

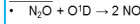
## Introduction

With the successful regulation of halogen containing ozone depleting substances (ODS) in the Montreal Protocol and its amendments, today nitrous oxide (N<sub>2</sub>O) is the most important ozone depleting species emitted by anthropogenic activity (e.g., Portmann et al., 2012). The future increase of carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>), however, will have a mitigating effect on the ozone depleting potential of N<sub>2</sub>O (e.g., Stolarski et al., 2015; see also box on the right). Thus, the future ozone depletion due to N<sub>2</sub>O strongly depends on the emission scenarios of CO<sub>2</sub> and CH<sub>4</sub>.

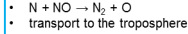
In our study we quantify the contribution from N<sub>2</sub>O to ozone loss and thus, the future potential of N<sub>2</sub>O to harm the stratospheric ozone layer under the extreme RCP8.5 scenario at the end of the 21<sup>st</sup> century when the stratospheric halogen loading will have returned to pre-1980 levels. Based on the analysis of multi-year simulations with the chemistry-climate model EMAC we examine the impact of increasing N<sub>2</sub>O on ozone and the feedbacks with greenhouse gas (GHG) induced temperature and circulation changes as well as CH<sub>4</sub> induced changes in stratospheric chemistry.

Sources and sinks of NO<sub>y</sub> in the stratosphere:

Source:



Sink:



N + NO reaction rate increases with lower temperature because the concentration of N atoms is increased (N+O<sub>2</sub> → 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 <sub>2</sub>	CH <sub>4</sub>	N <sub>2</sub> 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<sub>y</sub> changes

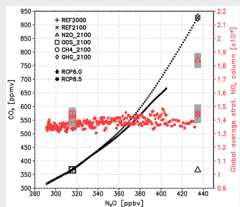


Fig. 1 N<sub>2</sub>O and CO<sub>2</sub> boundary conditions for the timeslice and transient simulations (black symbols) and the relation between surface N<sub>2</sub>O and stratospheric NO<sub>y</sub> column in 10<sup>15</sup> molecules/cm<sup>2</sup> (red symbols, y-axis on the right, gray shading indicates ±1σ).

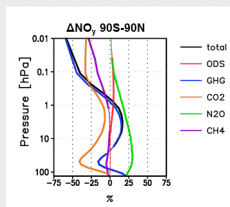


Fig. 3 Global mean annual mean relative differences in NO<sub>y</sub> mixing ratio between 2000 and 2100 and the contributions from ODS, GHG, CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O.

- The future increase of N<sub>2</sub>O emissions affects the amount of inorganic nitrogen (NO<sub>y</sub> = NO+NO<sub>2</sub>+reservoir species) in the stratosphere (Fig. 1, 2, 3).
- The NO<sub>y</sub> increase caused by a certain N<sub>2</sub>O increase depends on the concentration of the GHGs CO<sub>2</sub> and CH<sub>4</sub> (Fig. 2, 3)
- 37% higher N<sub>2</sub>O levels lead to an increase of stratospheric NO<sub>y</sub> by 28% if the CO<sub>2</sub> and CH<sub>4</sub> are not changed (= year 2000 level; Fig. 1 and 2).
- Accounting for the increase of CO<sub>2</sub> and CH<sub>4</sub> that is projected for the RCP8.5 scenario, the increase of stratospheric NO<sub>y</sub> is reduced to less than 1%.
- The largest impact on the NO<sub>y</sub> change is found for CO<sub>2</sub>, while CH<sub>4</sub> mainly affects NO<sub>y</sub> in the mesosphere.

## Future ozone changes

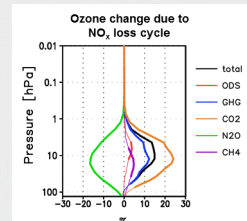


Fig. 4 Global mean annual mean relative differences in ozone mixing ratio due to changes of the loss rate in the catalytic NO<sub>y</sub>-cycle between 2000 and 2100 and the contributions from ODS, GHG, CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O.

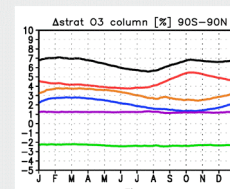


Fig. 5 Annual cycle of global mean relative differences in the stratospheric ozone column between 2000 and 2100 and the contributions from ODS, GHG, CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O.

