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Stratospheric ozone depletion from future nitrous oxide increases

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Abstract

We have investigated the impact of assumed nitrous oxide (N_2O) increases on stratospheric chemistry and dynamics by a series of idealized simulations. In a future cooler stratosphere the net yield of NO_y from a changed N_2O is known to decrease, but NO_y can still be significantly increased by the increase of N_2O . Results with a coupled chemistry-climate model (CCM) show that increases in N_2O of 50%/100% between 2001 and 2050 result in more ozone destruction, causing a reduction in ozone mixing ratios of maximally 6%/10% in the middle stratosphere at around 10 hPa. This enhanced destruction could cause an ozone decline in the second half of this century in the middle stratosphere. However, the total ozone column still shows an increase in future decades, though the increase of 50%/100% in N_2O caused a 2%/6% decrease in TCO compared with the reference simulation. N_2O increases have significant effects on ozone trends at 20–10 hPa in the tropics and at northern high latitude, but have no significant effect on ozone trends in the Antarctic stratosphere. The ozone depletion potential for N_2O in a future climate depends both on stratospheric temperature changes and tropospheric N_2O changes, which have reversed effects on ozone in the middle and upper stratosphere. A 50% CO_2 increase in conjunction with a 50% N_2O increase cause significant ozone depletion in the middle stratosphere and lead to an increase of ozone in the upper stratosphere. Based on the multiple linear regression analysis and a series of sensitivity simulations, we find that the chemical effect of N_2O increases dominates the ozone changes in the stratosphere while the dynamical and radiative effects of N_2O increases are insignificant on average. However, the dynamical effect of N_2O increases may cause large local changes in ozone mixing ratios, particularly, in the Southern Hemisphere lower stratosphere.

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5 efficiency of N₂O for global ozone depletion is expected to decrease in the stratosphere due to projected increases in CO₂ (Rosenfield and Douglass, 1998). Some other studies argued that CO₂-induced cooling could cause stratospheric ozone to recover to values greater than 1980 levels during this century (Chipperfield, 2009; Wuebbles, 2009) and lead to a so-called “super recovery”. Fleming et al. (2011) further carried out a comprehensive study of long-term stratospheric effects of some source gases including N₂O and CO₂ through series of two-dimensional (2-D) chemistry-climate model simulations. They predicted that in the latter half of the 21st century CO₂, N₂O, and CH₄ loading will all have significant impacts on global total ozone while the effect of CO₂ loading on global total ozone has twice the magnitude of the effect of N₂O.

10 Although previous studies have investigated the impact of N₂O on ozone depletion, the relative importance of the radiative and chemical effect of N₂O remains unclear and the net effect of increased N₂O on the ozone layer in the changing climate is still under debate. Previous studies have mostly been based on mechanistic analysis, with the main focus being the chemical effect of N₂O (Ravishankara et al., 2009), or have used relatively simple two-dimensional chemical-dynamical models (e.g. Portmann and Solomon, 2007; Chipperfield, 2009; Fleming et al., 2011) to diagnose the effect of N₂O changes on the ozone layer. In this study, we re-examine the effect of N₂O increases on ozone using a fully coupled 3-D chemistry-climate model (CCM). We attempt to clarify 20 quantitatively the radiative and chemical effects of N₂O increases on the ozone layer and to diagnose the ozone changes in a changing climate when CO₂ and N₂O are both increasing. The details of the model and numerical experiments are given in Sect. 2. The effect of N₂O increases on ozone depletion under different scenarios is addressed in Sect. 3. In Sect. 4, we analyze contributions of various factors associated with N₂O increases in modulating ozone depletion. Conclusions and a discussion are presented in Sect. 5.

2 Models and numerical experiments

We have used a fully coupled chemistry-climate model, the Whole Atmosphere Community Climate Model 3 (WACCM3) to investigate the chemical and thermal-dynamical responses to a linear future increase in N_2O . WACCM3 has 66 vertical levels from the ground to 4.5×10^{-6} hPa (approximately 145 km geometric altitude) with detailed interactive chemistry (Garcia et al., 2007) and a finite volume dynamical core (Lin, 2004). For this study simulations were carried out at $4^\circ \times 5^\circ$ horizontal resolution. The WACCM3 chemistry module is derived from the three-dimensional (3-D) chemical transport Model for Ozone and Related chemical Tracers (MOZART) (Brasseur and Hitchman, 1988; Hauglustaine et al., 1998; Horowitz et al., 2003). More details of WACCM3 model can be found in Garcia et al. (2007).

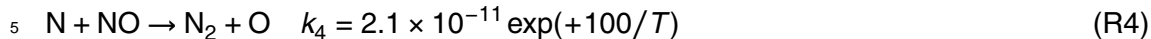
Four long-term WACCM simulations were performed in this study and detailed configurations of those simulations are listed in Table 1. The surface volume mixing ratios (vmrs) of GHGs in the control run (E0) are taken from IPCC A1b scenario. In runs E1 and E2, the surface vmrs of N_2O were increased linearly with time from 2001 to 2050 at a rate of $1\% \text{ yr}^{-1}$ in run E1 and $2\% \text{ yr}^{-1}$ in run E2. More specifically, N_2O values in E1 are increased by 1 % in 2001, 2 % in 2002, and 50 % in 2050. It should be pointed out that the percentage increases of N_2O in each year are based on N_2O values of A1B scenario. For instance, in year 2050, N_2O value of A1B scenario is 344 ppbv in run E0, therefore, it is 517 ppbv in run E1 and 688 ppbv in run E2. This increasing approach makes the N_2O variation in our simulations close to A1B scenario during the first half of the simulations with small additional increases in each year compared to A1B scenario, but far off A1B scenario with much larger N_2O increases during the later half of the simulations for the purpose of amplifying response signals of the atmosphere. In run E3, both N_2O and CO_2 are increased linearly with time from 2001 and increased by 50 % in 2050 with an increasing rate of $1\% \text{ yr}^{-1}$. The sea surface temperature (SST) and sea ice fields used in four runs are the same (HadGEM1) and derived from the SST and sea ice fields prepared for the Chemistry-Climate Model Validation activity 2

