IMPACT OF HISTORICAL CHANGES IN WELL-MIXED GREENHOUSE GASES ON TROPOSPHERIC COMPOSITION

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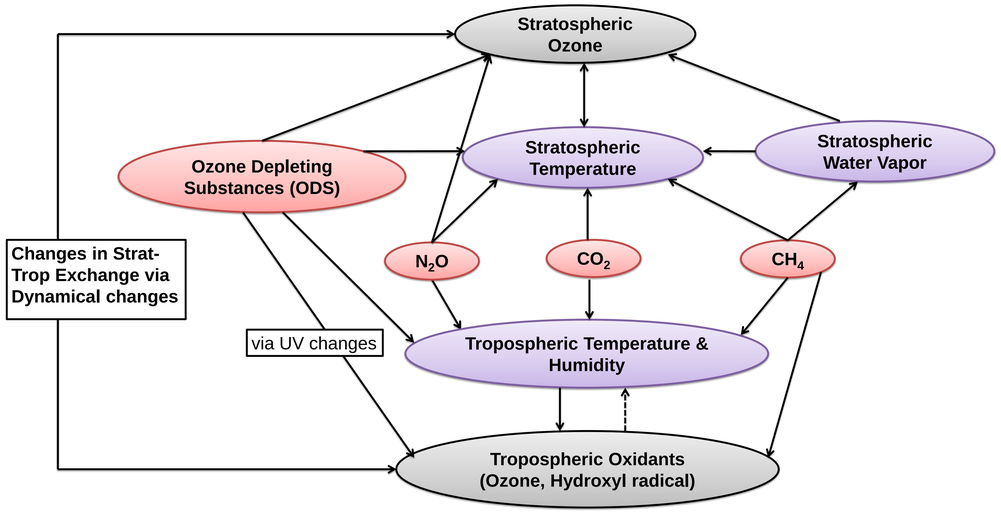
1. Introduction

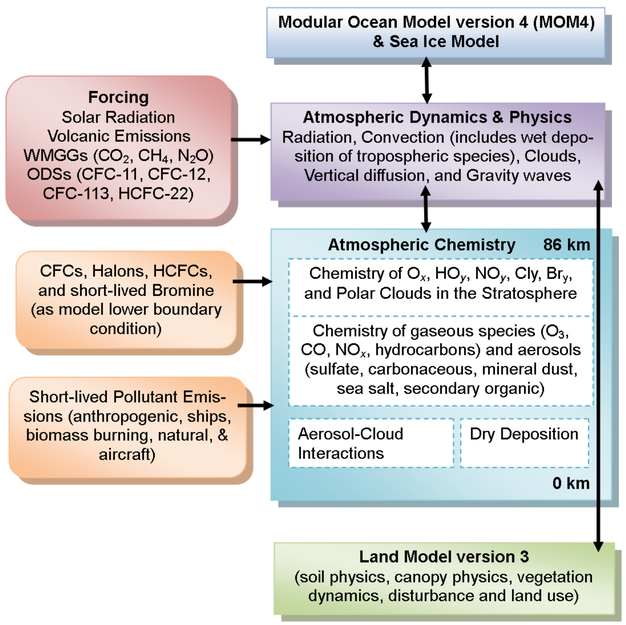
- Increasing atmospheric abundances of well-mixed greenhouse gases (WMGGs – CO_2, N_2O and CH_4), including halogenated ozone depleting substances (ODSs), since the preindustrial times have been shown to significantly impact both the stratospheric (WMO 2011) and the tropospheric (IPCC, 2007) climate and composition.
- Since the stratosphere-troposphere is a coupled system, any radiative, chemical, and dynamical changes in the stratosphere affect the troposphere and vice-versa. For example, CO_2-induced cooling of the stratosphere leads to an increase in the upper stratospheric ozone (O_3) resulting in enhanced stratospheric O_3 transport into the troposphere particularly at mid latitudes (e.g. Fleming et al., 2011). Changes in stratospheric O_3 alter tropospheric photochemistry by affecting the incoming UV radiation (e.g. Voulgarakis et al., 2013).
- The following schematic depicts the impact of changes in WMGGs and ODSs on tropospheric composition with feedbacks from changes in stratospheric climate and composition:



Objective: Investigate the changes in tropospheric composition (with focus on ozone and hydroxyl radical) from combined and individual increases in CO_2, CH_4, N_2O and ODSs over the historical period (1860-2005) using a coupled chemistry-climate model.

