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# Effects of stratospheric ozone recovery on tropospheric chemistry and air quality

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## Abstract

The stratospheric ozone has decreased greatly since 1980 due to ozone depleting substances (ODSs). As a result of the implementation of the Montreal Protocol and its amendments and adjustments, stratospheric ozone is expected to recover towards its pre-1980 level in the coming decades. We examine the implications of stratospheric ozone recovery for the tropospheric chemistry and ozone air quality with a global chemical transport model (GEOS-Chem). Significant decreases in surface ozone photolysis rates due to stratospheric ozone recovery are simulated. Increases in ozone lifetime by up to 7 % are calculated in the troposphere. The global average OH decreases by 1.74 % and the global burden of tropospheric ozone increases by 0.78 %. The perturbations to tropospheric ozone and surface ozone show large seasonal and spatial variations. General increases in surface ozone are calculated for each season, with increases by up to 5 % for some regions.

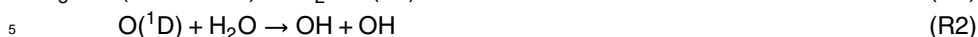
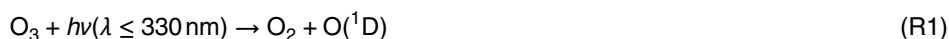
## 1 Introduction

Significant decreases in stratospheric ozone driven by ozone depleting substances (ODSs), in particular over the high latitudes, have been observed since the 1980s (Farman et al., 1985). The decreases in stratospheric ozone have allowed increasing solar UV radiation, especially the UV-B (280–315 nm) radiation to reach into the troposphere and Earth surface. Previous studies (McKenzie et al., 1991; Bais et al., 1993; Kerr and McElroy, 1993) have shown the general increases in ground-level solar UV-B radiation associated with the reduction of stratospheric ozone at various sites around the world.

Stratospheric ozone is expected to recover towards its pre-1980 level (generally defined as the ozone recovery level) due to the reduction in ODSs as a result of the implementation of Montreal Protocol as well as its Amendments and Adjustments (WMO, 2003). The change in solar UV-B radiation associated with the stratospheric ozone recovery would alter the tropospheric chemistry through its effect on photolysis rates

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of many tropospheric gas species. First, the photochemical destruction of tropospheric ozone would be affected. The photolysis of ozone followed by reaction with water vapor is the most important pathway for ozone destruction in the troposphere:



The above reactions also provide the dominant source for hydroxyl radical (OH) in the troposphere which controls the oxidizing capacity of the troposphere and hence the lifetimes of many atmospheric species (Thompson, 1992). On the other hand, the changes in UV radiation can also affect the photochemical production of ozone through the photolysis of  $\text{NO}_2$ :



Liu and Trainer (1988) used a box model to study the response of tropospheric ozone and OH to increasing UV radiation due to total ozone reduction with fixed  $\text{NO}_x$  concentration. It was found that tropospheric ozone changes in the same direction as column ozone while in polluted regions, an inverse relationship was seen. In addition, the change of tropospheric OH was independent of  $\text{NO}_x$  level. Thompson et al. (1989) also found that tropospheric ozone will increase with stratospheric ozone depletion at high  $\text{NO}_x$  level and decrease at low  $\text{NO}_x$  level applying a one-dimension model. Fuglestedt et al. (1994) found that the decline of stratospheric ozone would result in an increase in tropospheric OH and a general reduction of tropospheric  $\text{O}_3$  using model simulated stratospheric ozone. Schnell et al. (1991) showed that a 17% decrease in measured surface ozone concentrations at the South Pole in the austral summer was mostly caused by the increases in surface radiation resulting from stratospheric ozone hole where  $\text{NO}_x$  level is low.

The changes in tropospheric chemistry associated with the stratospheric ozone recovery also imply perturbations to ozone air quality. Ground level ozone has been regarded as one of the six criteria air pollutants by EPA as it could cause respiratory

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health problems (Desqueyroux et al., 2002; Peden, 2001) and damage vegetation and the ecosystem (Mauzerall and Wang, 2001; Oksanen and Holopainen, 2001). Because of the inhomogeneous distribution of  $\text{NO}_x$  and NMHCs and the nonlinear chemistry, the response of ground level ozone to change in surface UV radiation should vary between different locations depending on the  $\text{NO}_x$  level as studied by (Isaksen et al., 2005).

