

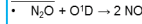
Introduction

With the successful regulation of halogen containing ozone depleting substances (ODS) in the Montreal Protocol and its amendments, today nitrous oxide (N₂O) is the most important ozone depleting species emitted by anthropogenic activity (e.g., Portmann et al., 2012). The future increase of carbon dioxide (CO₂) and methane (CH₄), however, will have a mitigating effect on the ozone depleting potential of N₂O (e.g., Stolarski et al., 2015; see also box on the right). Thus, the future ozone depletion due to N₂O strongly depends on the emission scenarios of CO₂ and CH₄.

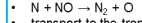
In our study we quantify the contribution from N₂O to ozone loss and thus, the future potential of N₂O to harm the stratospheric ozone layer under the extreme RCP8.5 scenario at the end of the 21st century when the stratospheric halogen loading will have returned to pre-1980 levels. Based on the analysis of multi-year simulations with the chemistry-climate model EMAC we examine the impact of increasing N₂O on ozone and the feedbacks with greenhouse gas (GHG) induced temperature and circulation changes as well as CH₄ induced changes in stratospheric chemistry.

Sources and sinks of NO_y in the stratosphere:

Source:



Sink:



• transport to the troposphere

N + NO reaction rate increases with lower temperature because the concentration of N atoms is increased (N+O → NO+O reaction rate decreased with cooling)

Model & Experiments

- Chemistry-climate model EMAC v2.50.7 (ECHAM/MESy Atmospheric chemistry; Jöckel et al., 2015, GMDD)
- T42L47 resolution (up to 0.01 hPa)
- Time slice simulations for the years 2000 and 2100
- Integration over 40 years (+5 years spin-up)
- Boundary conditions (overview in Table 1):
 - > Future GHG levels: RCP8.5 scenario (Meinshausen et al., 2011)
 - > Future ODS concentrations: A1 scenario (WMO, 2007)
 - > Prescribed fields for sea surface temperature (SST) and sea ice concentration (SIC) from simulations with the MPI-ESM (Schmidt et al., 2013)
 - > Nudged QBO, no solar variability

Simulation	CO ₂	CH ₄	N ₂ O	ODS	Trop. precursor	SST	SIC
REF2000	2000	2000	2000	2000	2000	1995-2004	1995-2004
REF2100	2100	2100	2100	2100	2100	2095-2104	2095-2104
N2O_2100	2000	2000	2100	2100	2000	1995-2004	1995-2004
CH4_2100	2000	2100	2000	2100	2000	1995-2004	1995-2004
GHG_2100	2100	2100	2100	2000	2000	2095-2104	2095-2104
ODS_2100	2000	2000	2000	2100	2000	1995-2004	1995-2004

Table 1 Overview and boundary conditions of the simulations.

- + two transient simulations (1960-2100, EMAC v1.10): RCP6.0 and RCP8.5

Future NO_y changes

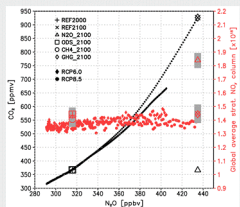


Fig. 1 N₂O and CO₂ boundary conditions for the timeslice and transient simulations (black symbols) and the relation between surface N₂O and stratospheric NO_y column in 10¹⁶ molecules/cm² (red symbols, y-axis on the right, gray shading indicates ±1σ).

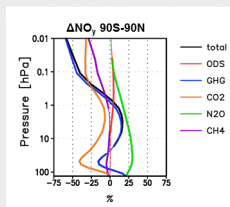


Fig. 3 Global mean annual mean relative differences in NO_y mixing ratio between 2000 and 2100 and the contributions from ODS, GHG, CO₂, CH₄ and N₂O.

