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Comment on acp-2021-149

Susan Solomon (Referee)

Referee comment on "Effects of enhanced downwelling of NO_x on Antarctic upper-stratospheric ozone in the 21st century" by Ville Maliniemi et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-149-RC2, 2021

Review of the paper by Maliniemi et al. by Susan Solomon

{Note: due to errors in the ACP system, this review may appear twice).

This paper uses WACCM CMIP6 simulations of the 21st century to examine the future behavior of ozone in the upper stratosphere using different scenarios.   The primary conclusion is that while a cooler stratosphere leads to ozone increases over much of the upper stratosphere, an exception occurs in the Antarctic, where the authors argue that increased downwelling within the strong polar vortex deposits greater abundances of NOx from EEP, which deplete ozone. The paper makes some interesting points but requires revisions before publication.   My comment are as follows:

- The paper doesn't what happens to the total ozone column, which is a key quantity and is frequently what is meant when people talk about recovery. Because much of the ozone loss is in the lowermost stratosphere (especially in the Antarctic), I would suspect that Antarctic total ozone still undergoes recovery, perhaps even super-recovery.   Please add a figure showing what happens to the global total ozone as well in this model.
- The use of the term 'recovery' or 'super-recovery' in this paper raises the question of recovery from what – it should be recovery from ozone depletion due to CFCs. Figure 1 shows values of average ozone mixing ratios from 1-10 hPa and from 10-100 hPa in different months and regions. These are very broad swaths, too broad to be appropriate. The mixing ratio at 10 hPa is much larger than at 1 hPa, so this quantity is heavily weighted towards lower altitudes and is hard to interpret.   The problem is even worse in Figure 2, where ClOx abundances for 1-100 hPa are presented. I suspect that there was not much ozone depletion from 1960-2000 (if any) at for example 1 hPa, so it's not clear that super-recovery is the right word there…it may not have been "depleted" in the first place. Please provide an additional figure showing contour plots of the changes in ozone and ClOx from -90 to 90 degrees, 1000 to 0.01 hPa, for 1960-2000 and clarify where depletion occurs versus where the ozone changes of

interest here occur. If there are regions or months where little or no ozone depletion occurred from 1960-2000 in the first place, then recovery or super-recovery is the wrong language and should be altered. These plots should probably be in percent units rather than mixing ratio, so that we can see the extent of changes everywhere.

- Please elaborate what is in the model with regard to solar proton events, and whether those could modify the picture. See Stone et al., GRL, 2018.
- Figure 6 shows that the maximum NOx increase occurs in August. It is essentially gone by October. Please explain why this occurs. I would have expected a longer residence time. Is it being mixed out of the vortex? Chemically destroyed? Or?
- There are many mistakes in basic English in the paper. I note that at least one of the authors is a native speaker of the English language and request that attention be paid to proper English to make the paper more readable. "The" is missing in many places, for example, and those mistakes should be fixed by the authors, not the reviewers.
- The title seems unclear, for the reasons noted above. Please rephrase. Something like "Effects of enhanced downwelling of NOx on Antarctic upper stratospheric ozone in the 21st century" might be suitable.
- Line 22 and later (e.g., 106). Garcia and Randel 2008 was a modelling study; Butchart et al. 2014 was a review but stated that while models showed BDC increases, the data was unclear. The statement that the strength of the BDC is increasing should be edited if it is just based on models; if it's based on data then please provide appropriate references to back it up.
- Line 27. Where in WMO, 2018, and does this pertain to ozone in the upper stratosphere?
- Line 28-29. How can you be sure that EEP are the main cause? Please note the existence of other sources of NOx (SPE, but also simply downward transport from other sources besides EEP). Please clarify how it is you know that the NO increase you calculate is indeed due to EEP (versus e.g., photolysis and photoionization reactions at higher altitudes, transport from higher altitudes but originating from lower, sub-auroral latitudes, etc.); also are you referring to auroral electrons or others? There is a lot of NO at 110 km even in the tropics, so we need to be sure we understand the role of EEP versus e.g. transport of that. Also, is there a solar cycle in the signal?
- Line 55. Briefly describe what was done in the CMIP6 recommendations on solar activity that are relevant here.
- Line 63. Why 31 years?
- Figure 1. I don't understand why these figures are so spikey if they are treated with the 31 year mean/LOWESS approach as described. Please explain.
- Figure 2. Can you explain why the variability is greater in the future than the past?
- Line 97. The increased ozone throughout the upper stratosphere is likely caused mainly by colder temperatures, not just at the equator.
- Lines 103-104. I don't think this statement is quite right. While it seems likely that HOx production is important in the mesosphere, a key point is that mesospheric ozone loss chemistry is much less temperature sensitive than that at the stratospause – different reactions with different energies of activation are involved. Please rephrase.
- Lines 130-135. Here the authors raise some doubt about how much of the changes referred to are indeed due to EEP and NOx versus other causes (e.g., dynamical). This is quite concerning since it's the main conclusion of the paper. Can dynamical contributions be checked by looking at other quantities in the model? for example, you might use SF6 as a tracer for downward mesospheric transport and see how percent changes in SF6 compare to the percent changes in ozone, for example. A clearer analysis is needed to support the paper's conclusions.
- Presumably if the NOx changes are caused by increased downwelling, then they should increase with time through the time of the simulation. Please show that.
- Line 140. Earlier, you said the equatorial lower stratospheric changes were dynamical.