

Estimation of the Variation of Atmospheric Composition and Its Effect on the Climate Change by using a Chemical Transport Model

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An online chemical transport model that can estimate the global distributions of N₂O isotopomers in the stratosphere and the troposphere has been developed. Fractionation of isotopes during the photolysis and chemical reactions was estimated by using the temperature and radiative flux estimated in the model. To estimate the global distribution of N₂O and its isotopomers, 20-years calculations have been executed. The isotopomers fractions in the emissions were estimated by assuming the isotopic fractions in various sources. The model well reproduced the vertical gradient of N₂O concentration and the isotopomers fractions observed at Sanriku, Kiruna, and Showa-station. The averaged value of the isotopomers fraction in the model was close to that from the emissions from the anthropogenic emissions, especially cultivated soil.

Keywords: greenhouse gases, isotopes, general circulation of atmospheric composition, chemical transport model

1. Introduction

Nitrous oxide (N₂O) in the troposphere is an important greenhouse gas anthropogenically enhanced since industrialization starting in the 19th century, accounting for about 5% of radiative forcing in the troposphere except water vapor (Forster et al., 2007). N₂O is also regarded as the most influential ozone depleting substance (ODS) being an exclusive source of nitrogen oxide (NO) in the stratosphere in this century (Ravishankara et al., 2009). However, it is difficult to immediately reduce N₂O emission because the main source of anthropogenic N₂O is agricultural activities (e.g. fertilization) to feed increasing population on the globe and the atmospheric lifetime of N₂O is more than a hundred years (114 years) (Forster et al., 2007). Quantitative details of N₂O emission from various natural and anthropogenic sources are still quite uncertain on regional to global scales despite much effort of the bottom-up and top-down estimations in the past. N₂O also has five different isotopologues with the mass of 44, 45, 46, 47 and 48, and some isotopomers in each isotopologues. The isotopomers are very useful to know origin of the N₂O, because individual isotopomers show different behaviors in biogeochemical processes, due to differences of the mass and of the molecular location. ¹⁴N¹⁴N¹⁶O, ¹⁴N¹⁵N¹⁶O, ¹⁵N¹⁴N¹⁶O, and ¹⁴N¹⁴N¹⁸O account more than 99.9% of all N₂O species. There are many observation studies for them, but only a few global modeling studies to understand the observed results. In this

study, we simulated these four isotopomers as different tracers for understanding N₂O behaviours in both the stratosphere and the troposphere, using an atmospheric general circulation model driven chemistry transport model.

2. Model description

We have developed an atmospheric chemical transport model (ACTM) based on the CCSR/NIES/FRCGC atmospheric general circulation model (AGCM) to simulate atmospheric N₂O concentration and its isotopes. A detailed and consistent meteorology as represented by the grid- and subgrid-scale processes, surface processes (e.g. PBL height and mixing), above-PBL dynamics (e.g. convection) were generated by the AGCM. The meteorological field in the AGCM was assimilated with the European Center for Medium-range Weather Forecasts (ECMWF) 40 year reanalysis and National Centers for Environmental Prediction (NCEP) version 2 reanalysis data via nudging for the comparison of temporal variations of N₂O and its isotopologues with ground-based and air-borne observations. We use T42 truncation in the horizontal and 67 vertical sigma-pressure layers up to 90 km for the present study. Feedbacks between dynamics-radiation-chemistry are taken into account in the model. For example, the photolysis rate of N₂O is estimated by using the shortwave radiative flux calculated in the radiation scheme of the model, and the radiation flux is calculated with cloud distribution and temperature in each

model time step (typically 20 minutes).

N_2O ($^{14}N^{14}N^{16}O$) inventory consists of three components of anthropogenic (Emission Database for Global Atmospheric Research (EDGAR) version 3.2 Fast Track 2000, annual mean), ocean (Global Emission Inventory Activity (GEIA), monthly mean) and soil (EDGAR version 2.0, annual mean) emissions. Inventories for isotopomers were estimated by applying isotopic values of anthropogenic and natural sources by Röckmann et al. (2003) and Ishijima et al. (2007). Fractionation coefficients in photolysis were incorporated, using experimental results by Kaiser et al. (2002, 2003) and von Hessberg et al. (2004). For fractionation in oxidation, coefficient values by Kaiser et al. (2002) were implemented. Dependencies on wavelength and temperature were considered for all isotopic fractionations. Parameterization lines for fractionation constants, which were originally calculated following to von Hessberg et al. (2004) with a factor of 1.2 for the better representations of observational variations within the range of uncertainty of the fractionation parameterisation.