In this study, we investigate the responses of tropospheric chemistry and surface ozone air quality to stratospheric ozone recovery by carrying out sensitivity studies with a global chemical transport model.

## 2 Methodology and model description

GEOS-Chem is a global 3-D chemical transport model, which is driven by meteorological data from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation office. Here we use GEOS-Chem version v8-03-01 driven by GEOS-5 meteorology with temporal resolution of 6 h (3 h for surface meteorological variables).

In this study, the simulations are run at a resolution of 4° latitude by 5° longitude with 47 vertical layers. We use the tropospheric Ozone- $\text{NO}_x$ -VOC full chemistry simulation with GEOS-Chem, which has been described in previous studies (Yuhang et al., 1998a, b; Wang et al., 1998). The Linoz stratospheric ozone chemistry package (McLinden et al., 2000) is used. Photolysis rates in the troposphere are calculated using fast-J mechanism, which takes into account the effects of clouds and aerosol on photolysis rates (Wild et al., 2000). 7 wavelength bins with different widths cover the wavelength range from 291 nm to 850 nm. Below 289 nm, Fast-J2 was developed to compute the photolysis rates in stratosphere (Bian and Prather, 2002).

To derive the sensitivity of troposphere photochemistry to stratospheric ozone recovery, two groups of simulations are performed: one as the control run and the other as the sensitivity run where the stratospheric ozone is assumed to fully recover to its pre-1980 levels. For both simulations, a whole year spin-up using 2005 meteorology was

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done followed by a three year simulation using meteorology for 2006 to 2008. Unless noted otherwise, all the model results discussed in this study refer to the 3 yr averages. We follow the WMO report (Randel, 2003; Fioletov, 2006) for the stratospheric ozone depletion in the past decades, which varies significantly with season and latitudes. The expected increases in stratospheric ozone column associated with the ozone recovery in the coming decades are shown in Table 1.

### 3 Results

To quantify the responses of photolysis rates, tropospheric chemistry, and surface ozone to stratospheric ozone recovery, a sensitivity analysis was performed by using the sensitivity factor as defined by (Fuglestedt et al., 1994):

$$S_x = \frac{\frac{\Delta X}{X}}{\frac{\Delta \sum O_3}{\sum O_3}} \quad (1)$$

Where  $X$  is the variable of interest (e.g, it can be the photolysis rates, concentrations or global burden of any tropospheric species),  $\sum O_3$  is the total column ozone.  $\Delta X$  and  $\Delta \sum O_3$  represents the differences between results from the sensitivity run and the control run. Therefore, the value of  $S_x$  indicates the percentage change in  $X$  resulting from 1% increase in total ozone column.

#### 3.1 Impacts on photolysis rates in troposphere

The photolysis of  $O_3$  and  $NO_2$  are critical for the chemical destruction and production of troposphere ozone respectively, therefore we focus our analysis on the photolysis rates of these two species –  $J(O_3)$  and  $J(NO_2)$ . Figures 1 to 4 show the zonal mean average  $J(O_3)$  in winter months, spring months, summer months and fall months. Tables 2 and 3 show the model simulated changes in  $J(O_3)$  and  $J(NO_2)$  resulted from the

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stratospheric ozone recovery as well as their sensitivity factors for different latitudes and seasons. The largest percentage change of ( $J(O_3)$ ) by up to 22% is found in spring time over the southern polar region associated with the strong stratospheric ozone recovery expected.

All of the sensitivity factors show negative values indicating that the stratospheric ozone recovery would lead to less solar radiation in the troposphere as expected. The calculated  $S_x$  for  $J(O_3)$  is about 20–80 times greater than  $S_x$  for  $J(NO_2)$ , reflecting the different absorption spectrum for  $O_3$  and  $NO_2$ . This implies that the stratospheric ozone recovery would have much stronger effects on the photochemical ozone destruction than the ozone production in the troposphere. The absolute values for  $J(O_3)$  are always larger than 1, implying that the percentage change of  $J(O_3)$  would be always larger than the percentage change for stratospheric ozone.