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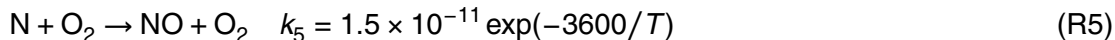
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Only the minor channel of Reaction (R3) leads to NO_y ($= \text{NO}_x + \text{NO}_3 + \text{HNO}_3 + \text{HO}_2\text{NO}_2 + \text{ClONO}_2 + 2\text{N}_2\text{O}_5 + \text{BrONO}_2 + \dots$) production. NO_y is removed from the stratosphere by transport to the troposphere and washout or by local photochemical loss at high altitudes via:



The abundance of N is determined by photolysis of NO and by the reaction:



Because of the temperature dependence of the reactions controlling the chemical loss of NO_y , NO_y tends to decrease in a future cooler stratosphere as Reaction (R4) speeds up while Reaction (R5) slows down (Rosenfield and Douglass, 1998). Without significant stratospheric cooling, N_2O increases in the atmosphere will cause NO_y and NO_x increases in the stratosphere. Figure 2 shows the time series of global mean NO_x and NO_y fields from the four experiments and corresponding changes in global mean column NO_x and NO_y in runs E1, E2 and E3 relative to that in control run E0. Note that a 100% increase in N_2O by the end of 2050 causes an increase in the global mean column NO_y and NO_x by $12 \times 10^{15} \text{ molecules cm}^{-2}$ and $4 \times 10^{15} \text{ molecules cm}^{-2}$, respectively. However, when CO_2 is also increased by 50% in conjunction with a 50% N_2O increase at the end of 2050 in run E3, which implies a cooler stratosphere than that in run E1 in which only N_2O is increased by 50% at the end of 2050, the NO_y and NO_x increases are smaller than those in run E1 (Fig. 1a and c). Even though the yield of NO_x from N_2O decreases in a cooler stratosphere, the efficiency of ozone loss due to NO_y increases so that N_2O increases in run E3 still causes an increase in ozone loss due to the increasing NO_y in an atmosphere with decreasing halogens. These results imply that the ozone depletion potential for N_2O in a future climate will actually depend both on stratospheric temperature changes and tropospheric N_2O changes, which have reversed effects on ozone in the middle and upper stratosphere. Figure 1 clearly indicates that the destruction of ozone associated with N_2O increases in run E3

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is reduced relative to that in runs E1, particularly in the middle and upper stratosphere, consistent with previous findings that CO₂-induced cooling tends to increase ozone in the future (e.g. Haigh and Pyle, 1982; Shepherd, 2008; Chipperfield, 2009; Wuebbles, 2009; Oman et al., 2010a) by slowing down the gas-phase chemical reactions associated with ozone.

In the lower stratosphere, increased N₂O could reduce the efficiency of Cl_y and Br_y for ozone destruction by tying up more of these halogens in the ClONO₂ and BrONO₂ reservoir species, consequently, N₂O increases lead to more ozone. Meanwhile, projected decreases in Cl_y and Br_y tend to tie up less NO_y in ClONO₂ and BrONO₂, and increase the efficiency of N₂O in ozone destruction processes (e.g. Ravishankara et al., 2009; Daniel et al., 2010), but Fig. 1 suggests that this effect is outweighed by the effect of N₂O increases. On the other hand, in the lower stratosphere/upper troposphere UV radiation is enhanced due to the ozone decreases in the middle and upper stratosphere, resulting in more ozone production through the so-called “self-healing” effect. A notable feature in Fig. 1 is that this ozone increase in the lower stratosphere/upper troposphere can be seen mostly at Southern Hemisphere mid-latitudes. Ravishankara et al. (2009) pointed out that the “self-healing” effect is most significant at tropical latitudes. As will be discussed later, this significant ozone increase in the Southern Hemisphere is partly caused by the enhanced vertical transport of ozone from the upper atmosphere to the lower stratosphere/upper troposphere, which may not be well simulated in 2-D models. Below 100 hPa in the troposphere, N₂O increases result in more ozone with a maximum 10 % increase at southern high latitudes when N₂O is increased by 100 % from 2001 to 2050 (Fig. 1b).

Due to significant ozone decreases in the middle and upper stratosphere caused by increased N₂O, the TCO in the two WACCM simulations (E1 and E2) decreases compared with control run E0 (Fig. 1d and e), particularly at high latitudes. A 50 % increase in N₂O causes a maximum 6 % decrease in TCO (Fig. 1d). When N₂O is increased by 100 % in run E2, TCO changes become larger with a maximum decrease of 10 % at high latitudes. The combined effect of N₂O and CO₂ increases results in

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a slight TCO decrease (1–2 %) in the Northern Hemisphere but an increase (2–6 %) in the Southern Hemisphere (Fig. 1f), due to the offsetting impact of CO₂-induced cooling.

