

Interactive comment on “Stratospheric ozone depletion from future nitrous oxide increases” by W. Wang et al.

Anonymous Referee #3

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The goal of this work is to quantify the radiative and chemical effects of N₂O increases separately.

Weakest part of the paper has to do with the abstract/introduction discussion of ODP. There are two reasons for this. First – the ODP is not defined in this work, and as I see it the definition most people use is not being followed. I realize that ODP has a bit of a slippery definition, but it is pretty clear from the early work that the ODP itself addresses perturbations in various chlorofluorocarbons relative to F11, without explicitly stating the presumption that errors in the 1D model of the era would cancel out since the area of interest was the upper stratosphere and chlorine would not care where it came from. The big uncertainty for HCFCs had to do with how much would be destroyed in the troposphere. Later work uses the same definition, without regard loss of cancellation of errors when processes involved were different. In ALL cases, the ODP has been used for a single base atmosphere – thus you might calculate the ozone change due to a perturbation in N₂O in 2000 and then the same thing for a perturbation in N₂O in 2050, with a cooler stratosphere. Using the classic definition, the ODP would be nearly the same (as shown by Ravishanka’s paper and also by Fleming). Clearly this paper has a strong sense of the other processes that are neglected by this approach, and I think they are important. However, this paper in the introduction uses ODP in a somewhat different matter, implicitly including impacts of cooling on the NO_y/N₂O as part of the ODP. This part of the paper requires substantial revision. Clarification will also lead to some changes in organization which will help the overall structure.

Presentation could be organized much more cleanly – even in the abstract, the per- cents are difficult to understand (increases of N₂O 50%/100% cause ozone reductions, but then TCO still increases but N₂O causes a 2%/6% decreases). This needs careful construction to avoid confusion. Author intent becomes clear when reading the whole paper but the abstract should be understandable on its own.

Thanks for the good comment. The term ‘ODP’ is indeed ambiguous and the abstract needs to be further polished. In the revised paper, we avoid using the concept of ODP in the abstract. The ‘classic’ definition of ODP is introduced in the Introduction section, and a discussion about the dependence of ODP on N₂O and CO₂-induced cooling is given at the end of section 4.

SPECIFIC COMMENTS

Abstract: As discussed above, the ozone depletion potential for N₂O in a future climate does not depend on temperature or tropospheric N₂O level using the ‘classic’ definition of ODP, even though as stated earlier in the abstract the NO_y per N₂O yield is a function of temperature. In my opinion this points up the inadequacy of the classic ODP to address this problem, but at the same time the authors need to take care to use terms like ODP in the same way that they are used by

others in the community, most importantly Ravishankara et al. (2009), Portman et al. (2012) and Fleming et al. (2011).

See the reply above.

Last sentence in the abstract – it is very surprising that the dynamical effects of N₂O increases can be large locally anywhere.

We will remove the last sentence in the abstract, since it might be model dependent. The revised abstract is rewritten as:

We have investigated the impact of assumed nitrous oxide (N₂O) increases on strato- spheric chemistry and dynamics by a series of idealized simulations with a coupled chemistry-climate model (CCM). In a future cooler stratosphere the net yield of NO_y from a changed N₂O is known to decrease, but NO_y can still be significantly increased by the increase of N₂O. **In the middle stratosphere at around 10 hPa**, increases in N₂O of 50% between 2001 and 2050 result in more ozone destruction, causing a reduction in ozone mixing ratios of maximally 6%. This enhanced destruction could cause an ozone decline in the second half of this century in the middle stratosphere. The increase of 50% in N₂O cause a 2% decrease in the total ozone column (TCO) compared with the reference simulation. **However, for the long-term trend, TCO still shows an increase in future decades.** N₂O 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 **effect of N₂O** increases on ozone in a future climate depends both on stratospheric temperature changes and tropospheric N₂O changes, which have reversed impact on ozone in the middle and upper stratosphere. A 50% CO₂ increase in conjunction with a 50% N₂O increase relative to the current CO₂ and N₂O condition 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 found that the chemical effect of N₂O increases dominates the ozone changes in the stratosphere while the dynamical and radiative effects of N₂O increases are insignificant on average.

p. 29453 L 20 “Even though the yield of NO_x from N₂O decreases in a cooler stratosphere, the efficiency of ozone loss due to NO_y increases so that N₂O increases in run E3 still causes an increase in ozone loss due to the increasing NO_y in an atmosphere with decreasing halogens.” This point needs explanation.