#### 3.2 Impacts on tropospheric ozone and OH

Due to the reduced photolysis rates in the troposphere associated with the stratosphere ozone recovery, the global tropospheric ozone burden is calculated to increase by 0.78% (from  $383 \text{ Tg yr}^{-1}$  in the control run to  $386 \text{ Tg yr}^{-1}$  in the sensitivity run). In the mean time, the global average tropospheric OH concentration is found to decrease by 1.74% (from  $1.15 \times 10^6 \text{ molecules cm}^{-3}$  in the control run to  $1.13 \times 10^6 \text{ molecules cm}^{-3}$ ).

Figures 5 to 8 show the zonal mean concentrations for tropospheric ozone as well as the impacts due to stratospheric ozone recovery for each season. As we can see from the figures, the effects of stratospheric ozone recovery present a seasonal and spatial variation.  $O_3$  concentration in the lower troposphere will be mostly affected. The largest increases by up to 4% are found in the lower troposphere over southern mid-latitudes in summer time.

Figures 9 and 10 show the annual mean surface OH concentration and annual zonal mean OH concentration respectively. Surface OH decreases on a global scale as a result of reduced solar radiation onto the surface of the earth because of stratospheric ozone recovery that leads to reduced ozone photolysis to produce OH. OH in the tro-

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posphere also decreases, which should be also due to the less penetration of solar radiation into the troposphere.

The changes in photolysis rates of ozone would affect the life time of tropospheric ozone against chemical destruction. The calculated average ozone lifetime in the troposphere are 32.39 days and 32.79 days for control run and sensitivity run, respectively, with a 1.23 % increase. In the lower troposphere over the oceans, the chemical lifetime of ozone increases by up to 7 %. The longer life time would enhance the intercontinental transport of ozone.

### 3.3 Impacts on surface ozone air quality

General increases in surface ozone due to stratospheric ozone recovery are calculated for each season (Figs. 11 to 14). The largest increases (up to 5 %) are always found over remote ocean regions. The largest percentage change of surface ozone is found in summer time while the least change in winter time, reflecting the strongest photochemical activities during summer time.

## 4 Conclusions and discussions

We examine the potential impacts on tropospheric chemistry and ozone air quality from the expected stratospheric ozone recovery in the coming decades with a chemical transport model. With the full recovery of stratospheric ozone to its pre-1980 level, we find significant decreases in photolysis rates for tropospheric ozone, with the surface  $O_3$  photolysis rates decrease by up to 22 %. The sensitivity factor for surface ozone photolysis rate, defined as the percentage changes in surface ozone photolysis rate for 1 % change in stratospheric ozone, is always larger than one. Compared to tropospheric ozone, tropospheric  $NO_2$  shows much weaker sensitivity to stratospheric ozone recovery. As a consequence, the stratospheric ozone recovery would lead to de-

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creases in both the photochemical destruction and production of tropospheric ozone, but the decreases in destruction would be much stronger than that for production.

The global tropospheric ozone burden is calculated to increase by 0.78 % while the average tropospheric OH concentration would decrease by 1.74 %. The perturbations to tropospheric chemistry and surface ozone show strong seasonal and spatial variations. The largest perturbations are generally found in the lower troposphere and during summer time when there are strong photochemical activities. General increases in surface ozone are calculated for each season associated with the stratospheric ozone recovery, in particular over oceans where surface ozone can increase by up to 5 % during summertime. The general increases in background ozone and the longer lifetime of tropospheric ozone imply that the stratospheric ozone recovery would enhance the intercontinental transport of ozone pollution and therefore has significant implications for air quality policy and management.

In this sensitivity study, we focus on the effects of stratospheric ozone recovery on tropospheric chemistry and ozone air quality through the changes in photochemical reactions. The recovery of stratospheric ozone could also affect tropospheric ozone by enhancing the ozone flux into the troposphere through stratosphere–troposphere exchange (STE) (Zeng et al., 2010) but this effect is not accounted for in this study. The STE effect could be particularly important for ozone in the upper troposphere while our study show that the photochemical effect is very important for ozone in the lower troposphere and surface air.

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**Table 1.** Increase in stratospheric ozone column due to expected ozone recovery in the sensitivity run.