2. GFDL Coupled Chemistry-Climate Model (CM3) & Simulations

- GFDL CM3 (Donner et al., 2011; Golaz et al., 2011) is a fully coupled global atmosphere-ocean-land-sea ice model with seamless representation of tropospheric and stratospheric chemistry (Naik et al., 2013; Austin et al., 2013) and explicit physical representation of aerosol-cloud indirect effects (Ming and Ramaswamy, 2008; Levy et al., 2013).
- Coupled transient (1860-2005) CM3 simulations are performed on a cubed-sphere grid (~2° x 2°) with 48 vertical layers extending up to 86 km (0.01 hPa).



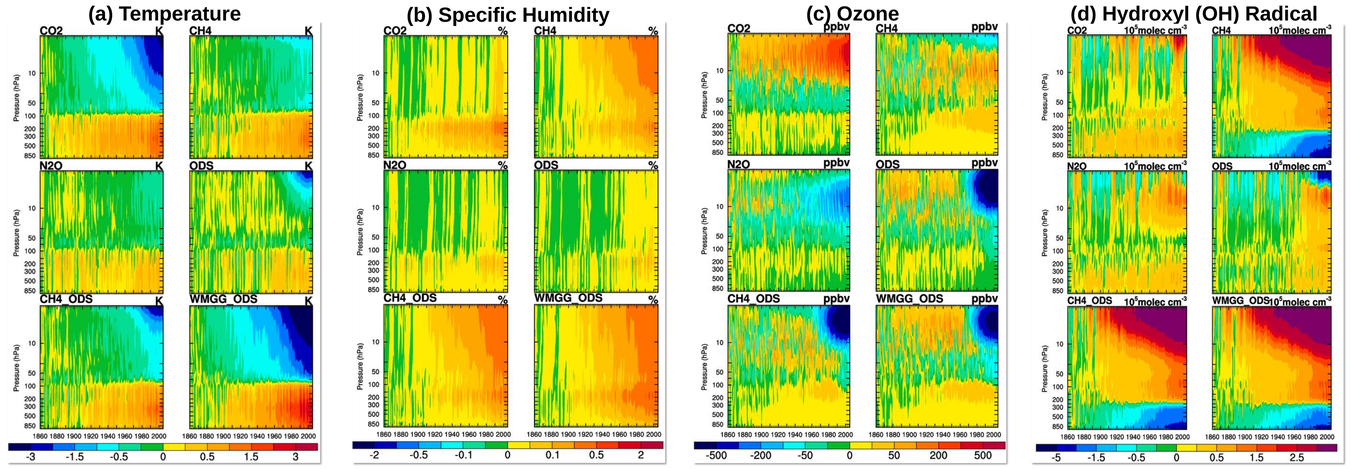
Simulations	Forcings (1860-2005)	Chemistry
CO2	Time varying CO_2	1860 CH_4, N_2O, ODS
CH4	Time varying CH_4	1860 N_2O, ODS
N2O	Time varying N_2O	1860 CH_4, ODS
ODS	Time varying ODSs	1860 CH_4, N_2O
CH4_ODS	Time varying CH_4 and ODS	1860 N_2O
WMGG_ODS	Time varying CH_4, CO_2, N_2O, and ODS	Time varying CH_4, CO_2, N_2O, and ODS

For all simulations:

- Short-lived pollutant emissions are set to 1860 levels,
- Volcanic emissions are turned off and solar radiation is set to 1860 level,
- Dynamic vegetation is used,
- Biogenic volatile organic compound (BVOC) emissions are fixed, and lightning NO_x, dimethyl sulfide (DMS), oceanic organic carbon, dust and sea-salt emissions vary with meteorology.
- Historical global abundances of WMGGs and ODSs are from Meinshausen et al. (2011). WMGGs increase steadily from 1850 while ODSs begin increasing after 1950.

3. Results and Analysis

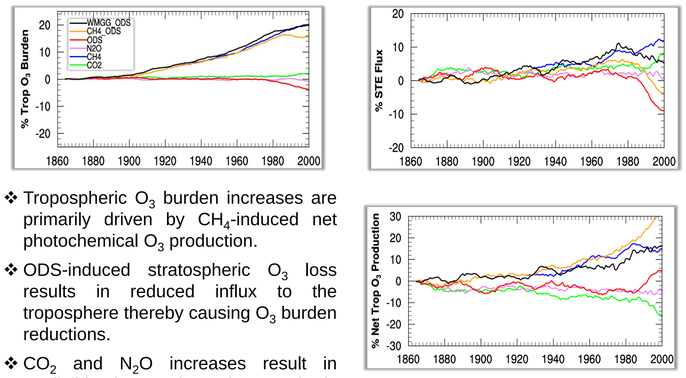
Figure 1. Evolution of annual mean (a) temperature, (b) specific humidity, (c) ozone, and (d) OH expressed as difference from mean 1860-69.



