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A MODEL STUDY OF THE IMPACT OF WELL-MIXED GREENHOUSE GASES (GHGS) ON TROPOSPHERIC CHEMISTRY FROM THE PREINDUSTRIAL ERA TO THE PRESENT DAY

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1. INTRODUCTION

Changes in the atmospheric abundance of greenhouse gases (GHGs; carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O)) and ozone depleting substances (ODSs) have been shown to significantly affect the chemical and dynamical structure of both the stratosphere (WMO 2011) and the troposphere (IPCC 2013). It has been shown that GHGs concentrations have been increasing since preindustrial times Meinshausen, Smith et al. (2011). Increasing atmospheric concentrations of GHGs and ODSs since preindustrial time (Figure 1 based on data from Meinshausen et al. 2011) have the potential to alter the distribution and evolution of tropospheric ozone (O_3). Previous modeling studies have examined how long-term increases in GHGs and ODSs impact stratospheric O_3 and dynamics. For example, Fleming et al. (2011) investigated the relative contributions of individual and combined increases in driving changes in stratospheric O_3. They applied numerous sensitivity simulations using two-dimensional models focusing on stratospheric changes and did not consider changes in the troposphere. Lang et al. (2012), on the other hand, examine GHGs and ODSs applied a three-dimensional model chemistry-climate model with combined tropospheric and stratospheric chemistry to investigate the impact of past changes in GHGs and ODSs on tropospheric O_3. However, their study was based on a series on time-slice integrations combining the impact of CO_2 N_2O and SSTs into one simulation and isolating the role of ODSs and CH_4 into two different simulations.

In this study we apply transient simulations of the GFDL coupled chemistry climate model (CM3) to analyze how combined and individual increases in CO_2, CH_4, N_2O and ODSs over the historical period (1860-2005) have impacted tropospheric chemistry, particularly tropospheric O_3.

In section 2 we describe the model used and the simulations performed for this study. In section 3, we present the results from our sensitivity simulations focusing on total ozone column (TOC), surface O_3, specific humidity, and hydrochloric acid (HCl). Illustrations and tables are presented in section 4 and a summary and future work is in section 5.

2. METHODOLOGY

2.1. *Model Description and Simulations*

We apply the GFDL coupled chemistry-climate model (CM3),a fully coupled global atmosphere-ocean-land-sea ice model (Donner et al., 2011) that simulates tropospheric and stratospheric chemistry (Austin et al., 2013; Naik et al., 2013) seamlessly (Figure 2).

We performed four coupled transient (1860-2005) CM3 simulations on cubed-sphere grid (~2º x 2º) with 48 vertical layers extending up to 86 km (0.01 hPa). To isolate the impact of each of the greenhouse gases and ODSs on tropospheric chemistry, we varied the concentrations of CO_2, CH_4, N_2O and ODSs individually while keeping constant all other long-lived forcings at their 1860 levels (Table 1). In all the simulations:

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- short-lived species emissions (i.e., aerosols and O_3 precursors other than CH_4) were set at 1860 levels,
- volcanic emissions (i.e., SO_2) were turned off,
- dynamic vegetation was applied,
- solar radiation was set to 1860 levels,
- biogenic emissions of VOCs were fixed, and
- natural emissions of a few species varies with meteorology (i.e., lightning, NO_x, DMS, oceanic organic carbon, dust, and sea-salt).

In the next section, we describe the impact of increases in CO_2, CH_4, N_2O, and ODSs individually on tropospheric chemistry with a focus on tropospheric O_3 driven by changes in chemistry and dynamics.

3. RESULTS

3.1. *Historical Evolution of Global Total Ozone Column (TOC)*

Figure 3 shows the transient change in absolute and percent change in TOC relative to mean over the 1860-1869 period. TOC increases in response to CH_4 increases driven by CH_4-induced net photochemical tropospheric O_3 production with sharp increase beginning in the 1920s. Doubling of CH_4 in the present day relative preindustrial results in about 4% increase in TOC by 2005. TOC decreases in response to ODS-induced stratospheric O_3 loss beginning in the 1970s. TOC decreases from N2O-induced stratospheric O3 loss beginning earlier (in the 1940s) than those induced by ODSs. However, the reductions in the last 30 years are smaller than those induced by ODSs indicating that N_2O is less efficient in destroying stratospheric O_3. Increases in CO_2 produce negligible changes in TOC. CO_2 changes alter the atmospheric transport and climate producing small changes in O_3 throughout the atmosphere.