Figure 3a–c shows modeled ozone trends as a function of latitude and pressure from 2000 to 2050 under different assumed N₂O changes. Also shown are the time series of TCO averaged over different latitude bands, i.e., 90° N–70° N, 22° S–22° N, and 70° S–90° S. Consistent with results in previous studies (e.g. Tian and Chipperfield, 2005; Austin and Wilson, 2006; Newman et al., 2006; Eyring et al., 2007; WMO, 2007), the global ozone will generally recover in the next half century under the IPCC A1b GHGs scenario with a maximum increasing trend of 0.3 ppmv/10 yr at 10–1 hPa (Fig. 3a). However, if N₂O is further increased by 50 % relative to the IPCC A1b projection, the ozone in the middle stratosphere at around 20 hPa decreases with a maximum trend of –0.2 ppmv/10 yr, although the ozone in the upper stratosphere still increases (Fig. 3b). Note that a steadily declining trend in ozone in the tropical lower stratosphere was also simulated in previous simulations (Oman et al., 2010a; SPARC-CCMVal, 2010; Eyring et al., 2010a), but there is no significant ozone trend in the tropical lower stratosphere in our simulations through 2050. A possible reason is that the upwelling in our 50 yr simulations has not significantly increased in the lower stratosphere/upper troposphere (Table 3) though we used an increased GHG scenario and time varying SSTs. As in Fig. 1, CO₂ increases tend to offset the effect of N₂O on ozone trends in the middle stratosphere (Fig. 3c). When N₂O and CO₂ are both increased by 50 %, the declining trend of ozone in the middle stratosphere becomes less significant. It is worth noting that the effect of increased N₂O on long-term trends of ozone mixing ratios at about 20–10 hPa have evident spatial variations. Ozone mixing ratios decrease most significantly in the tropics with a trend up to –0.1 ppmv/10 yr when N₂O is increased by 50 %. N₂O increases have no significant effect on ozone trends in the Antarctic stratosphere but tend to slow down the ozone recovery in the Arctic stratosphere. Previous studies also found that ozone evolution over Antarctic shows small sensitivity to changes in GHGs (Eyring et al., 2010a). Similar features can be noted in the TCO time series shown in Fig. 3d–f. Figure 3 also indicates that the higher N₂O corresponds to a less

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becomes much larger when CO₂ is also increased by 50 % together with a 50 % N₂O increase (Fig. 5c). Also shown in Fig. 5 are the water vapor changes between the WACCM runs. Stratospheric water vapor depends on tropopause temperatures and production from CH₄ oxidation. As temperature changes in the upper troposphere and lower stratosphere (UTLS) region (Fig. 5a–c), a general decrease of water vapor on the order of 0.05 ppmv is simulated in the upper stratosphere and a significant increase of 0.15 ppmv can be noted in the mid-latitude lower stratosphere in the SH if N₂O is increased by 50 %. When N₂O is increased by 100 %, water vapor increases in the atmosphere due to warming of the tropopause, as is evident in Fig. 5b. Due to the significant cooling of the stratosphere caused by CO₂ and N₂O increases, the stratospheric water vapor decreases in run E3 (Fig. 5f).

Stratospheric methane and water vapor are closely related through chemical oxidation and dynamical transport processes. Figure 5g, h, j show the CH₄ changes caused by N₂O and CO₂ increases. Throughout the stratosphere, water vapor changes are anti-correlated with methane changes. More CH₄, which is shown as a round 0.05–0.07 ppmv in the middle to upper stratosphere (10 hPa to 1 hPa) and maximises in the tropics, is transported to the middle and upper stratosphere as a result of an enhanced BD circulation in response to N₂O and CO₂ increases. Compared with CH₄ increases in E1, in which N₂O is increased by 50 % (Fig. 5g), the CH₄ increases seem to be smaller in magnitude when N₂O increased by 100 % in run E2 (Fig. 5h). This is partly caused by transport differences in the tropical lower stratosphere between runs E1 and E2, since the BD circulation in E2 is stronger than that in E1. Also, a more significant decrease of CH₄ can be found in E2 in the lower stratosphere compared with in E1. CH₄ is oxidized by reactions involving OH, O(¹D) and Cl radicals (e.g. Röckmann et al., 2004), which are perturbed by the GHG increases. The non-linear responses of CH₄ to N₂O increases indicate that the changes in CH₄ are due to the competing effects of transport and chemistry.

As polar stratospheric clouds (PSCs) are closely related to ozone loss in the polar stratosphere (e.g. Rex et al., 2004; Chipperfield et al., 2005; Austin et al., 2010b), it

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is useful to examine the effect of N₂O increases on the occurrence of PSCs. Figure 6 shows the vertical distribution of the modeled change in the PSC surface area in the Antarctic and Arctic. The modeled PSC area is enlarged by 1–2 × 10⁶ km² in the Arctic between 200 and 20 hPa, when N₂O is increased by 50–100 %. In the Antarctic lower stratosphere, however, N₂O increases cause no significant changes in the modeled PSC area. An exception occurs in the Antarctic mid-stratosphere at about 10 hPa, at the upper limit of PSC occurrence, where additional cooling can cause more PSCs. Figure 6 suggests that the N₂O increases have a different impact on the modeled PSC area in the Arctic and Antarctic stratosphere. In the Arctic stratosphere, temperatures are close to the threshold for widespread PSC formation and the cooling caused by N₂O increases can result in more PSCs. In the Antarctic lower stratosphere, temperatures in winter and spring are essentially always low enough for PSC formation, therefore, a small cooling caused by N₂O increases has no significant impact on PSC formation. This difference in the change in modeled PSC area between the Arctic and Antarctic makes a significant contribution to the hemispheric asymmetry of ozone destruction discussed in Fig. 3. However, previous studies (SPARC-CCMVal, 2010) showed that the potential for chlorine activation (PACI) in the Arctic simulated by WACCM is overall larger than expected, though it performs well for polar ozone depletion processes. This may lead to an overestimate for the ozone depleting effects of N₂O increases in the Arctic. Also noticeable is that compared to run E1, the impact of an additional 50 % N₂O increase (run E2) causes a larger increase in PSC area in the Arctic than an additional 50 % CO₂ increase. Here the increased NO_y from N₂O promotes the formation of nitric acid trihydrate.

4 Contributions to ozone depletion from different factors due to N₂O changes

The attribution of ozone variations to dynamical, radiative, and chemical effects resulting from changes in GHGs and ODSs is an important problem in stratospheric climate. Multiple linear regression (MLR) is typically used to estimate statistical model param-

ters (e.g. WMO, 2007; Shepherd, 2008; Stolarski et al., 2010; Oman et al., 2010b). For a given location and time, an ozone change (ΔO_3) can be expressed as follows:

$$\Delta O_3(t) = \sum_j c_j \Delta X_j(t) + \varepsilon(t) \quad (1)$$

where the variables X_j are the factors that can affect ozone, the coefficients c_j are the sensitivity of ozone to the factor X_j , and ε is the error in the fit. Here, ΔO_3 and ΔX_j are the changes in ozone and other quantities between different model experiments with respect to the control experiment (E0) (i.e., $\Delta O_3 = O_{3,E1} - O_{3,E0}$, where $O_{3,E1}$ and $O_{3,E0}$ represent modeled ozone data from runs E0 and E1, respectively).