3. Result and discussion

We have prepared two emission scenarios and two initial values for each isotopomers: each isotopomer has four historical cases in a model run to cover ranges of observed N_2O concentration and isotopomer ratios at the Neumayer station. Isotopomer ratios of $^{14}N^{15}N^{16}O$, $^{15}N^{14}N^{16}O$, $^{14}N^{14}N^{18}O$ are denoted by $\delta^{15}N^{central}$, $\delta^{15}N^{terminal}$, and $\delta^{18}O$ in permil unit (‰), respectively. Here, $\delta = (R/R_{reference} - 1) \times 10^3$ ($R = [N_2O]/[N_2O]_{standard}$). For the estimation of source of emissions, following denotations are also used; $\delta^{15}N^{Bulk} = (\delta^{15}N^{central} + \delta^{15}N^{terminal})/2$ and $SP = \delta^{15}N^{central} -$

$\delta^{15}N^{terminal}$.

Vertical profiles of observed and modeled concentration of N_2O and ratio of isotopomers at three stations (Sanriku, Kiruna, and Showa) are shown in Fig. 1. General features of N_2O concentration (*e.g.* vertical gradient) observed using balloon are well captured by the model, but fine structures are not well simulated. For example, the model cannot reproduce steep decrease of N_2O concentration at around the height of 20km at Sanriku, which was corresponding with the eventual intrusion of upper stratospheric air. The difference of meteorological field (NCEP2 and ERA40) does not strongly affect the vertical profiles of N_2O concentration. Model also roughly reproduces observed isotopomer ratios. Rayleigh plots show that N_2O isotopic fractionations caused by stratospheric chemical reactions are well simulated, although the fractionation of $\delta^{15}N^{terminal}$ seem to be underestimated in almost cases. Model significantly underestimates the concentration and the fractionation over Kiruna, indicating the air-age is younger than real in the polar vortex in boreal winter, which is not realistically reproduced by this model.

$\delta^{15}N^{Bulk}$ and SP estimated in this study is shown in Fig. 2, with the observed values from various sources (Toyoda et al., 2004). It seems that $\delta^{15}N^{Bulk}$ and SP for global total N_2O sources estimated in this study are reasonably in the range of measured values for land sources (temperate soil and cultivated soil). It is possible that model overestimate isotopomer ratios in the global total sources, because model tends to overestimate heavier isotopomers losses in the stratosphere, which causes underestimation in isotopomer ratios.