3.2. *Preindustrial (PI) to present-day (PD) Surface Ozone Change*

Figure 4 shows the changes in surface O_3 concentrations from preindustrial to present day in response to the individual increases in CO_2, CH_4, N_2O and ODSs. Consistent with previous studies, CH_4 increases lead to uniform increases in surface O_3 with a global average increase of about 4 ppbv. Increases in ODSs result in global average surface O_3 decrease of about 1.3 ppbv with strong reductions simulated over the southern ocean driven by stratospheric O_3 loss. PI to PD CO_2 increase produces surface O_3 reductions in the tropics of up to 1.5 ppbv and increases in the extratropics with a small change in the global average. N_2O increases also produce negligible changes in surface O_3 concentration.

3.3. *PI to PD Temperature and Specific Humidity Change*

We next analyze the PI to PD changes in atmospheric temperature to explain changes in O_3. Figure 5 shows the change in annual mean zonal average temperature for the surface to about 1 hPa for PI to PD. The stratosphere cools while troposphere warms in response to individual increases in greenhouse gases and ODSs. The changes in temperature over the historical period are strongest for CO_2 increases and weakest for N_2O increases. Figure 6 shows the PI to PD change in specific humidity for each of the four simulations. Tropospheric water vapor increases in response to GHG and ODS-induced warming, with the largest percent increases occurring in the upper troposphere in response to CO_2 and CH_4 increases. Since CH_4 is a precursor to stratospheric water vapor, increases in CH_4 lead to significant specific humidity enhancement in the stratosphere.

3.5. *PI to PD Ozone Change*

In this section we analyze the change in both stratospheric and tropospheric O_3 concentrations to help further diagnose the tropospheric O_3 response. Figure 7 shows the PI to PD changes in annual mean zonal average O_3 for each of the four simulations. CO_2 increases enhance upper stratospheric O_3 (induced by cooling) but decrease tropospheric O_3, particularly in the lower tropical troposphere, driven by chemistry and circulation changes (will be discussed in more detail in a journal article). CH_4 increases O_3 uniformly (by about 10 ppbv) in low to mid-troposphere, despite constant NO_x emissions. Lower to mid-stratospheric O_3 increases from the combined effect of CH_4 – induced cooling and converting active chlorine (Cl) to longer-lived reservoir species HCl. Stratospheric O_3 losses induced by ODS and N_2O increases cause tropospheric reductions. As mentioned previously stratospheric O_3 losses are stronger for ODS increase compared to N_2O increase.

3.6. *PI to PD HCl Change (Figure 8)*

Figure 8 shows the PI to PD changes in hydrochloric acid (HCl). Increases in CH_4 and ODSs produce dominant changes in HCl. CH_4 combines with reactive Cl to produce longer-lived HCl (a chlorine reservoir species), resulting in a buildup of stratospheric O_3. (Fleming et al., 2011). For the ODS simulation, significant amounts of ClO and other short-lived reservoir species (e.g. $ClONO_2$ and HOCl) are also produced in addition to HCl which lead to stratospheric O_3 loss.

4. ILLUSTRATIONS AND TABLES

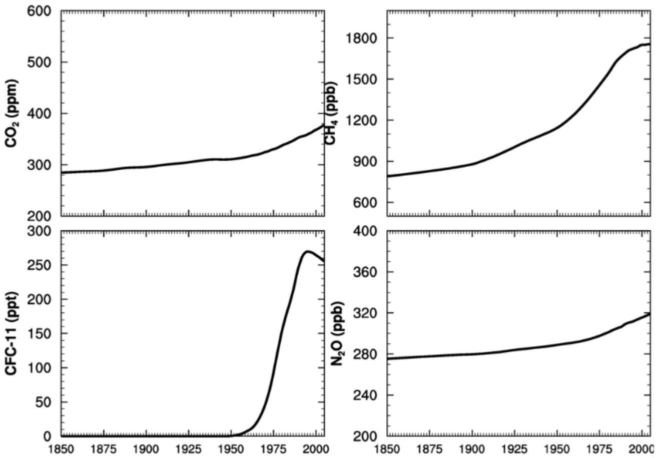

Figure 1. Evolution of GHGs (i.e., CO2, CH4, N2O) and ODSs (i.e., CFC-11) over the historical period. Data from Meinshausen et al. (2011).