	90° S–60° S	60° S–35° S	35° S–25° S	25° S–25° N	25° N–35° N	35° N–60° N	60° N–90° N
DJF <sup>a</sup>	4 %	6.3 %	3.15 %	1 %	2.8 %	5.6 %	12 %
MAM <sup>a</sup>	4 %	5.4 %	2.7 %	1 %	3.0 %	6 %	8 %
JJA <sup>a</sup>	16 %	5.9 %	2.95 %	1 %	1.75 %	3.5 %	4 %
SON <sup>a</sup>	16 %	6.2 %	3.1 %	1 %	2.1 %	4.2 %	4 %

<sup>a</sup> DJF = December–February for Northern Hemisphere winter; MAM = March–May for Northern Hemisphere spring; JJA = June–August for Northern Hemisphere summer; SON = September–November for Northern Hemisphere fall.

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**Table 2.** Change in surface O<sub>3</sub> and NO<sub>2</sub> photolysis rates due to stratospheric ozone recovery (%).

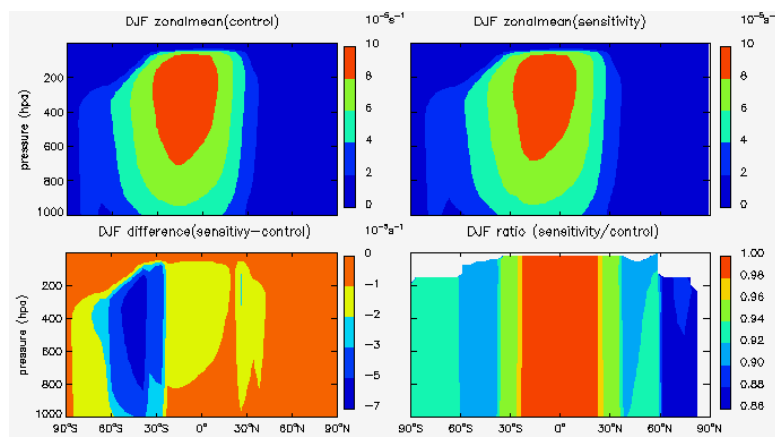
	90° S–60° S	60° S–35° S	35° S–25° S	25° S–25° N	25° N–35° N	35° N–60° N	60° N–90° N
DJF ( <i>J</i> O <sub>3</sub> )	-6.31	-7.79	-4.17	-1.45	-4.16	-7.79	-12.94
MAM ( <i>J</i> O <sub>3</sub> )	-5.79	-6.61	-3.73	-1.53	-4.25	-8.60	-11.03
JJA ( <i>J</i> O <sub>3</sub> )	-18.29	-6.80	-4.27	-1.45	-2.42	-4.98	-5.93
SON ( <i>J</i> O <sub>3</sub> )	-21.78	-7.85	-4.27	-1.44	-3.00	-6.02	-5.62
DJF ( <i>J</i> NO <sub>2</sub> )	-0.097	-0.110	-0.058	-0.020	-0.052	-0.131	-0.449
MAM ( <i>J</i> NO <sub>2</sub> )	-0.114	-0.098	-0.050	-0.021	-0.057	-0.125	-0.217
JJA ( <i>J</i> NO <sub>2</sub> )	-0.513	-0.116	-0.056	-0.019	-0.034	-0.069	-0.095
SON ( <i>J</i> NO <sub>2</sub> )	-0.330	-0.115	-0.058	-0.020	-0.039	-0.084	-0.110

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**Table 3.** Sensitivity factors of average surface  $J(O_3)$  and  $J(NO_2)$ .

	90° S–60° S	60° S–35° S	35° S–25° S	25° S–25° N	25° N–35° N	35° N–60° N	60° N–90° N
DJF ( $J(O_3)$ )	-1.58	-1.24	-1.32	-1.45	-1.48	-1.39	-1.08
MAM ( $J(O_3)$ )	-1.45	-1.22	-1.38	-1.53	-1.42	-1.43	-1.38
JJA ( $J(O_3)$ )	-1.14	-1.15	-1.45	-1.45	-1.39	-1.42	-1.48
SON ( $J(O_3)$ )	-1.36	-1.27	-1.38	-1.44	-1.43	-1.43	-1.40
DJF ( $J(NO_2)$ )	-0.024	-0.017	-0.018	-0.020	-0.019	-0.024	-0.037
MAM ( $J(NO_2)$ )	-0.028	-0.018	-0.021	-0.019	-0.019	-0.021	-0.027
JJA ( $J(NO_2)$ )	-0.032	-0.020	-0.019	-0.019	-0.019	-0.020	-0.024
SON ( $J(NO_2)$ )	-0.021	-0.018	-0.019	-0.020	-0.019	-0.020	-0.027

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**Fig. 1.** DJF average zonalmean  $J(O_3)$  due to stratospheric  $O_3$  recovery.