We will remove this sentence, since it is ambiguous.

p. 29453 L 24 – inconsistent use of ODP compared with previous authors. This needs discussion earlier in the paper.

The term ‘OPD’ is dropped out to avoid confusion.

29454 L 20 Specifics of the changes in vertical transport are not completely robust across all 3D models (CCMVal report) – sign change in the tropics is the same, but the rate of change differs and the latitude dependence of downward transport increase in the extratropics differs. This is important for the context of this discussion.

This is a good point. The following discussion is added in the revised text:

‘However, there are still relatively large uncertainties for the vertical transport in 3D CCMs (i.e. large spread between different CCMs for the latitude dependence of downward transport, SPARC CCMVal report 2010), which indicate that the dynamical effects on ozone still need further investigation.’

29455 L 15 ff - The discussion of the lack of increase in tropical w^* (causing an ozone decrease) is weak. ‘perhaps it didn’t change in our simulation?’ - that is a big difference compared with WACCM results in CCMVal and almost certainly happens because of some other change compared with prior simulations. This is a hot topic and must be addressed. The mention of SSTs later (p 29457) is not really sufficient – why use something different, and why is there little trend in the SSTs? If it is due to using SSTs without trend, then you need to reference the papers that talk about the influence of SST trend on w^* .

The CCMs in CCMVal used two different sets of the ocean surface forcing in REF-B2 simulations. CAM3.5, GEOSCCM, ULAQ, and WACCM all use CCSM3 SST data, while E39CA, UMSLIMCAT, and the UMUKCA models use HadGEM1 SST data (Morgenstern et al., 2010, Table 6). We used the WACCM model, but chose the HadGEM1 data. A comparison between these two SSTs data sets shows that the long-term increase of SSTs in future century from CCSM3 data is much stronger than the HadGEM1 data. It is a good suggestion that we should point out the influence of SST trend in w^* . We discuss the importance of SSTs in modulating the tropical upwelling by citing previous work by Xie et al., 2008, in P29457 L9. The similar result can be found in a recent work by Oberländer et al., 2013. Their work shows that the warming in SSTs contributes almost the entire strengthening of B-D circulation in the lower stratosphere, and is also an important factor influencing the B-D circulation in the middle and upper stratosphere.

29455 L 25 – slow down ozone recovery – this is jargon! - and imprecise at that. Some segment of the community thinks of ‘recovery’ simply as ‘ozone increase to prior levels’ but another segment thinks of ‘recovery’ as ‘no more loss due to anthropogenic chlorine’. N_2O can affect one definition of ‘recovery’ but not the other.

Good point! In the revised text, ‘ozone recovery’ is changed to ‘ozone increase’.

29456 L 20 PSCs jump out of now here. If this is an important part of the feedback, many aspects of PSC formation and parameterization need to be discussed.

Thanks for the suggestion. The discussions related to PSC are dropped out here.

29458 L 10 why does cooling cause the water vapor decrease in the stratosphere (away from the troposphere).

According to the suggestion by reviewer 2, Figure 5d-5i and the related discussions are deleted in the revised text. See details in the response to the comments from reviewer 2.

29459 L 3 To support confidence in the projected change in PSC area – how do simulations for present compare with the PSC climatology developed from CALIPSO? Is there also an increase in HNO_3 that would affect PSC formation (and not just cooling associated with N_2O ? If CO_2 is increasing, isn’t the effect of N_2O cooling in the noise? You do get to the NO_y effect as the last sentence of the paragraph – confusing presentation.

We did not do such kind of comparison as there have already been some work (i.e. Tilmes et al., 2007, SPARC CCMVal report 2010) showing that the simulated potential of activated chlorine (PACI, a measure similar to PSC volume) for Antarctic is in good agreement with observations. Those previous work also showed that the temperatures are biased high and the vortex volume and are significantly smaller than observed in the Arctic stratosphere. We has also discussed

about this uncertainty in P29459 L16-L20.