Simulation	Long-lived Forcings (1860-2005)	Chemistry
CO_2	Time varying CO_2, constant CH_4, N_2O, and ODSs at 1860 levels	CH_4 and N_2O set to 1860 levels
CH_4	Time varying CH_4, constant CO_2, N_2O, and ODSs at 1860 levels	N_2O and ODSs set to 1860 levels
N_2O	Time varying N_2O, constant CO_2, CH_4, and ODSs at 1860 levels	CH_4 and ODSs set to 1860 levels
ODS	Time varying ODSs, constant CO_2 and CH_4 at 1860 levels	CH_4 and N_2O set to 1860 levels

Table 1. Simulations performed using GFDL-CM3

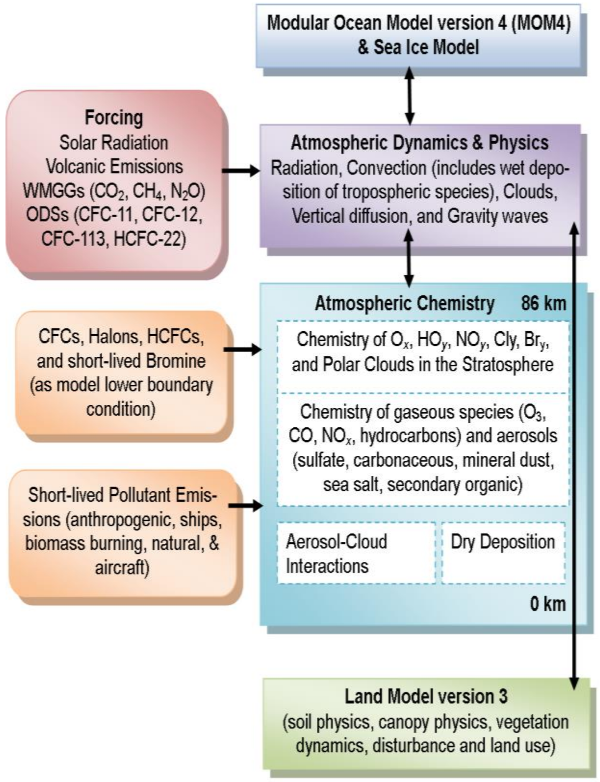

Figure 2. Schematic diagram of how the GFDL CM3 model operates (Donner et al., 2011; Austin et al., 2013; Naik et al., 2013)