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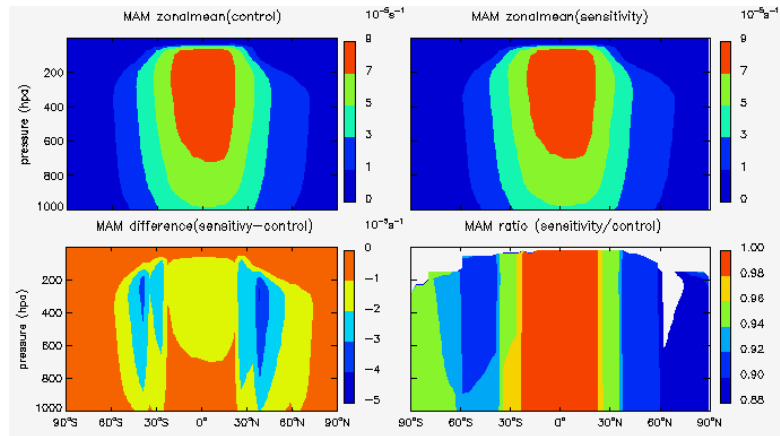


Fig. 2. MAM average zonalmean  $J(O_3)$  due to stratospheric  $O_3$  recovery.

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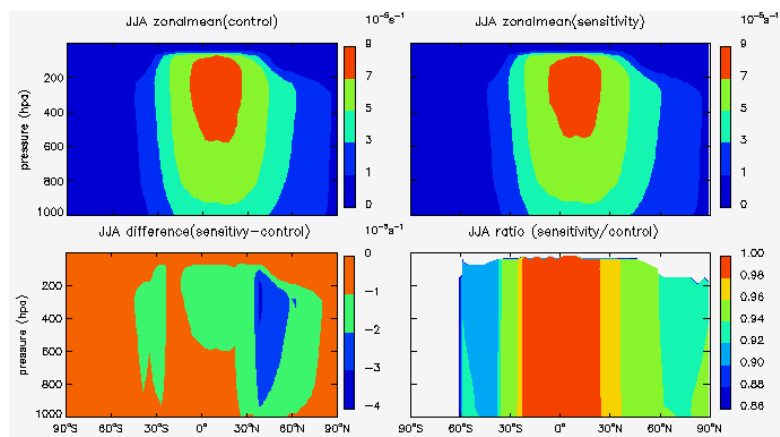


Fig. 3. JJA average zonalmean  $J(O_3)$  due to stratospheric  $O_3$  recovery.

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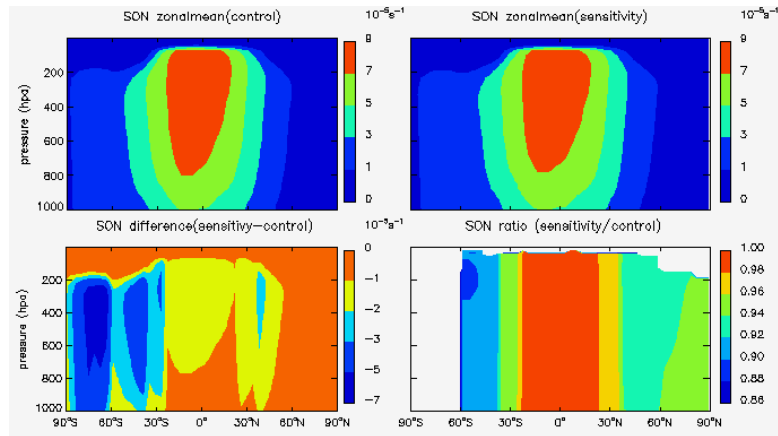


Fig. 4. SON average zonalmean  $J(O_3)$  due to stratospheric  $O_3$  recovery.

