A Model Study of the Impact of Source Gas Changes on the Stratosphere, 1860-2100

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1. OVERVIEW/OBJECTIVES

- The greenhouse gases (GHGs) CO_2, CH_4, and N_2O, and the chlorine- and bromine-containing ozone depleting substances (ODSs) as represented by the Equivalent Effective Stratospheric Chlorine (EESC), undergo large changes over the 1860-2100 time period (Figure 1). These perturbations have significant impacts on the time evolution of the stratosphere.
- In this study, simulations from the GSFC 2D interactive chemistry-dynamics-radiation model are used to separate the individual impacts of CO_2, CH_4, N_2O, and the ODSs. To do this, we ran a series of experiments in which the surface concentrations of only the ODSs or the individual GHGs are varied time dependently for 1860-2100 (Fig. 1), while all other source gases are fixed at 1860 levels. We compare these with a simulation in which all source gases are varied simultaneously (full simulation).
- We also compare the 2D model with observations and with full simulations of the Goddard 3D chemistry-climate model (GEOSCCM) to illustrate that the 2D model captures the basic processes responsible for long term stratospheric changes.
- We focus on globally averaged ozone, temperature, and age of air. The age of air is the time required for air parcels to travel from the tropical tropopause to a given location in the stratosphere.
- We also examine the photochemical lifetimes of N_2O and several chloroflurocarbons.

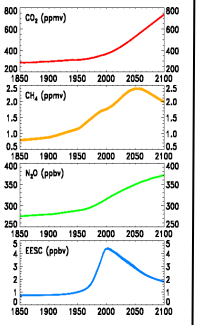

Figure 1. Changes in source gases for 1850-2100. Greenhouse gases are from the IPCC A1b scenario for 1950-2100, and Hanson and Sato [2004] for 1850-1950. The EESC curve (Equivalent Effective Stratospheric Chlorine) represents the accumulation of stratospheric chlorine and bromine due surface emissions of ozone depleting substances (ODSs) based on WMO [2003, 2007] and Butler et al. [1999].

4. Temperature (global average)

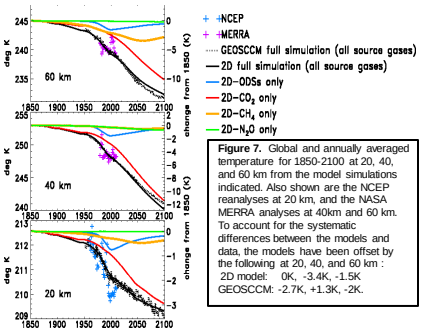

Figure 7. Global and annually averaged temperature for 1850-2100 at 20, 40, and 60 km from the model simulations indicated. Also shown are the NCEP reanalyses at 20 km, and the NASA MERRA analyses at 40km and 60 km. To account for the systematic differences between the models and data, the models have been offset by the following at 20, 40, and 60 km : 2D model: 0K, -3.4K, -1.5K GEOSCCM: -2.7K, +1.3K, -2K.

Major points:

- The 2D and GEOSCCM full simulations have similar rates of stratospheric cooling, and generally agree with the rate of cooling seen in the data.
- The CO_2 cooling has the largest impact, especially by 2100.
- The ODS-induced ozone depletion results in less stratospheric heating, with a maximum impact in 2000.
- The CH_4-induced H_2O cooling has a significant effect at 60 km.
- There are relatively small temperature changes prior to ~1960.

5. Age of Air:

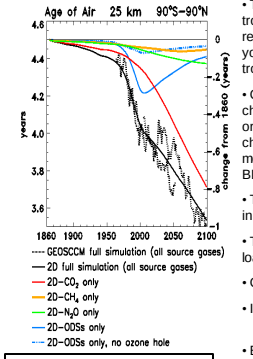

Figure 8. Global and annually averaged age of air at 25 km for 1860-2100, from the model simulations indicated. The blue dashed-dotted curve is a 2D ODS-only simulation with all heterogeneous chemical reactions turned off so that no ozone hole is simulated. The right hand axis shows the change relative to 1860.

Major points:

- The age of air is the time required for air parcels to travel from the tropical tropopause to a given location in the stratosphere. Changes in the age of air reflect changes in the strength of the Brewer-Dobson circulation (BDC), e.g., a younger age of air implies a stronger BDC and faster transport from the tropical tropopause.
- Changes in the atmospheric loading of CO_2, CH_4, N_2O, and the ODSs change the temperature distribution through radiative changes, either directly or indirectly via the changes in ozone and water vapor. These temperature changes modify the zonal wind (via thermal wind balance), which in turn modifies the planetary wave driving. This ultimately induces changes in the BDC and stratospheric age of air.
- The 2D model and GESOCCM full simulations show similar rates of decrease in age of air during the 1950-2100 time period.
- The impacts prior to 1960 are relatively small, and are mainly due to CO_2 loading.
- CO_2 and ODS loading have the largest impacts during ~1970-2060.
- In the 2D full simulation (all source gases), the total age of air decrease is ~1 year from 1860-2100.
- By 2100 CO_2 forcing is dominant; N_2O and CH_4 have secondary impacts, contributing 13% and 6%, respectively, to the total change from 1860-2100.
- Comparing the simulations with the ozone hole (blue solid) and without the ozone hole (blue dashed-dotted) shows that the age of air changes induced by ODS loading are mainly due to the ozone hole, via the associated changes in temperature, zonal wind, and planetary wave driving at high latitudes of the southern hemisphere.