- The increase of N<sub>2</sub>O emissions and the concomitant increase in stratospheric NO<sub>y</sub> (under year 2000 conditions for CO<sub>2</sub> and CH<sub>4</sub>) leads to a larger loss rate in the catalytic NO<sub>y</sub>-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 21<sup>st</sup> century.
- Largest strat. ozone column increase due to ODSs (4-6%) and CO<sub>2</sub> (3-4%).

## Sensitivity of NO<sub>y</sub> changes to CO<sub>2</sub>

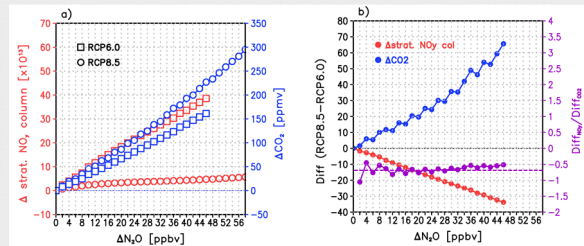


Fig. 6 a) Change of stratospheric NO<sub>y</sub> column in 10<sup>15</sup> molecules/cm<sup>2</sup> (red) and CO<sub>2</sub> mixing ratio in ppmv (blue, right y-axis) for a given N<sub>2</sub>O increase (bins of 2 ppbv, starting in the year 2040) in the transient simulations RCP6.0 (square) and RCP8.5 (circle). b) Differences between the RCP6.0 and RCP8.5 runs in the NO<sub>y</sub> column (red) and CO<sub>2</sub> (blue) change per N<sub>2</sub>O 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 CO<sub>2</sub> levels on stratospheric NO<sub>y</sub> with identical N<sub>2</sub>O changes, we use the transient simulations RCP6.0 and RCP8.5.
- By binning the changes in NO<sub>y</sub> and CO<sub>2</sub> in both simulations to equal changes in N<sub>2</sub>O, the sensitivity of NO<sub>y</sub> increase to CO<sub>2</sub> changes is analysed (Fig. 6a).
- The difference between the RCP6.0 and RCP8.5 runs (Fig. 6b) shows that the larger the difference between CO<sub>2</sub>, the larger the difference in the stratospheric NO<sub>y</sub> column: A N<sub>2</sub>O increase of 40 ppbv causes a 7 times larger increase in stratospheric NO<sub>y</sub> if the CO<sub>2</sub> increase is 30% smaller.
- The relationship between CO<sub>2</sub> and NO<sub>y</sub> 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<sub>y</sub> changes depending on N<sub>2</sub>O and CO<sub>2</sub> changes

- > Assuming linear relation between CO<sub>2</sub>, temperature and NO<sub>y</sub> changes:

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

with S<sub>N<sub>2</sub>O</sub> and S<sub>CO<sub>2</sub></sub> representing sensitivity parameters for the changes.

- > S<sub>N<sub>2</sub>O</sub> is derived from the timeslice simulations for N<sub>2</sub>O 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<sub>CO<sub>2</sub></sub> 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 N<sub>2</sub>O emissions on stratospheric NO<sub>y</sub> and ozone in simulations with the CCM EMAC. As shown in previous studies the NO<sub>y</sub> change resulting from increased N<sub>2</sub>O strongly depends on the level of GHGs, in particular CO<sub>2</sub>. We find that NO<sub>y</sub> is increased by 28%, which leads to an ozone reduction by up to 18%, due to an enhanced catalytic NO<sub>y</sub> cycle in the middle stratosphere if N<sub>2</sub>O emissions rise following the RCP8.5 scenario for the year 2100 but CO<sub>2</sub> and CH<sub>4</sub> levels are fixed at the year 2000. The corresponding decrease of stratospheric ozone is 2%. Accounting for increasing CO<sub>2</sub> and CH<sub>4</sub> the NO<sub>y</sub> increase is considerably reduced due to stratospheric cooling and a more effective NO<sub>y</sub> loss. All in all, the N<sub>2</sub>O induced stratospheric ozone loss is overcompensated by the effect of the other GHGs.

The sensitivity of NO<sub>y</sub> to CO<sub>2</sub> 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 CO<sub>2</sub> changes on NO<sub>y</sub>, 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<sub>y</sub>. However, this provides a rough estimation of the stratospheric NO<sub>y</sub> change for potential N<sub>2</sub>O and CO<sub>2</sub> changes: For a N<sub>2</sub>O increase as projected in the RCP8.5 scenario but a smaller CO<sub>2</sub> increase (as in RCP6.0) the resulting increase of stratospheric NO<sub>y</sub> is more than 60 times larger compared to the increase with the RCP8.5 CO<sub>2</sub> increase.

## References & Acknowledgments

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