Key Points:

- The stratosphere cools while troposphere warms in response to individual or combined WMGG and ODS increases.
- The changes in temperature over the historical period are strongest for CO_2 increases and weakest for N_2O increases.
- Tropospheric water vapor increases in response to WMGG and ODS-induced warming, with largest percent increases occurring in the upper troposphere.
- CH_4 increase leads to enhanced water vapor in the stratosphere with maximum increases from 1980 onwards.
- CO_2 increases enhance upper stratospheric O_3 (induced by cooling) but decreases tropospheric O_3, particularly in the low troposphere, driven by chemistry and circulation changes (see below).
- Since it is a major precursor of tropospheric O_3, increasing CH_4 increases O_3 uniformly (10 ppb) in low to mid-troposphere in the 20th century, despite constant NO_x emissions.
- Stratospheric O_3 losses induced by ODS and N_2O increases cause tropospheric O_3 reductions. ODS-induced changes are significant only after 1960.
- Tropospheric OH increases in response to CO_2-induced water vapor increases over the historical period.
- Water vapor increases combined with enhanced UV penetration into the troposphere resulting from N_2O and ODS-induced stratospheric loss leads to tropospheric OH increases.
- CH_4 increase causes significant tropospheric OH decreases, particularly in the last two decades, as it is a major sink of OH. CH_4 increase also leads to significant increases in upper stratospheric OH induced by enhanced water vapor abundance.

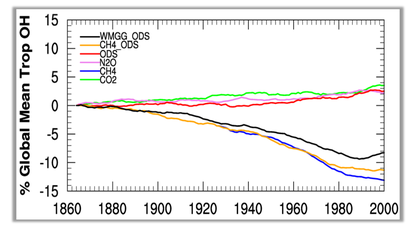
Figure 2. Tropospheric O_3 Burden, Strat-trop exchange (STE), and Net chemical production relative to mean 1860-69 plotted as 10-year running mean



- Tropospheric O_3 burden increases are primarily driven by CH_4-induced net photochemical O_3 production.
- ODS-induced stratospheric O_3 loss results in reduced influx to the troposphere thereby causing O_3 burden reductions.
- CO_2 and N_2O increases result in negligible changes in total tropospheric O_3 burden over the historical period.

Figure 3. Global Annual Mean Tropospheric OH relative to mean 1860-69 plotted as 10-year running mean

- CH_4 causes largest decreases in global mean tropospheric OH over the historical period.
- Increasing CO_2, N_2O and ODS offset the CH_4-induced tropospheric OH reductions only by a small amount.



4. Preliminary Conclusions and Future Work

- Increases in CO_2, CH_4, N_2O, and halogenated ozone depleting substances (ODSs) affect tropospheric composition either directly through chemistry changes or indirectly through radiative and dynamical influences on the stratosphere-troposphere coupled system.
- Sensitivity simulations indicate that of the WMGGs, increasing CH_4 is the dominant driver of global tropospheric O_3 increases over the last century. ODS-induced stratospheric O_3 loss after ~1970 leads to tropospheric O_3 decreases, however, the influence of CH_4 dominates when both CH_4 and ODSs increase simultaneously.
- CH_4 is the dominant driver of OH changes both in the stratosphere and in the troposphere over the historical period.
- CO_2 and N_2O increases have only a small impact on tropospheric O_3 and OH, although changes in the biosphere (e.g., BVOCs, soil NO_x) induced by these gases, not included in the current modeling setup, could potentially cause larger changes in O_3 and OH (Isaksen et al., 2009).
- Examine circulation changes to gain a better understanding of changes in tropospheric composition that are induced by dynamical responses to increasing WMGGs and ODSs.
- Analyze seasonal and spatial distribution of changes in tropospheric composition induced by increasing WMGGs and ODSs with a particular focus on surface pollution.
- Apply similar sensitivity simulations to project future changes in atmospheric composition from individual and combined changes in WMGGs and ODSs.

5. References

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