2. 1860-2100 Ozone:

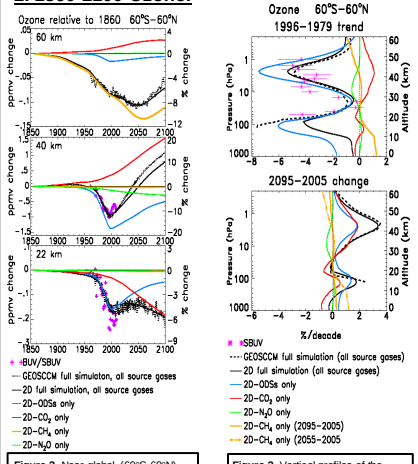

Figure 2. Near-global (60°S-60°N) annually averaged ozone time series for 1860-2100, relative to 1860, for 22, 40, and 60 km from the model simulations as indicated, and the BUV/SBUV satellite data. Values are in ppmv change (left axes) and percent change (right axes).

Figure 3. Vertical profiles of the near-global average (60°S-60°N) ozone trend for 1996-1979 (top) and 2095-2005 (bottom). The red asterisks are the trends derived from SBUV data, and the colored lines depict the trends from the model simulations as indicated. The CH_4-only simulation is shown for two time periods.

Major points:

- During 1960-2000, ozone changes in the lower and upper stratosphere and in the total column are dominated by ODS loading.
- CO_2 cooling increases ozone in the upper stratosphere due to the slower temperature dependent ozone loss rates. This has the largest impact at 40 km and in the total column, causing increases of 20% and 4% (12.5 DU), respectively, from 1860-2100.
- CO_2 loading also accelerates the BDC (see Figure 8). This *increases* ozone at mid-high latitudes and *decreases* ozone in the tropics by advecting ozone-poor air upward from the troposphere (see Figure 5, red curve). This mechanism also results in a 5% ozone decrease from 1860-2100 in the near-global average at 22 km (Figure 2, bottom panel).
- N_2O loading and the subsequent increase in NOx-ozone loss maximizes near 35 km (-0.5%/decade), and causes a 2.7% (8 DU) global total ozone decrease from 1860-2100.

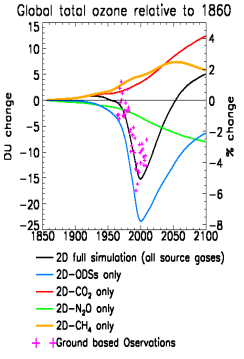

Figure 4. Global and annually averaged total ozone time series for 1860-2100, relative to 1860, from the model simulations indicated. Also shown are ground-based data updated from Fioletov et a. [2002]. Values are in Dobson unit (DU) change (left axes) and percent change (right axes).

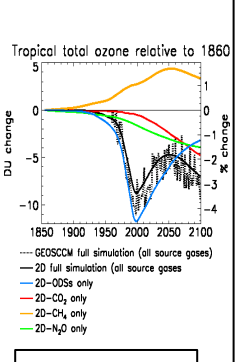

Figure 5. Annually averaged tropical (10°S-10°N) total ozone time series for 1860-2100, relative to 1860, from the model simulations indicated. Values are in Dobson unit (DU) change (left axes) and percent change (right axes).

Major points (continued):

- CH_4 loading impacts ozone by three mechanisms:
  1) tropospheric ozone *increases* due to NO_x-induced ozone production;
  2) stratospheric ozone *decreases* due to the conversion of active chlorine to the reservoir HCl by the reaction CH_4+Cl->CH_3 +HCl;
  3) upper strat./mesospheric ozone *decreases* due to the increase in H_2O and HO_x-ozone oss.
- The CH_4-induced HO_x-ozone loss dominates the 60 km ozone changes during 1860-2100, and is significant prior to 1950 (Figure 2, top panel).
- The net effect of CH_4 loading is to increase total column ozone throughout 1860-2100; the future CH_4 impact is larger in 2055 than in 2095 following the surface boundary condition (Figure 1).
- The net effect of all sources in the 2D full simulation is a 1.6% increase in global total ozone from 1860-2100 (Figure 4). However in the tropics (Figure 5), total ozone in the full simulation increases from 2000-2050, but then decreases by ~1% over the last half of the 21st century.
- The 2D model ozone changes in the full simulation are in mostly good agreement with the observations and the 3D GEOSCCM full simulation in Figures 2-5.

3. 1860-1960 Ozone:

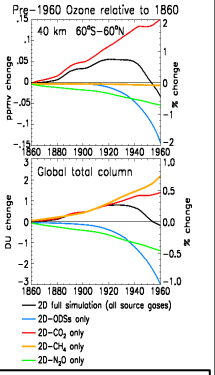

Figure 6. Ozone time series for 1860-1960, relative to 1860, for the 60°S-60°N average at 40 km (top) and the global average total column (bottom) from the model simulations indicated. Values are in ppmv or Dobson unit (DU) change (left axes) and percent change (right axes).