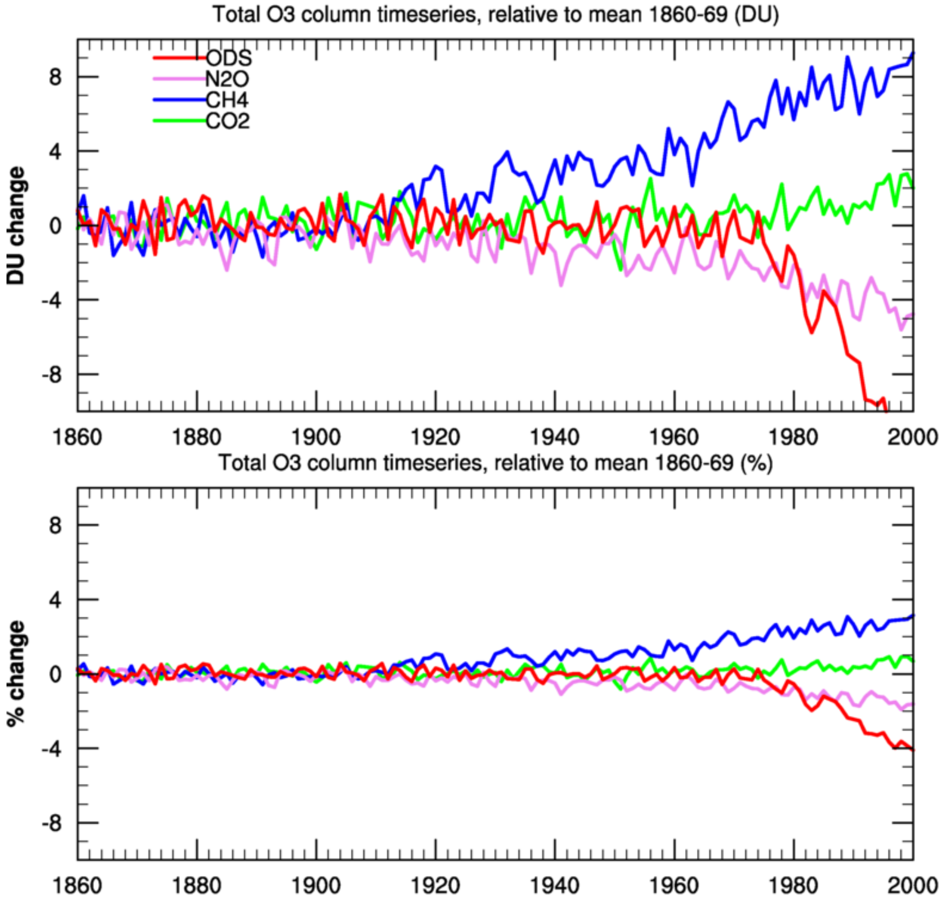

Figure 3. Total ozone column (TOC) changes (up) and percent changes (down), relative to mean 1860-69

