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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Explicit calculation of indirect global warming potentials for halons using atmospheric models" *by* D. Youn et al.

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Anonymous Referee 2

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General comments:
This manuscript is the first to use a CTM to estimate the indirect GWPs of any of the halocarbons due to the stratospheric ozone they destroy. The authors have made a good choice to look at the two halons with the greatest atmospheric concentration, as these compounds likely have the most important indirect effects. Indirect GWPs due

to the ozone destruction associated with halocarbons have been calculated in the past with a simple parameterization based on equivalent effective stratospheric chlorine. The value for the 100-year indirect GWP of Halon 1301 agrees rather well with the 2007 WMO estimate, which uses the simple parameterization, but the value here is somewhat smaller in magnitude. The indirect value for Halon 1211 is also of smaller magnitude than the WMO value. Nevertheless, both values fall within the single standard deviation quoted in WMO (2007) and so can be interpreted as confirmation of the simple approach. The authors should be commended for embarking on this study. There are many simplifying assumptions in the previous approach based on EESC and so it is very useful to have a more thorough calculation with which to compare. Nevertheless, there are a few issues of substance that I believe should be addressed.
→ We thank the reviewer for the invaluable and constructive comments.

First, the authors make a point that their calculated value for the 100-year indirect GWP of Halon 1211 is smaller than the values quoted before using the EESC parameterization. While their value is smaller than the previously-quoted central values, it falls within a single standard deviation of the WMO (2007) value and so is most likely not different. The tone of the manuscript would seem to require change to reflect this.
→ Agreed. We changed the corresponding sentences to reflect the reviewer's comment.

Second, the majority of the uncertainty associated with the WMO (2007) value is likely due to the assumed ozone radiative forcing from halocarbons and the value of alpha. Without knowing how these values from the models used in the manuscript compare to what was assumed in the WMO assessment, it is impossible for the reader to determine why the values might be different. Specifically, with the high degree of uncertainty in the ozone radiative forcing (see, e.g., IPCC (2007)), it would be helpful

if the authors could provide more justification for why the reader should believe the values from these particular models. The error bars quoted in the WMO assessment are quite large, so if the authors were able to reduce the uncertainty in some way, that could be an improvement over our current understanding. This would not be easy, however. In the absence of this, the paper is still useful and should be published. But, it would need to be primarily a confirmation of our current understanding rather than presented as providing a new, different value.
→ The uncertainty range in stratospheric ozone radiative forcing in IPCC (2007) is based on 1) observationally based ozone RF change which is dependent on considered time period and 2) model assessed results between pre-industrial and 2005. Furthermore, the estimate takes into account causes of stratospheric ozone change in addition to those due to the Montreal Protocol gases (IPCC, 2007). In this study, we calculate year-by-year temporal changes in radiative forcing following model-determined changes in perturbed ozone distribution using a radiative transfer model, which means our approach does not require assumed RF from halocarbons and the value of alpha. However, both the 2-D and 3-D models produce alpha values that are close to 60 (WMO, 2007). The ozone RF depends on the change in ozone distribution and we have calculated the ozone RF due to changes in chlorine and bromine from 1979 and 1997 using our 2-D CTM and RTM which is -0.11 W m-2 smaller than the value of -0.15$\pm$0.10 W m-2 used in the previous analyses (WMO, 2007). The ozone RF could possibly explain the smaller Indirect GWP derived here since alpha values are similar. In section 4.3, we added sentences as below:
Both the 2-D and 3-D models produce alpha values that are close to 60 (WMO, 2007). The ozone RF depends on the change in ozone distribution and we have calculated the ozone RF due to changes in chlorine and bromine from 1979 and 1997 using our 2-D CTM and RTM which is -0.11 W m-2 smaller than the value of -0.15$\pm$0.10 W m-2 used in the previous analyses (WMO, 2007). The ozone RF could possibly explain the smaller Indirect GWP derived here since alpha values are similar.