Major points:

- ODS loading, driven mainly by CCl_4 emissions, causes a 1% total column depletion by 1960.
- By 1960, the total column changes due to CO_2 and N_2O are roughly equal and opposite (+0.5%,-0.5%).
- CH_4 loading causes +0.75% increase in total ozone from 1860-1960.
- At 40 km, CO_2 cooling induces a 2% ozone increase from 1860-1960, approximately equal and opposite to the ozone depletion caused by ODSs.
- Ozone in the full simulation reaches a broad maximum during 1920-1940. This precedes the rapid decline after ~1970 driven mainly by the ODS loading (Figures 2 and 4).

6. Time Dependent

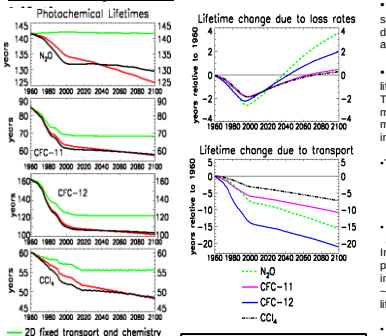

Figure 9. Time dependent photochemical lifetimes of N_2O, CFC-11, CFC-12, and CCl_4 from the 2D model with the chemistry and transport fixed at 1960 levels (green curves); the chemistry fixed at 1960 levels (red curves); and the full simulation (black curves).

Figure 10. (Top) Difference between the full and fixed chemistry simulations (black minus red curves in Figure 9), showing the lifetime changes due to the changing loss rates. **(Bottom)** Difference between the fixed chemistry and fixed chemistry and transport simulations (red minus green curves in Figure 9), showing the lifetime changes due to changes in the Brewer-Dobson circulation transport.

Major points:

- The lifetime is important in determining the length of time over which a molecule of a substance will have a significant impact on ozone depletion or global warming, and in deriving surface mixing ratio boundary conditions from emissions estimates for use in atmospheric models.
- The black curves in Figure 9 show the full 2D model simulation, and the present day lifetimes are 132, 61, 108, and 51 years, respectively for N_2O, CFC-11, CFC-12, and CCl_4. These are older than those cited in WMO-2007: 114, 45, 100, and 26 years. This is due mainly to the older age of air simulated by the 2D model (Figure 8 above), compared to the models used to obtain the lifetimes for WMO-2007. Also, our older lifetime for CCl_4 does not include soil or ocean losses.
- The lifetimes are controlled by:
  1) the photochemical loss rates (photolysis, reaction with $O(^1D)$;
  2) the rate of transport through the stratospheric loss region;
  3) the changing atmospheric burden (total number of molecules).
- The green curves in Figure 9 are from a simulation with the transport and chemistry fixed at 1960 levels, showing the effect of the changing atmospheric burden of each compound. In this simulation, the computed lifetimes of CCl_4 and the CFCs decrease during the latter part of the 20th century due to the disequilibrium between the surface emissions, which are increasing rapidly from zero, and the stratospheric loss. As this equilibrium is reached after ~2020, the lifetimes become constant . N_2O has a significant background source and it's lifetime is not impacted by this effect.
- The lifetime impact due to the changing chemical loss rates is shown in Figure 10 (top). This is the difference between the full simulation and the simulation with the chemistry fixed at 1960 levels (black minus red curves in Figure 9). This effect follows the changing overhead burden of ozone, with lifetime decreases of ~ 2 years from 1960-2000, and increases of 2-6 years from 2000-2100. These changes are all small (<4%) over the 1960-2100 period.
- The impact due to the changing transport is shown in Figure 10 (bottom). This is the difference between the fixed chemistry and fixed chemistry and transport simulations (red minus green curves in Figure 9). This follows the changes in the BDC (Figure 8), and these lifetime decreases due to the transport changes are ~12% over the 1960-2100 time period for all four compounds.

7. MAIN CONCLUSIONS:

- **Time dependent changes in the emissions of CO_2, CH_4, N_2O, and the ODSs have a substantial impact on the time evolution of the stratosphere throughout the 1860-2100 time period.**
- **The stratospheric ozone changes during 1860-1960 were small compared to 1960-2000, but not insignificant. Ozone reached a broad maximum during 1920-1940 due to the combined effects of the GHG and ODS loading. This preceded the rapid decline during ~1970-2000 driven mainly by ODSs.**
- **By 2100, CO_2, CH_4, and N_2O will all have significant impacts on global total ozone; CO_2 will have the largest individual impact, causing a 4% increase from 1860-2100.**
- **Model calculations show that the time dependent photochemical lifetimes of N_2O, CFC-11, CFC-12, and CCl_4 all decrease by ~12% during 1960-2100 due to the acceleration of the Brewer-Dobson circulation; smaller lifetime changes of <4% occur due to the changing loss rates.**
- **Comparisons of the 2D model with observations and the Goddard 3D chemistry-climate model (GEOSCCM) illustrate that the 2D model captures the basic processes responsible for long term stratospheric changes.**

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