- The future increase of N₂O emissions affects the amount of inorganic nitrogen (NO_y = NO+NO₂+reservoir species) in the stratosphere (Fig. 1, 2, 3).
- The NO_y increase caused by a certain N₂O increase depends on the concentration of the GHGs CO₂ and CH₄ (Fig. 2, 3)
- 37% higher N₂O levels lead to an increase of stratospheric NO_y by 28% if the CO₂ and CH₄ are not changed (= year 2000 level; Fig. 1 and 2).
- Accounting for the increase of CO₂ and CH₄ that is projected for the RCP8.5 scenario, the increase of stratospheric NO_y is reduced to less than 1%.
- The largest impact on the NO_y change is found for CO₂, while CH₄ mainly affects NO_y in the mesosphere.

Future ozone changes

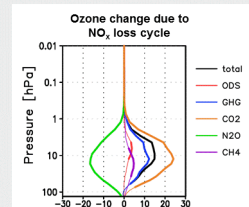


Fig. 4 Global mean annual mean relative differences in ozone mixing ratio due to changes of the loss rate in the catalytic NO_y cycle between 2000 and 2100 and the contributions from ODS, GHG, CO₂, CH₄ and N₂O.

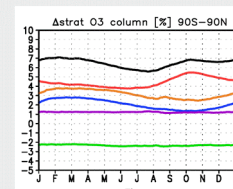


Fig. 5 Annual cycle of global mean relative differences in the stratospheric ozone column between 2000 and 2100 and the contributions from ODS, GHG, CO₂, CH₄ and N₂O.

- The increase of N₂O emissions and the concomitant increase in stratospheric NO_y (under year 2000 conditions for CO₂ and CH₄) leads to a larger loss rate in the catalytic NO_y-cycle and an ozone decrease by up to 18% (Fig. 4).
- The stratospheric ozone column is reduced by 2% throughout the year (Fig. 5).
- No other species emitted by anthropogenic activity (considered in this study) causes an ozone decrease at the end of the 21st century.
- Largest strat. ozone column increase due to ODSs (4-6%) and CO₂ (3-4%).

Sensitivity of NO_y changes to CO₂

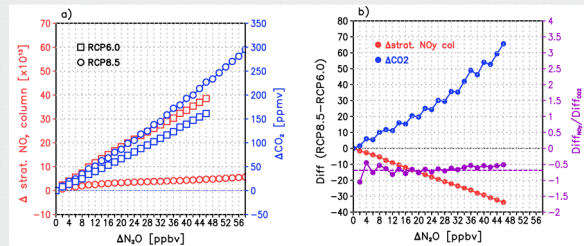


Fig. 6 a) Change of stratospheric NO_y column in 10¹⁶ molecules/cm² (red) and CO₂ mixing ratio in ppmv (blue, right y-axis) for a given N₂O increase (bins of 2 ppbv, starting in the year 2040) in the transient simulations RCP6.0 (square) and RCP8.5 (circle). b) Differences between the RCP6.0 and RCP8.5 runs in the NO_y column (red) and CO₂ (blue) change per N₂O increase. The purple line shows the quotient between the red and the blue line (y-axis on the right). The purple dashed line represents the quotient but derived from the timeslice simulations.

- To analyse the effect of different CO₂ levels on stratospheric NO_y with identical N₂O changes, we use the transient simulations RCP6.0 and RCP8.5.
- By binning the changes in NO_y and CO₂ in both simulations to equal changes in N₂O, the sensitivity of NO_y increase to CO₂ changes is analysed (Fig. 6a).
- The difference between the RCP6.0 and RCP8.5 runs (Fig. 6b) shows that the larger the difference between CO₂, the larger the difference in the stratospheric NO_y column: A N₂O increase of 40 ppbv causes a 7 times larger increase in stratospheric NO_y if the CO₂ increase is 30% smaller.
- The relationship between CO₂ and NO_y differences is not constant (from ~1 to ~0.5) indicating a nonlinear behaviour or the possible influence of other drivers (e.g., differences in the chlorine loading).