Another question raised by the results here involves the implication of the smaller mean value of the halons when compared to the central values from WMO (2007). If the total ozone forcing due to halocarbons were the same in the models used here as assumed in WMO, this might imply that the indirect forcing of the other gases should be more significant. Or, on the other hand, the total forcing of the ozone lost due to halocarbons may simply be somewhat smaller in magnitude in these models. If the authors were able to address this issue, it would be informative. The models used here underestimate the lifetime of Halon 1211 compared to WMO (2007). How much does this account for the indirect GWP differences? It would seem to go in the right direction, but not fully account for the difference.
→ Our current results will not be much affected by the current smaller mean values of Halons. Figures 1 and 2 are presented to prove it with another Halon-1211 experiment in which surface fluxes are increased to see the sensitivity to Halon-1211 tropospheric concentrations. The increased concentrations in a steady-state atmosphere (Figure 1) make little differences in resulting ozone RFs and indirect GWP (Figure 2). Baseline tropospheric mixing ratios of Halons (starting concentrations of halons) thus would little affect our indirect GWPs.
A shorter lifetime should somewhat reduce the derived indirect GWPs but the differences between the 1211 lifetime from WMO (2007) and from the model is within the uncertainty of the WMO value. As indicated by the longer lifetime we derive for Halon-1301, the large decrease in the indirect GWPs would still be derived. The shorter lifetime of halons in our model is due to our chemistry (JPL 2002 data, see Sander et al., 2002), which leads to faster decay/loss of halons after a pulse perturbation and thus smaller direct GWP. The faster halon removal means faster bromine release and ozone destruction in the stratosphere, which possibly results in smaller indirect GWP. To get a quantitative sense for this, it requires further study. As mentioned above, our ozone RF due to halocarbons are smaller in magnitude and thus it would possibly explain the smaller indirect GWPs. In section 4.3, we added sentences as below:
A shorter lifetime should somewhat reduce the derived indirect GWPs but the dif-

ferences between the 1211 lifetime from WMO (2007) and from the model is within the uncertainty of the WMO value. As indicated by the longer lifetime we derive for Halon-1301, the large decrease in the indirect GWPs would still be derived.

Specific comments: Page Line Comment Abstract 22-23 Yes, they are smaller, but within the error bars of the previous estimate (WMO, 2007)
→ We changed the sentence accordingly.

15514 13 Provide latest IPCC estimate, which suggests that the net forcing from halocarbons is likely positive
→ One of the points of the paper (and the last WMO assessment) is that we should not directly add the direct and indirect GWPs because the ozone effect is not well mixed so its effects on climate may not directly correspond to the RF value.

15514 16 While there have been many papers and assessments in the past that have made this statement, it is possible (within 1 error bar, IPCC (2007)) for this indirect forcing to be positive and increase the GWP of the halocarbons.
→ As we discussed above, the indirect RF (IPCC, 2007) is based on the estimate accounting for causes of stratospheric ozone change in addition to those due to the Montreal Protocol gases. It would be different from our calculated RF for the ozone change resulting from specific halons.

15514 21 It is exactly linear except for the threshold assumption
→ We corrected it.

15514 22 Another limitation of the EESC approach that should be listed is that the indirect GWP has been calculated assuming that there is a threshold for ozone

depletion such that ozone loss ends when EESC falls below 1980 levels
→ We included the limitation of the EESC approach as below:
However, the traditional approach is based on two assumptions: (1) a linear relationship between the changes in equivalent effective stratospheric chlorine (EESC) and in radiative cooling due to the lower stratospheric ozone loss and (2) a threshold for ozone depletion such that ozone loss ends when EESC falls below 1980 levels.

15517 19 can you briefly discuss the advantages/drawback to using these CTMs rather than a GCM?
→ In the section of discussion and conclusions, we discussed those as below:
Recent development of coupled chemistry climate models such as the Whole Atmosphere Community Climate Model (WACCM) from the National Center for Atmospheric Research (NCAR) enable us to study interactions between ozone and climate in the stratosphere and the WACCM can be used for Halon perturbation simulations. However, huge computational demands of the WACCM at present would keep us from using it for this study. Another aspect involves the requirement of better understanding changes in meteorological fields related to Halons and resultant ozone and feedback among them. Further study with the WACCM is therefore needed although current 3-D CTMs of the stratosphere and the troposphere still requires huge computational resources. The other difficulty with using a GCM is that natural climatic variability will further complicate interpretation of the results.