We select NO_x , ClO_x , as well as temperature (T) as proxies to represent the direct chemical effect, indirect chemical effect and radiative effect result from N_2O increases, respectively. Reactive hydrogen (HO_x) and transport parameters are not included, since their effects are relatively small (not shown). The MLR analysis described above is performed on the modeled data from the control run E0 and run E1, in which N_2O is increased by 50 % by 2050. Ozone changes (ΔO_3) caused by the selected regression factors are examined over different latitude bands (i.e., $90^\circ N-70^\circ N$, $22^\circ N-22^\circ S$, and $70^\circ S-90^\circ S$) on all the model levels from surface to 1 hPa, and the results are shown in Fig. 7.

The contributions of different factors to ozone changes are strongly latitude and altitude dependent. In the Arctic, NO_x and temperature changes cause ozone decreases in the lower and middle stratosphere. NO_x changes, which represent the direct chemical effect of N_2O increases, contribute over 80 % or even higher to the 0.5 ppmv ozone decrease from 70 to 10 hPa (Fig. 7a). Figure 7a also shows that in the upper stratosphere, ClO_x changes result in more ozone. In the tropics (Fig. 7b) both temperature and ClO_x changes tend to result in more ozone in the mid-upper stratosphere. NO_x changes lead to large ozone depletion (about 150 % of total ozone changes) which are partly offset (about 50 %) by the T and ClO_x contributions. NO_x changes in the lower stratosphere cause more ozone on the order of 0.2 ppmv, while ClO_x and T have no

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significant effects on ozone there. The contribution of NO_x to ozone changes, which peaks at 10–3 hPa, is consistent with earlier studies (Oman et al., 2010b; Eyring et al., 2010b). In the Antarctic (Fig. 7c) the ozone changes caused by NO_x changes are still negative but relatively small compared to those in the tropical and Arctic stratosphere.

In contrast to those in the Arctic stratosphere, ozone changes associated with ClO_x are negative in the Antarctic stratosphere. Note also that temperature has a more significant contribution on ozone changes than NO_x in the Antarctic stratosphere. Similar features can be obtained when MLR analysis is performed on the modeled data from control run E0 and run E2 in which N₂O is increased by 100 % by 2050 (not shown).

This multiple regression analysis suggests that the direct chemical effect (indicated by NO_x) of N₂O on ozone changes contributes to the largest part of ozone changes in the lower stratosphere, while the indirect chemical effect (indicated by ClO_x) of N₂O, which is associated with enhanced chlorine-catalyzed ozone destruction, is more important in the upper stratosphere. The effect of temperature changes (indicated as *T*) is more pronounced in the tropical upper stratosphere but relatively less significant in the polar stratosphere. It should be pointed out that some non-linear or interdependent effects of N₂O increase on ozone, such as ozone depletion associated with PSCs, may not be well separated by the MLR analysis. Nevertheless, it provides us with some useful information on the contributions of different processes associated with N₂O increases to ozone changes.

We now discuss the results of the WACCM sensitivity runs which applied N₂O increases separately to the model's radiation and chemistry schemes. The changes in ozone and temperature caused by chemical and radiative effects of N₂O increases are shown in Fig. 8. Ozone decreases up to 10 % (1 ppmv) (Fig. 8a) when the 50 % N₂O increase is applied to the chemistry routine, while the same increase of N₂O in the radiation scheme causes no more than 1 % (0.1 ppmv) ozone decrease (Fig. 8b). When N₂O is increased in both the chemistry and radiation schemes, the ozone changes show no significant differences from those caused by the chemical effect of a 50 % N₂O alone (not shown). It is clear that the chemical effect contributes most to the ozone changes

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caused by N₂O increase, which is consistent with the MLR analysis. The ozone depletion causes significant subsequent stratospheric cooling, with a maximum of -1.7 K in the middle stratosphere (around 10 hPa) in the tropics (Fig. 8c). The direct radiative effect of N₂O is much smaller (below 1 K) than the indirect radiative effects (Fig. 8c and d) without a clear pattern in the spatial distribution.

Another factor which should also be considered is the dynamical effect of increasing N₂O. As shown in Table 3, the increase in N₂O is likely to strengthen the BD circulation. Meanwhile, this strengthened circulation influences the distribution and also contributes to the variability of O₃. Figure 9 shows the vertical ozone flux in the reference experiment E0 (Fig. 9a) as well as the corresponding changes in E1 relative to that in control run E0 (Fig. 9b). For a given grid cell $A[\text{lat}_i, \text{lev}_k]$ located at the latitude of lat_i and the pressure levels of lev_k , the vertical ozone flux is defined as the incoming ozone at the lower level lev_{k-1} ($O_3[\text{lat}_i, \text{lev}_{k-1}] \times w^*[\text{lat}_i, \text{lev}_{k-1}]$) minus the ozone coming out at higher level lev_{k+1} ($O_3[\text{lat}_i, \text{lev}_{k+1}] \times w^*[\text{lat}_i, \text{lev}_{k+1}]$). Consistent with the overall picture of the BD circulation, the ozone in the tropical lower stratosphere is transported to the upper stratosphere and ozone in the mid-latitude upper stratosphere is transported downward to the lower stratosphere (Fig. 9a). It is evident from Fig. 9b that the strengthened BD circulation caused by the increased N₂O leads to a net decrease of ozone in the tropical lower stratosphere and a net ozone increase at around 100 hPa at southern mid-to-high latitudes. The large ozone increases around 100 hPa at southern mid-latitudes exhibited in Fig. 1a–c may be mainly caused by this transport effect. The MLR analysis with w^* as an additional regression factor suggests that the dynamic effect of a N₂O increase on ozone changes is much less significant than its chemical and radiative effects in most of the areas (not shown). The result here implies that the dynamical effect of increasing N₂O on ozone is small on average, but may have a significant contribution to local ozone changes in the mid-latitude stratosphere.