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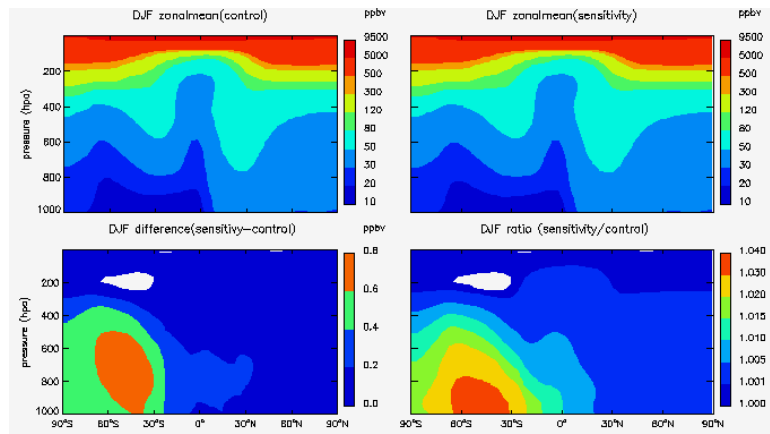


Fig. 5. DJF average zonalmean  $O_3$  concentration due to stratospheric  $O_3$  recovery.

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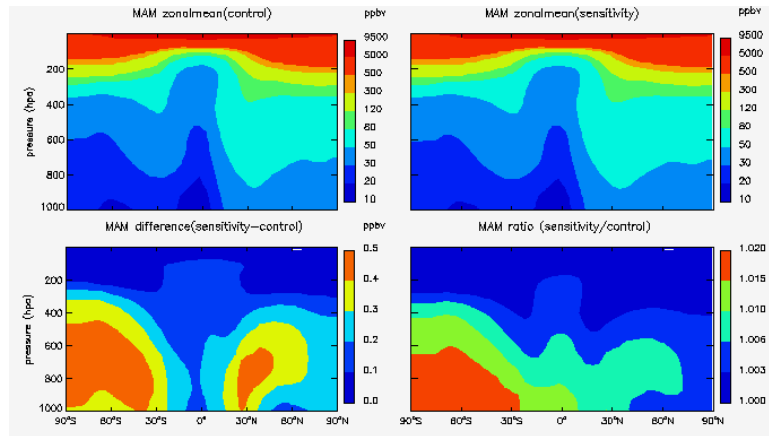


Fig. 6. MAM average zonalmean O<sub>3</sub> concentration due to stratospheric O<sub>3</sub> recovery.

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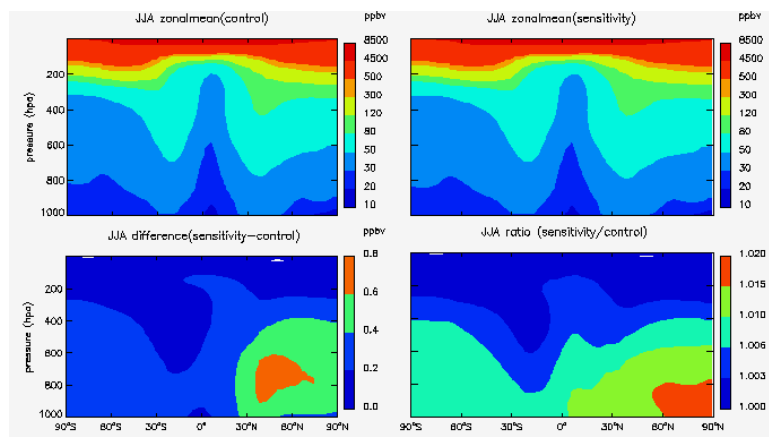


Fig. 7. JJA average zonalmean O<sub>3</sub> concentration due to stratospheric O<sub>3</sub> recovery.