15518 11 while this model provides some sensitivity results, you have fixed the winds from WACCM and don't allow this feedback; how significant is this limitation
→ Kinnison et al. (2007) showed the sensitivity of the MOZART-3 to several different meteorologies and Pan et al. (2007) showed the effect of meteorology-chemistry interaction on the tropopause. The most significant effect caused by changing meteorological fields will be changes in the tropopause, which would add sensitivity or

uncertainty in radiative forcing. A further study is needed to address this issue. See statement above for further complications to such an analysis.

15518 18-19 it seems that you are using a fixed mixing ratio for the Montreal Protocol gases. This would suggest that you would not capture any potential non-linear response between chlorine/bromine levels and ozone. Also, because the EESC approach of calculating the indirect GWP assumes the threshold, it would be useful to say something about the implications of your 1999 fixed assumption. Of course, we do know that alpha changes with chlorine and bromine levels.
→ We believe that the GWP definition consider the other gases concentration stay in current level. Direct and indirect GWPs for the two major halons could be changed if the Montreal Protocol gases are greatly changed. It would be good for us to see what if the chlorine/bromine levels are changed because the model automatically includes alpha variation. But, such feedbacks are likely to be minor and our results are unlikely to be significantly affected.

15520 7 again 'serious limitations due to the parameterization . . .' seems to overstate the issue since your calculations actually agree with the simple approach
→ We changed it to 'significant'. They only agree because very large error bars (that are being largely ignored by policymakers) were put onto the values from the simpler approach.

15521 3 it seems the wording should be 'is a key indirect effect' or 'are key indirect effects'
→ We changed it.

15521 1-16 I am confused by this justification of using an emission pulse rather than a

concentration one; why is the mixing faster using a mixing ratio BC?
→ It is our finding through various initial analyses with the CTMs that a model study on atmospheric response of a gas following its instantaneous pulse perturbation at the surface should be conducted with the model setup of using an flux emission pulse. Time-dependent changes in its atmospheric concentrations, associated with a pulse perturbation for the calculation of GWP, result from combined processes such as atmospheric mixing and transport as well as chemical processes. The traditional model setup of specifying lower boundary mixing ratios for long-lived greenhouse gases (LLGHGs) cannot be applied for the analyses of LLGHGs' GWPs although they are considered to be well-mixed gases in the atmosphere. It is because it in reality takes a while for the LLGHGs to reach the destruction height level in the stratosphere and reach steady horizontal/vertical distributions through mixing and transport processes in the atmosphere. Even for a steady continuous increase case of LLGHGs, their concentrations in the troposphere will show temporal variation and high spatial distribution during the first several years and begin to be well-mixed after some time. If a mixing ratio BC is used, the fixed mixing ratio over the globe will quickly over-ride the existing concentration ignoring the gradual influence of an added perturbation through realistic atmospheric processes. This is expressed as faster mixing at our text.

15524 22-24 The last phrase of this sentence is confusing to me
→ Is it about model derived chemical lifetime? If so, a lifetime of a gas in a model can be derived from model-determined total amount (loading) and chemical loss rate of the gas in the atmosphere simply by dividing the two. Also we can estimate a lifetime (e-folding) time of the gas from a time evolution of an added atmospheric loading. Time evolution of the gas is basically dependent on model chemical loss rate. Therefore, the estimated e-folding time should be same with the model-derived chemical lifetime for the validity of the perturbation experiment.

15526 10 Doesn't this just arise from the longer lifetime of Halon 1301? This would likely be the case for CFC-11 for an ODP calculation and is certainly the case for the CO2 forcing in a GWP calculation.
→ The effect of CFC-11 on ozone for an ODP calculation is different because ODP calculation does not require a pulse perturbed atmosphere but a sustained perturbed atmosphere. For ODP calculation, a mixing ratio BC can be used as long as total atmospheric loading is exactly known. About the CO2 forcing in a GWP calculation, we already seem to have shown that an absolute GWP (AGWP) for halons evaluated based on the traditional approach of using atmospheric concentration, lifetime, and radiative forcing efficiency are similar to our calculated AGWP using the model time series of tropospheric concentration. In other words, AGWP of CO2 and a gas for a GWP calculation could be the different case from the indirect GWP due to ozone destruction.