5 Discussion and conclusions

The distribution of ozone changes resulting from N_2O increases in a 3-D CCM shown as function of latitude and altitude and the chemical mechanism was discussed by examining various chemical species related to ozone destruction. Subsequent climate changes induced by these N_2O increases, such as changes in stratospheric temperatures and water vapor, BD circulation and area of PSCs, were also examined, helping us to understand the N_2O and ozone interactions with climate changes. A series of sensitivity simulations and MLR were performed to detect the dominant factors controlling the ozone changes caused by N_2O increases.

The CCM results suggest that N_2O increases by 50%/100% between 2001 and 2050 result in more ozone destruction, reducing in ozone mixing ratios of maximally 6%/10% in the middle stratosphere at around 10 hPa. However, the total ozone column still shows an increase in future decades. The increase of N_2O also results in significant temperature and H_2O changes, which are seen in a slight cooling in the stratosphere and an evident increase of stratospheric H_2O . In a future cooler stratosphere the net yield of NO_y is known to decrease, and our simulations indicate that the ozone-depleting impact of N_2O becomes smaller due to the cooling and fewer halogens in the stratosphere. The ozone depletion potential for N_2O in a future climate depends on whether the stratospheric cooling effect is larger than the effect of tropospheric N_2O changes. The increase of N_2O has only a small influence on chemical processes associated with Antarctic ozone destruction. However, increased emissions of N_2O are likely to delay recovery of the ozone in the Arctic stratosphere.

The depletion of ozone caused by nitrous oxide involves complex dynamical, chemical and radiative processes. The analyses based on a series of sensitivity simulations and MLR suggests that N_2O increases cause ozone changes mainly through chemical processes. The direct chemical effect of N_2O increases (via NO_x chemistry) contributes the majority of ozone changes in the lower and middle stratosphere, while the indirect chemical effect of N_2O increases (via halogen chemistry) is simulated as the dominant

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Table 1. The configurations of four CCM transient simulations.

Experiments	GHGs Scenario
E0	IPCC A1B scenario
E1	Linear 50 % increase in N ₂ O surface vmr from 2001–2050
E2	As E1 but with 100 % increase in N ₂ O 2001–2050
E3	As E1 with additional 50 % linear increase in CO ₂ vmr from 2001–2050

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Table 2. The configurations of four CCM sensitive simulations.

Simulations	GHGs Settings
S0	Fix N ₂ O as 2050 climatology in IPCC A1B scenario
S1	Increase N ₂ O by 50 % on S0 only in chemistry
S2	Increase N ₂ O by 50 % on S0 only in radiation
S3	Increase N ₂ O by 50 % on S0 interactively

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Table 3. The tropical (25° N–25° S) upwelling (w^*) averaged between 70–20 hPa over the last 10 yr of the four simulations and changes in w^* (Δw^*) relative to the first 10 yr of the corresponding simulations.

Experiments	$w^*(10^{-3} \text{ m s}^{-1})$	$\Delta w^*(\%)$
E0	0.029	5.5
E1	0.032	28.4
E2	0.037	37.3
E3	0.033	21.9

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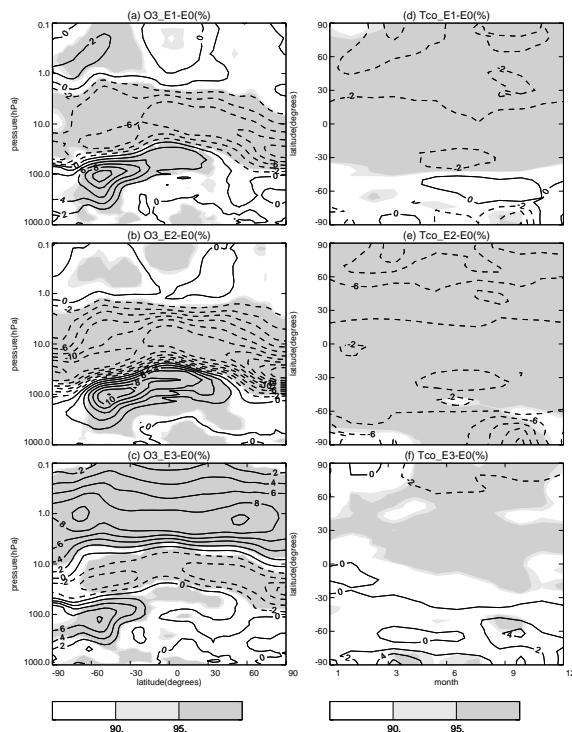


Fig. 1. Latitude-pressure cross sections of differences (%) in zonal mean ozone mixing ratios (2041–2050) between WACCM experiments **(a)** E1 and E0, **(b)** E2 and E0, and **(c)** E3 and E0. Time-latitude cross sections of differences (%) in zonal mean total column ozone climatology (2041–2050) between experiments **(d)** E1 and E0, **(e)** E2 and E0, **(f)** E3 and E0. The contour interval is 2% for percentage ozone changes. The grey colors represent significance levels for the mean state differences between different experiments by the Student's *T* test.