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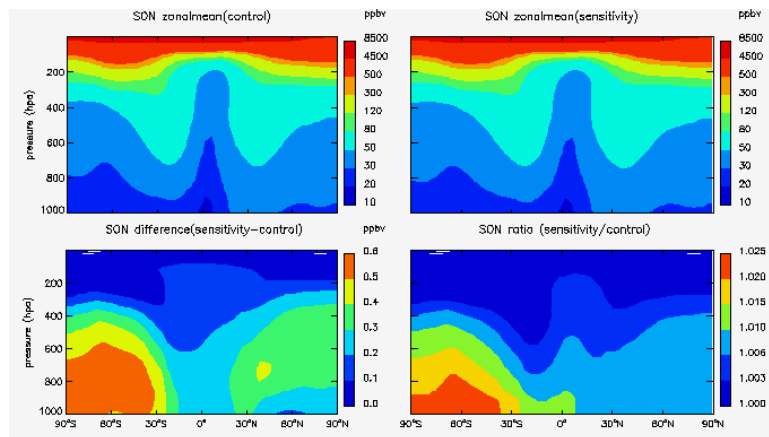


Fig. 8. SON average zonalmean O<sub>3</sub> concentration due to stratospheric O<sub>3</sub> recovery.

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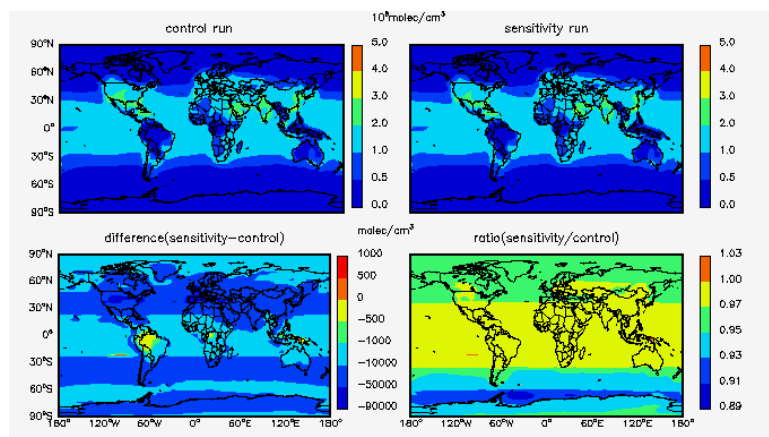


Fig. 9. Annual mean surface OH concentration due to stratospheric O<sub>3</sub> recovery.

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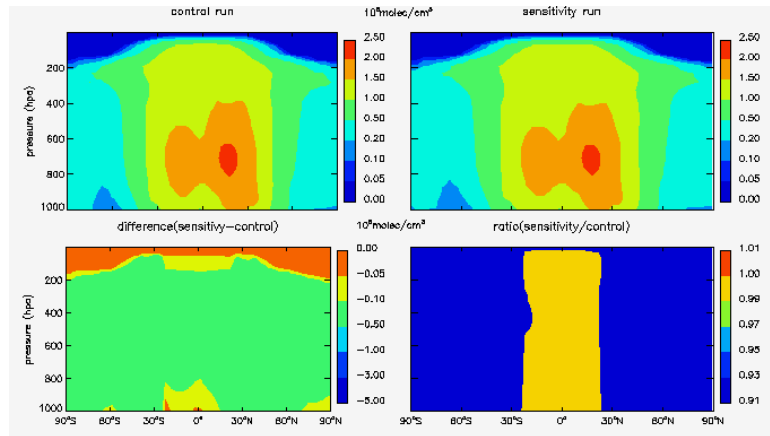


Fig. 10. Annual zonal mean OH concentration due to stratospheric O<sub>3</sub> recovery.

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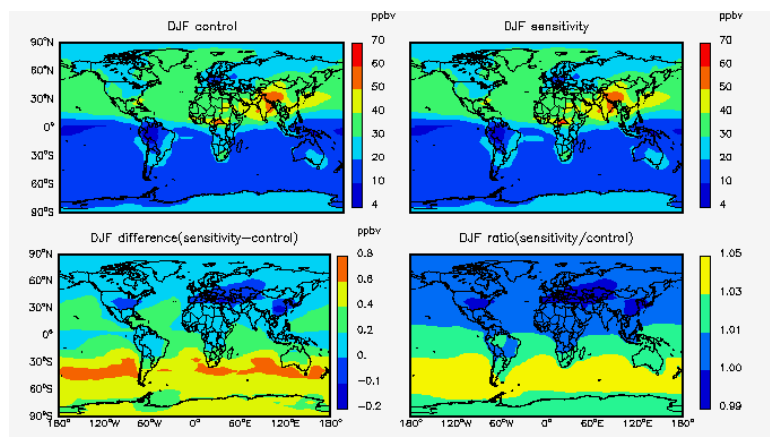


Fig. 11. DJF average surface O<sub>3</sub> concentration due to stratospheric O<sub>3</sub> recovery.