15529 8-11 Please provide more explanation for why this is partially guaranteed. It does not seem obvious.
→ As we discussed above, the time evolution of a gas following an pulse injection into the surface air should be dependent on the model chemical loss rate (or lifetime). Our calculated time-dependent changes in the gas show the agreement and thus guarantee the validity of our model simulations.

15536-37 why don't the masses end the run close to or above the starting mass? Is this because you are starting out of steady state? If so, a perturbation figure would be more meaningful. Otherwise, more explanation is needed to interpret this figure. Perhaps the only issue is that you have not included a point at year 0, which would make things more clear.
→ We revised the figure to include a point at year 0.

Figures

Figure 1. Changes in tropospheric mixing ratios of Halon-1211. Red line with circle marks denotes 2-D model outputs in default setup and blue line with cross does outputs with increased baseline emissions

Figure 2. Changes in radiative cooling change due to two Halon-1211 perturbation experiments shown in Figure 1. Red line denotes 2-D model outputs in default setup and blue line with cross marks does outputs with increased baseline emissions.

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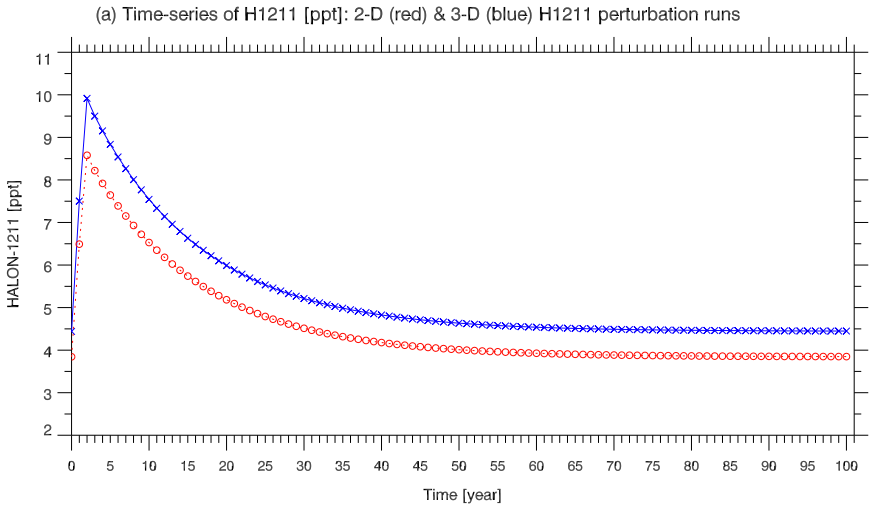

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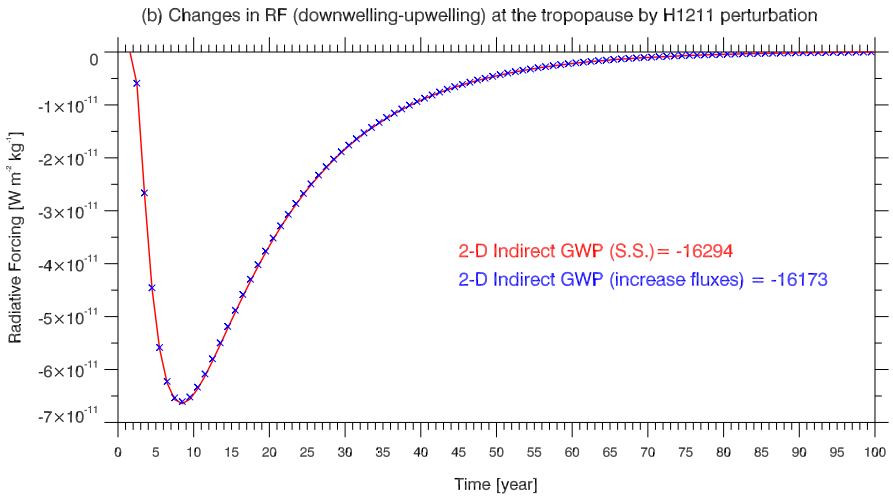

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