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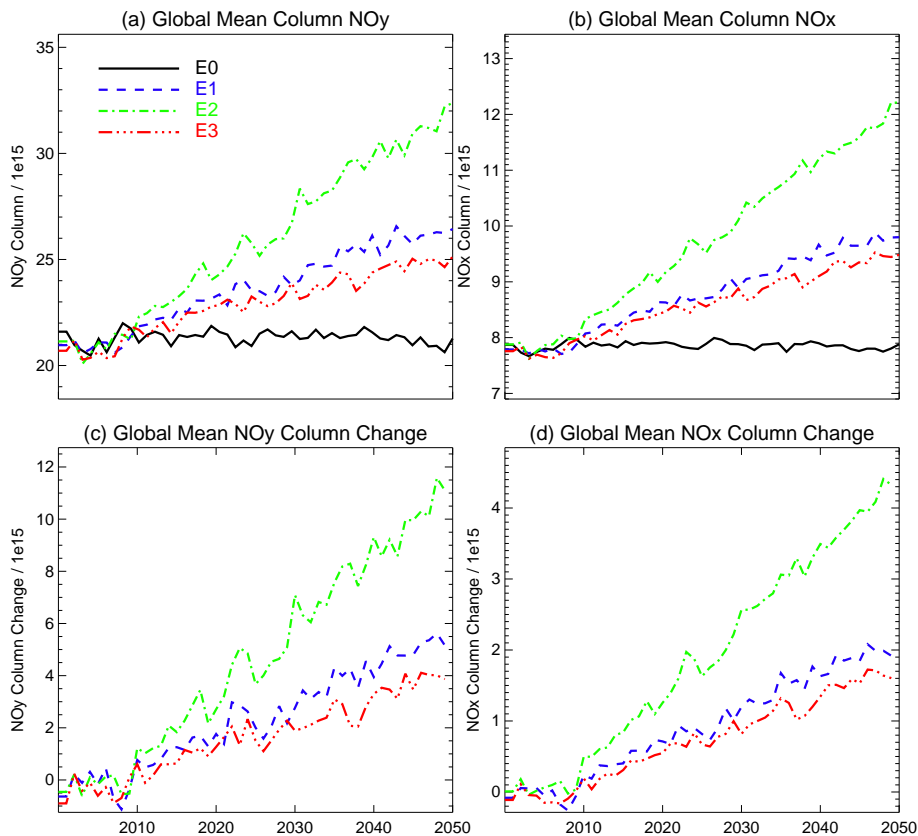


Fig. 2. Time series of global mean **(a)** column NO_y (molecules cm⁻²) and **(b)** column NO_x (molecules cm⁻²) in E0 (black line), E1 (blue line), E2 (green line) and E3 (red line). Difference in **(c)** column NO_y and **(d)** column NO_x between E1 and E0 (blue line), E2 and E0 (green line), E3 and E0 (red line).

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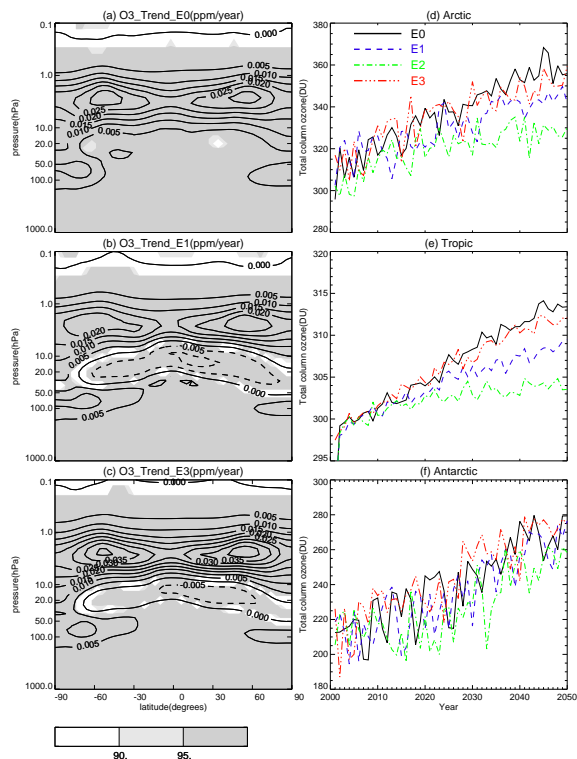


Fig. 3. (a, b, c) Latitude-pressure cross sections of the long-term (2001–2050) trends of zonal mean ozone mixing ratios (ppmvyr^{-1}) in WACCM experiments E0, E1 and E3. The contour interval is $0.005 \text{ ppmvyr}^{-1}$. Grey colors represent statistic significance levels of the trends. (d, e, f) Time series of total column ozone (DU) in different regions (Arctic, Tropics, Antarctic) in experiments E0 (black line), E1 (blue dashed line), E2 (green dashed-dotted line) and E3 (red dashed-dotted line).

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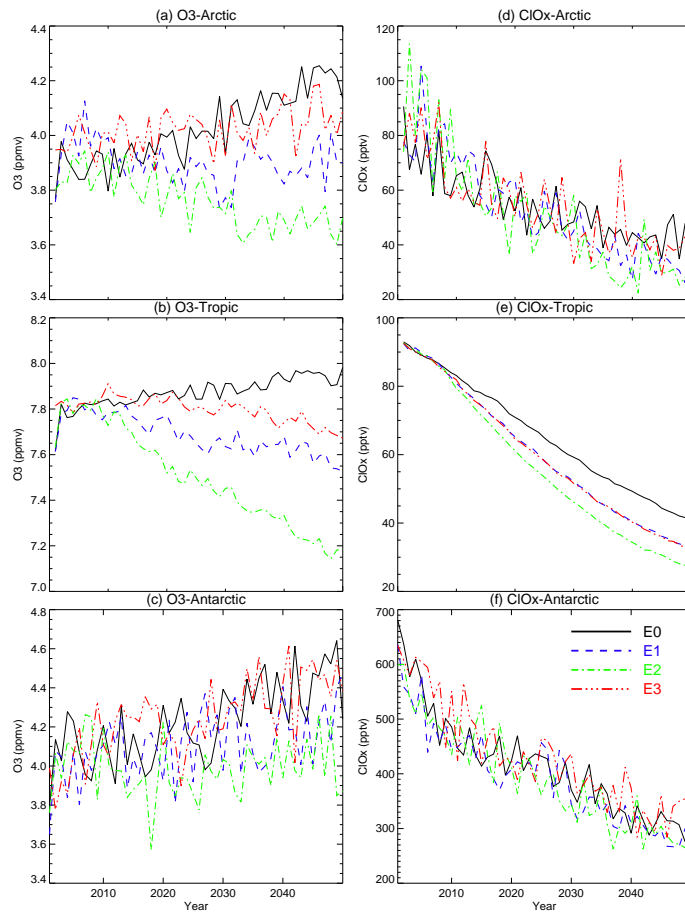


Fig. 4. Time series of mean **(a, b, c)** ozone and **(d, e, f)** Cl_x ($Cl_x = Cl + ClO + 2 Cl_2O_2$) between 20–10 hPa averaged over **(a)** Arctic (60° N–90° N), **(b)** Tropics (22° N–22° S) and **(c)** Antarctic (60° S–90° S) in E0 (black line), E1 (blue line), E2 (green line) and E3 (red line).

