ ,  $\delta^{18}\text{O}$  and  $\delta^{15}\text{N}_{\text{SP}}$  values of the anthropogenic emission were  $-13.1 \pm 3.7\text{‰}$ ,  $30.1 \pm 3.6\text{‰}$  and  $11.4 \pm 3.9\text{‰}$ , respectively during 2015 CE. Our new reconstructions provide the most comprehensive view of N<sub>2</sub>O trends during the last ~100 years, with an implication for understanding anthropogenic impact on the global nitrogen cycle and greenhouse gas radiative forcing.

**Primary author:** Dr GHOSH, Sambit (Seoul National University)

**Co-authors:** Prof. TOYODA, Sakae (Department of Chemical Science and Engineering, School of Materials and Chemical Technology, Tokyo Institute of Technology, Yokohama, Japan); Prof. BUIZERT, Christo (College of Earth, Ocean and Atmospheric Sciences, Oregon State University (OSU), Corvallis, OR, USA); JANG, Youngjoon (Seoul National University); Prof. ETHERIDGE, David (CSIRO, Aspendale, Australia); Dr TANS, Pieter (NOAA, Boulder, USA); Prof. YOSHIDA, Naohiro (Department of Chemical Science and Engineering, School of Materials and Chemical Technology, Tokyo Institute of Technology, Yokohama, Japan); AHN, Jinho (Seoul National University)

**Presenter:** AHN, Jinho (Seoul National University)

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