Estimation of stratospheric NO_y changes depending on N₂O and CO₂ changes

- > Assuming linear relation between CO₂, temperature and NO_y changes:

$$\Delta NO_y = \Delta N_{2O} [ppbv] \cdot S_{N_{2O}} + \Delta CO_2 [ppmv] \cdot S_{CO_2}$$

with S_{N₂O} and S_{CO₂} representing sensitivity parameters for the changes.

- > S_{N₂O} is derived from the timeslice simulations for N₂O increase only (see Fig. 1):

$$S_{N_{2O}} = \frac{\Delta NO_y}{\Delta N_{2O}} = \frac{0.399 \cdot 10^{16} \text{ molec/cm}^2}{119 \text{ ppbv}} = 3.35 \cdot 10^{13} \frac{\text{molec/cm}^2}{\text{ppbv}}$$

- > S_{CO₂} represents the quotient in Fig. 6 and is found to be

$$S_{CO_2} = \frac{Diff_{NO_y}}{Diff_{CO_2}} = -0.707 \cdot 10^{13} \frac{\text{molec/cm}^2}{\text{ppmv}}$$

in the timeslice simulations.

Conclusions

In this study we have analysed the effect of increasing N₂O emissions on stratospheric NO_y and ozone in simulations with the CCM EMAC. As shown in previous studies the NO_y change resulting from increased N₂O strongly depends on the level of GHGs, in particular CO₂. We find that NO_y is increased by 28%, which leads to an ozone reduction by up to 18%, due to an enhanced catalytic NO_y cycle in the middle stratosphere if N₂O emissions rise following the RCP8.5 scenario for the year 2100 but CO₂ and CH₄ levels are fixed at the year 2000. The corresponding decrease of stratospheric ozone is 2%. Accounting for increasing CO₂ and CH₄ the NO_y increase is considerably reduced due to stratospheric cooling and a more effective NO_y loss. All in all, the N₂O induced stratospheric ozone loss is overcompensated by the effect of the other GHGs.

The sensitivity of NO_y to CO₂ changes is estimated based on the timeslice simulations and tested with the help of transient simulations under the RCP6.0 and RCP8.5 scenarios. We introduce a sensitivity parameter for the effect of CO₂ changes on NO_y, which is constant when derived from the timeslice simulations (by definition) and has a slight slope when derived from the transient simulations. This indicates either nonlinearities or the impact of other processes affecting stratospheric NO_y. However, this provides a rough estimation of the stratospheric NO_y change for potential N₂O and CO₂ changes: For a N₂O increase as projected in the RCP8.5 scenario but a smaller CO₂ increase (as in RCP6.0) the resulting increase of stratospheric NO_y is more than 60 times larger compared to the increase with the RCP8.5 CO₂ increase.

References & Acknowledgments

Portmann, R. W., J. S. Daniel and A. R. Ravishankara, Stratospheric ozone depletion due to nitrous oxide: influence of other gases, *Phil. Trans. Soc. B*, 367, 1256-1264, 2012.

Stolarski, R. S., A. R. Douglass, L. Oman and D. W. Waugh, Impact of future nitrous oxide and carbon dioxide emissions on the stratospheric ozone layer, *Environ. Res. Lett.*, 10, 034011, 2015.

Jöckel, P. et al., Earth System Chemistry Integrated Modelling (ESCI-Mo) with the Modular Earth Submodel System (MESSy, version 2.51), *Geosci. Model Dev. Discuss.*, 2015.

Meinshausen et al., The RCP Greenhouse Gas Concentrations and their Extension from 1765 to 2300, *Climate Change (Special Issue)*, 2011.

World Meteorological Organization (WMO), Scientific Assessment of Ozone Depletion: 2006, *Global Research and Monitoring Project-Report Nr. 50*, 572 pp., Geneva, Switzerland, 2007.

Schmidt et al., Response of the middle atmosphere to anthropogenic and natural forcings in the CMIP5 simulations with the Max Planck Institute Earth system model, *J. Adv. Earth Syst.*, 5, 98-116, 2013.

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