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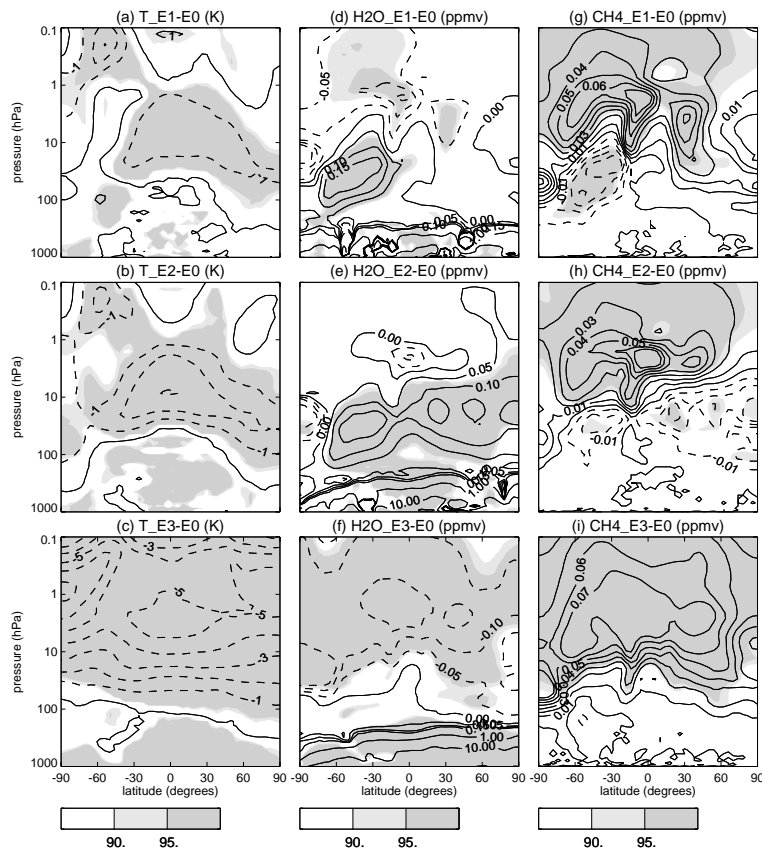


Fig. 5. Latitude-pressure cross sections of differences in **(a, b, c)** zonal mean temperature, **(d, e, f)** H₂O and **(g, h, i)** CH₄ climatology (2041–2050) between WACCM experiments. The contour intervals are 1 K for temperature, 0.01 ppmv for CH₄ and 0.05 ppmv for H₂O in the stratosphere where H₂O mixing ratios low. Grey colors represent statistical significance levels for the mean state differences between different experiments by the Student's *T* test.

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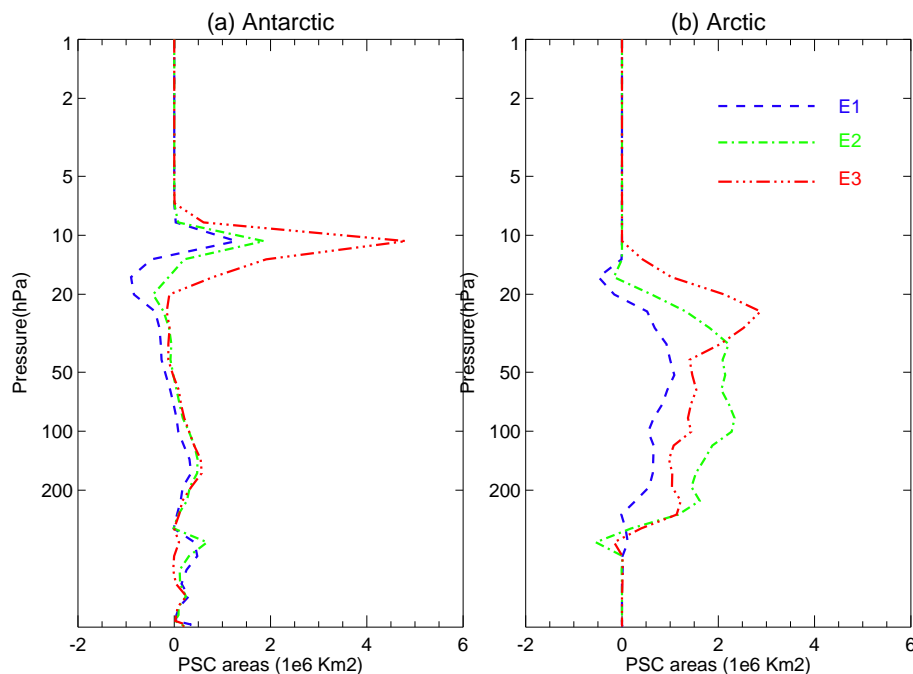


Fig. 6. Profiles of differences in annual mean PSCs area ($\times 10^6 \text{ km}^2$) between WACCM experiments E1 and E0 (blue dashed line), E2 and E0 (green dashed dotted line), and E3 and E0 (red dashed dotted line) in **(a)** Antarctic and **(b)** Arctic.