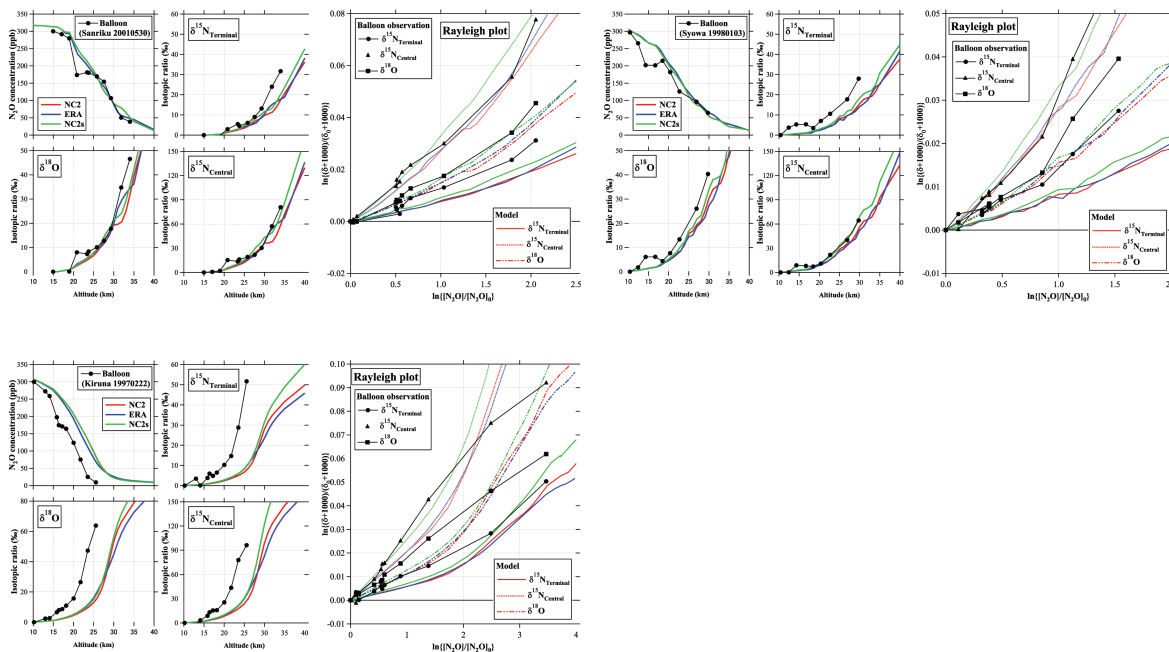


Fig. 1 Comparison of N_2O concentration, $\delta^{15}N^{terminal}$, $\delta^{15}N^{central}$ and $\delta^{18}O$, and Rayleigh plots of the isotopomers between balloon observations over Sanriku (39N, 142E, Japan), Kiruna (68N, 20E, Sweden) and Syowa (69S, 40E, Antarctica), and the model results. NC2, ERA and NC2s represent simulation assimilated with NCEP2, ERA-40 and NCEP2+ fractionation multiplied by 1.2, respectively.

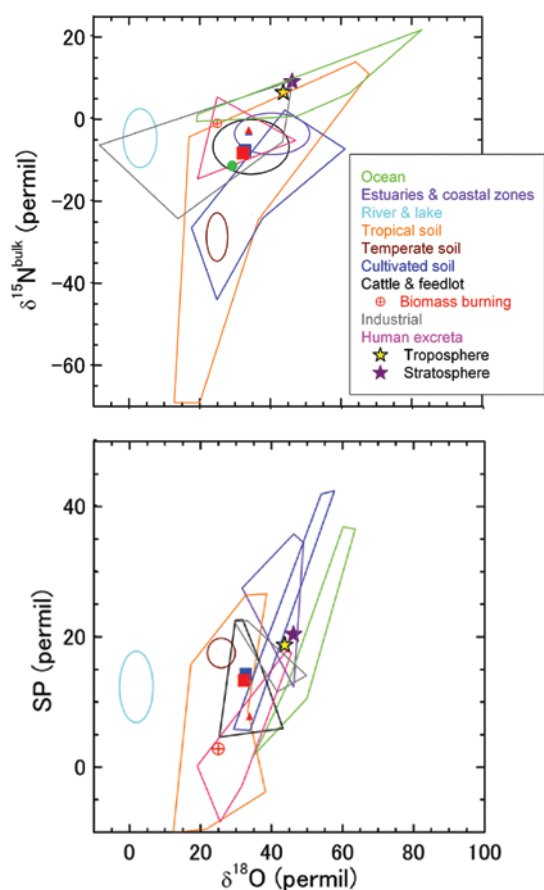


Fig. 2 $\delta^{15}\text{N}^{\text{bulk}}$ and SP for global total N_2O sources estimated in this study (blue and red squares). The measurement value ranges for various sources (Toyoda et al. 2004) are also shown for comparison.

4. Summary

An online chemical transport model that can estimate the global distributions of N_2O isotopomers in the stratosphere and the troposphere has been developed. Fractionation of isotopes during the photolysis and chemical reactions was estimated by using the temperature and radiative flux estimated in the model. To estimate the global distribution of N_2O and its isotopomers, 20-years calculations have been executed. The isotopomers fractions in the emissions were estimated by assuming the isotopic fractions in various sources. The model well reproduced the vertical gradient of N_2O concentration and the isotopomers fractions observed at Sanriku, Kiruna, and Showa-station. The averaged value of the isotopomers fraction in the model was close to that from the emissions from the anthropogenic emissions, especially cultivated soil.

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化学輸送モデルによる大気組成変動と気候影響の研究

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地球温暖化およびそれに伴う気候変動に大きな影響を与えられとされる温室効果気体の大気中濃度変動を解析するため、大気大循環モデル MIROC を基にした温室効果気体に特化した化学輸送モデル ACTM を開発した。温室効果気体のソース・シンク、および輸送過程に関する評価を行うため、一酸化二窒素の同位体を用いた地表放出源分布推定および輸送過程等の検証を行った。地表観測値と ACTM による複数の第一推定値とを組み合わせることで現在気候および産業化以前における地表放出源における全球平均した一酸化二窒素アイソトポマー比の推定を行った結果、モデルが北半球高緯度、中緯度および南極昭和基地でのアイソトポマー比の鉛直分布等を良く再現していること、および $\delta^{15}\text{N}^{\text{bulk}}$ (N^{15} 存在比) や SP (N^{15} が終端に存在する比率)、 $\delta^{18}\text{O}$ などの値から、人為起源放出源、とくに農耕地の N_2O アイソトポマー比に近いことなどが明らかとなった。

キーワード: 大気物質循環, 温室効果気体, 化学輸送モデル, 同位体