There's also an increase in HNO₃ in both Antarctic and Arctic when N₂O is increased. In the revised text, the factors affecting PSC area are discussed with caution with an emphasis both on the polar temperature and HNO_x/NO_y changes.

29460 L4 ff MLR is a wonderful tool for a lot of things, but it does not separate mechanisms unless they are sufficiently orthogonal – discussed by Oman et al. (2013) concerning QBO and ENSO during the Aura period and also by Stolarski et al. (2010) talk about needing a long enough time record for signals to have sufficient orthogonal-ity to separate. How do you think your proxies behave? You need to at least talk about this, and how the coefficients are less certain when the proxies are less orthogonal.

This is a good point. The proxies used in our MLR are not strictly orthogonal as they are interdependent to some extent, particularly in chemistry processes. We did not chose the ENSO and QBO as the proxies in our MLR although they are important factors impacting on total column ozone. Our main focus is not to predict ozone using MLR but to find the relative importance of the proxies we interested. We add some discussions about this issue in the paragraph P29461 L16-L20:

'It should be pointed out that large uncertainties may exist in our MLR analysis. This is because of the interdependent of different proxies. Actually, the time series of NO_x and ClO_x are strongly correlated to each other. Nevertheless, it provides us with some quantitative information on the relative importance of different processes associated with N₂O increases in causing ozone changes.'

29461 l17 – 'may not be separated' – you have a foundation to say something about this if you have taken the steps of discussing what you need to have a definitive answer from MLR.

Thanks for the point. The sentence is rephrased in the revised text.

29462 L 7 Curious that increasing N₂O increases the BDC whereas in prior discussion the change in upwelling was smaller than expected due to lack of trend in the SSTs. This bears discussion.

The revised paper will includes the following discussion:

In prior discussions, what we emphasis is the long term trends of the DB circulation in the simulations are small due to lack of trend in SSTs. However, N₂O increases can enhance the BD circulation due to its direct (radiative) and indirect (ozone feedbacks) cooling effect in the stratosphere.

Table 1 – not clear that the N₂O increases are percents relative to A1b (although that is clear in the discussion/examples). I think a plot would be much more clear than the table.

The table is explained in more details in the revised paper.

Grammar

29455 L 14 'simulated in simulations' There are other grammatical issues but because suggested revisions are substantial I do not take the time to identify more of them.

Change to be: pointed out in previous simulations.

We have carefully checked English in the revised paper to avoid those grammatical errors.

Additional discussion about the concept of ODP Add to the end of Section 4:

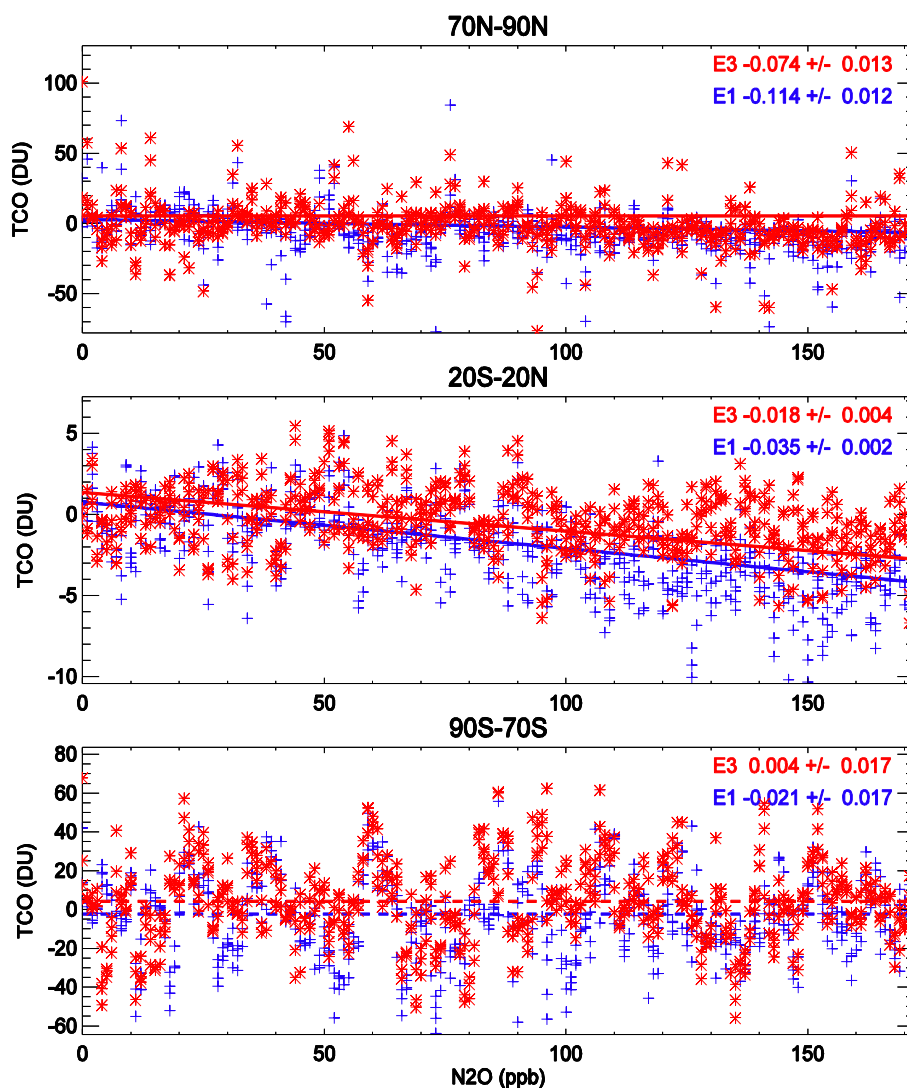


Fig. 10. TCO changes relative to that in control run E0 averaged over (up) Arctic, (middle) Tropics and (bottom) Antarctic caused by surface N_2O changes with different magnitudes. The blue + and the red * are from the runs of E1 and E3, respectively. The lines are fitted by a simple regression model, the solid and dashed lines represent the regression is over and below 95% confidence level, respectively.

At this stage, it is worthwhile to discuss the ODP of N_2O . Based on 'classic' definition the ODP (Ravishanka et al., 2009), N_2O ODP is temperature independent and will keep unchanged even though the chemical and dynamical environment changes. The ODP depends on the base state of the atmosphere and makes the assumption that the other source gases do not change during the time integration. However, it is impossible to keep the temperature and other sources of gases the same in the real atmosphere since there existing complex interactions between different reactions cycles. Meanwhile, some reaction rates and reaction times are affected by temperatures and dynamical transport. This 'classic' definition of ODP is challenged by other researches (Portmann et al. 2012; Revell et al. 2012a; 2012b).

Figure 10 shows TCO changes due to N₂O and CO₂ increases. The total ozone depletion ratios in run E1 (define as the slope of linear fit of the TCO changes vs surface N₂O changes) are -0.114, -0.035 and -0.021 DU ppb⁻¹ over the Arctic, tropics and Antarctic, respectively. The values for the Arctic and the tropics are in good agreement with the result in Revell et al. (2012b), but the value for the Antarctic is much smaller. The goodness of the linear regressions in Fig. 10 is above 95% significance level in the Arctic and tropics, suggesting that the ozone depletion ratio for N₂O is basically linear. When CO₂ and N₂O are increased together in the same increasing rate, the ozone depletions ratios change to -0.074, -0.018 and +0.004 over the Arctic, tropics and Antarctic, respectively. The result here suggests that the CO₂-induced cooling reduces the ozone depletion by 35% over the Arctic, 49% in the tropics and 119% over the Antarctic. The corresponding ozone changes at 10 hPa resulted from N₂O and CO₂ increases shows the similar results (not show), i.e., the CO₂-induced cooling can reduce ozone depletion associated with N₂O by 60%, 33% and 144% at 10 hPa over the Arctic, tropics and Antarctic, respectively. The high dependence of N₂O-induced ozone depletion on stratospheric temperatures suggests that a more reasonable ODP definition needs to be considered.

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