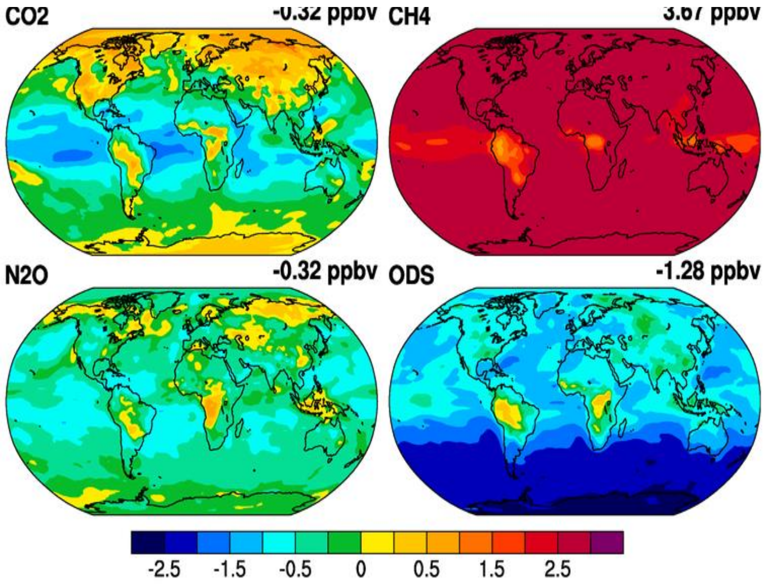

Figure 4. Preindustrial (PI) to present-day (PD) changes in surface O_3

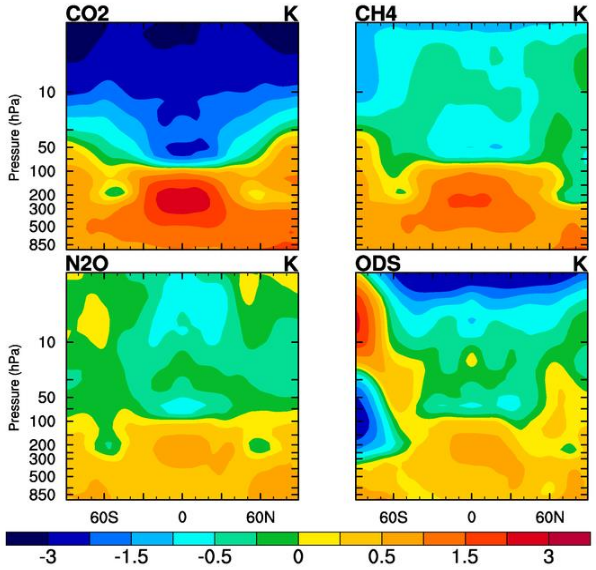

Figure 5. PI to PD changes in annual mean temperature

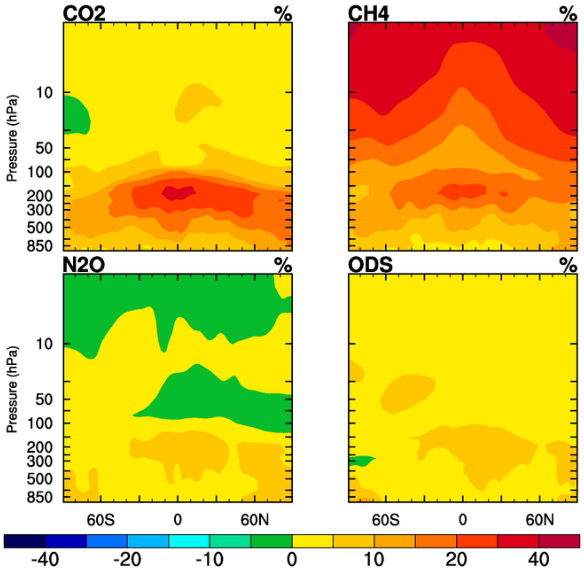

Figure 6. PI to PD changes in annual mean specific humidity

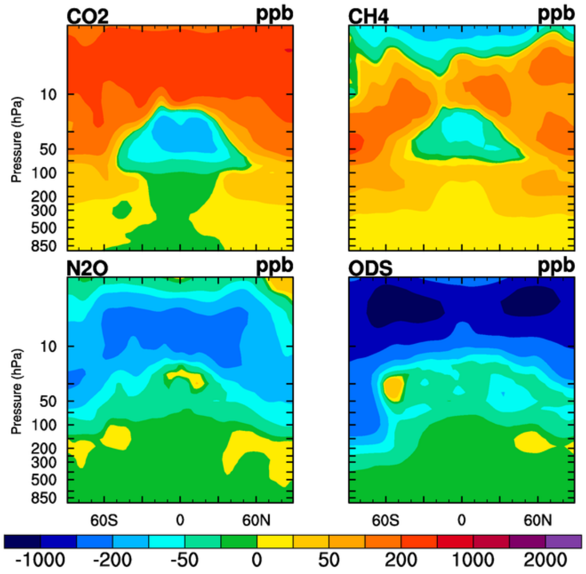

Figure 7. PI to PD changes in annual mean ozone

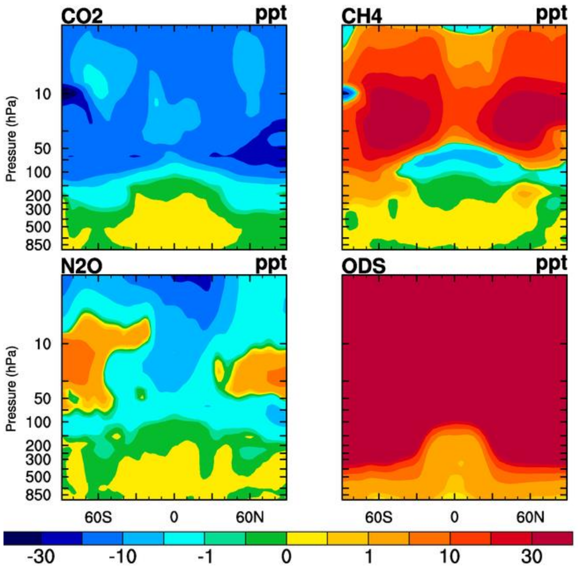

Figure 8. PI to PD changes in annual mean hydrochloric acid (HCl)

5. SUMMARY AND FUTURE WORK

We applied sensitivity simulations of the GFDL-CM3 model to explore the impacts of changes in individual GHGs (i. e., CO_2, CH_4, N_2O, and ODSs) on tropospheric composition with a focus on ozone (O_3). Increases in CH4 result in increases in TOC while N_2O and ODS increases cause TOC to decrease in the present day relative to preindustrial. CO_2 produces negligible changes in TOC. Increases in CH_4 cause globally uniform O_3 increases in the troposphere with global average surface O_3 concentrations increasing by ~4 ppbv from preindustrial to present day. CO_2 increases produce noticeable tropospheric ozone decreases in the tropics, particularly in the upper troposphere, and minimal increases in the extratropics from CO_2-induced changes in atmospheric circulation.

For future work we will like to explore changes in tropospheric O_3 in response to combined increases in CH_4 and ODSs that have offsetting impacts. Also, we are interested in exploring seasonal changes, tropospheric O_3 budget changes, and changes in brewer-dobson circulation.

6. REFERENCES

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