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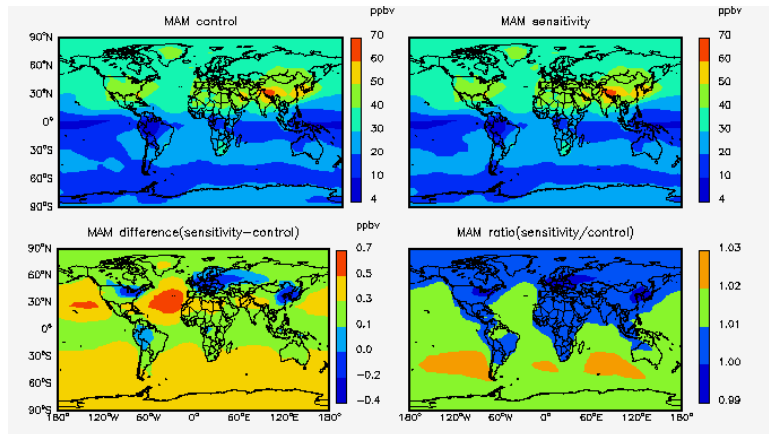


Fig. 12. MAM average surface O<sub>3</sub> concentration due to stratospheric O<sub>3</sub> recovery.

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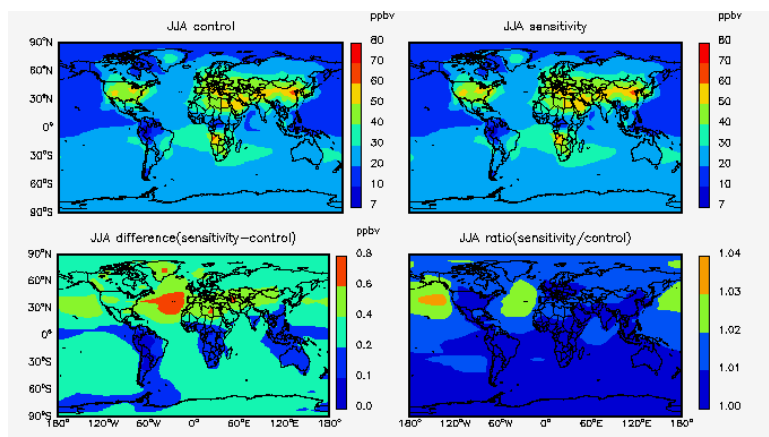
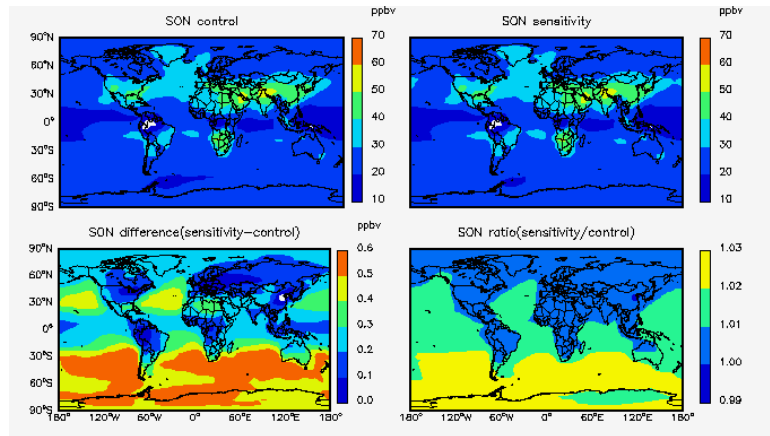


Fig. 13. JJA average surface O<sub>3</sub> concentration due to stratospheric O<sub>3</sub> recovery.

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**Fig. 14.** SON average surface  $O_3$  concentration due to stratospheric  $O_3$  recovery.