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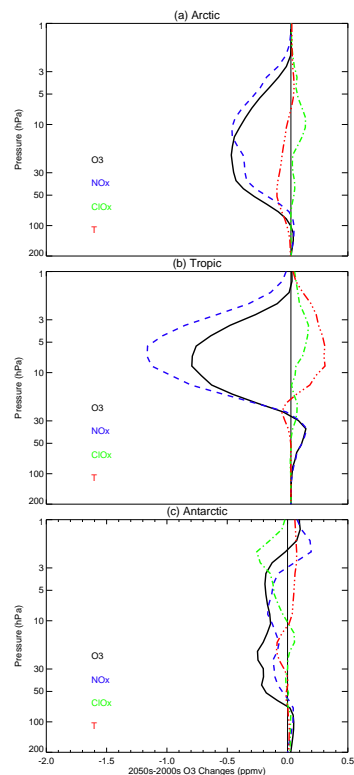


Fig. 7. Results of multiple regressions for spatially-averaged ozone change ($\Delta O_3 = O_3(E1) - O_3(E0)$) with spatially-averaged NO_x , ClO_x and T change. The averaging domains are $90^\circ N - 70^\circ N$, $22^\circ N - 22^\circ S$, and $70^\circ S - 90^\circ S$. The black thick line shows the changes of ΔO_3 from 2001 to 2050 ($d\Delta O_3$), calculated by a simple linear regression of ΔO_3 with year ($\Delta O_3 = a_0 + a_1 \times \text{year}$, $d\Delta O_3 = a_1 \times 50$). The blue dashed line, green dashed-dotted line and red dashed double point line show the NO_x , EESC and T contributions to ΔO_3 (i.e. $d\Delta O_{3,NO_x} = \Delta NO_x \times \text{Coefs}_{NO_x}$), respectively.

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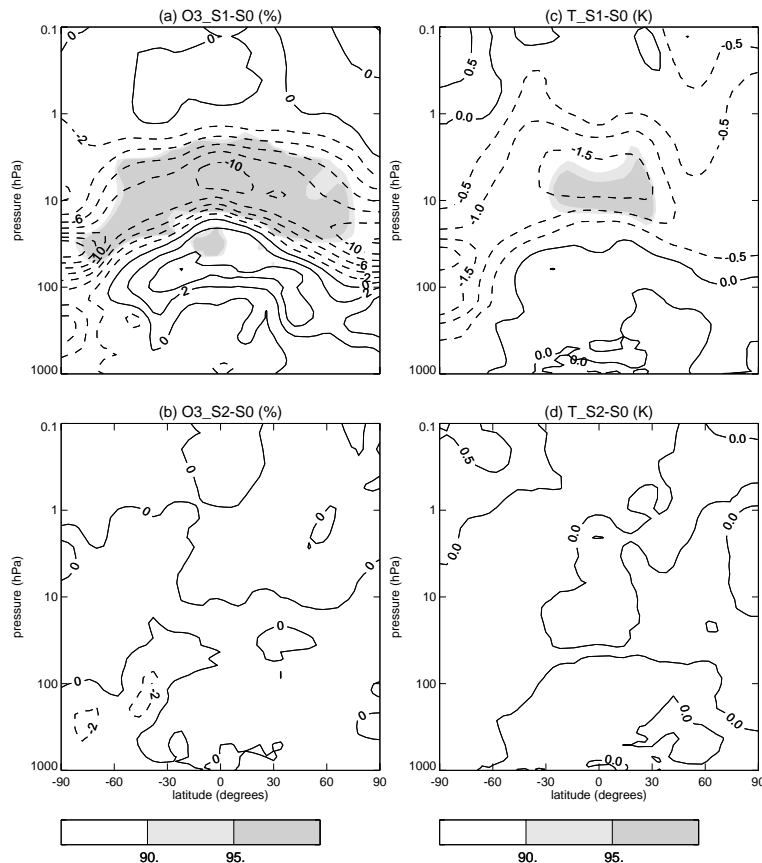


Fig. 8. Latitude-pressure cross sections of zonal mean ozone differences (%) between WACCM sensitivity experiments **(a)** S1 and S0, and **(b)** S2 and S0, and temperature differences (K) between sensitivity experiments **(c)** S1 and S0, and **(d)** S2 and S0. The contour interval for ozone is 2% and for temperature is 0.5 K. Grey colors represent statistical significance levels for the mean state differences between different simulations by the Student's *T* test.

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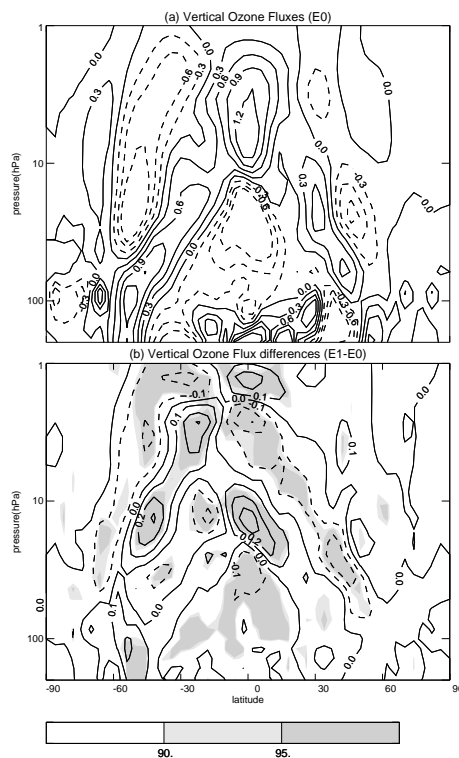


Fig. 9. Vertical ozone fluxes associated with the BD circulation (w^*) calculated from **(a)** reference experiment (E0) and **(b)** the differences between E1 and E0 (for more details of the definition of ozone flux see text). The contour intervals are $0.3 \times 10^3 \text{ kg s}^{-1}$ and $0.1 \times 10^3 \text{ kg s}^{-1}$ in **(a)** and **(b)**, respectively. Grey colors **(b)** represent the statistical significance levels for the mean state differences between different experiments by the Student's T test.

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