Rev of acp-2019-1205
Thornhill et al. "Effective Radiative forcing from emissions 1 of reactive gases and aerosols – a multimodel comparison"

This paper is a very important contribution to climate and atmospheric chemistry studies. It is critical for the current IPCC AR6 assessment. My apologies to the authors for the delay in my review – surprisingly, these isolation times do not make it easier to review. I am rushing to get this out and so there may be typos in this review.

AerChemMIP is a very important project that is trying to make sense of a complex, nonlinear system of chemistry and clouds. The design has some serious flaws, and we all knew that as it was being developed since there were obvious limitations in the number and complexity of experiments. First, there is the nonlinearity, which comes our clearly in these results: Exp(A) plus Exp(B) does not equal Exp(A+B). Further, and because of the nonlinear nature, the choice of reference atmosphere (1850) will produce quite different results than another (2014). Personally I would have much preferred to use the 2014 atmosphere as the reference atmosphere (at least we can compare the models to measurements) and then subtract the emission or concentration changes from 1850 to 2014. So, we live with AerChemMIP and use it. The analysis here is very good, but needs to develop more of an "assessment" view when reporting final numbers. They should reflect some (subjective) adjustment of values (for non-linearity) or bias (from 1850 atmosphere). Simply reporting the model average for Exp(A) and then Exp(B) will lead anyone who uses this paper to assume that the combined effect is the sum. That would not be good for either policy or attribution work. If the sum is the best answer (I would think that so), then the contributions of components may need ot be expressed in % of total rather than in W/m2. I do not recommend a substantial rewrite, but rather a self-assessment by the author team of how to use these results.

The abstract starts off being very careful and clear, but then I get lost between emissions and composition change. For example, SO2 is clearly an emission-based ERF, while CH4 is an abundance-based ERF. VOCs & NOx are obviously emissions (having direct RF) but O3 is not emitted. The HC (L40: halocarbons? as opposed to NMHC?) are negative presumably from the ozone depletion, but this is an odd way to list the CFCs. Methane is singled out at the end of the abstract as increasing ozone, but ozone is listed separately. Is the methane based only on the change in CH4 concentration? Further, I suspect that the methane includes stratospheric water vapor which is indirect as is ozone, so why the separate listings. This is just inconsistent and you really need to present the framework and rules for partitioning and assigning ERF. Basically, AerChemMIP is a complicated set of overlapping experiments and thus the abstract with simple results is very difficult to write correctly. Try to keep it clear and simple.

L53- aerosols are chemically reactive. try again – aerosols and gases that …

L56- climate feedbacks on natural emissions is a tough one for the non-CO2 species. I do not think AerChemMIP did anything on that.

L68- "conditions" do you mean SST, I doubt you prescribed different chlorophyll or DMS?

L69- why aerosols here? It seems more like and ie than and eg, why not just say aerosols and gases.

L71- again the contrast of 'aerosols and chemistry' is really a poor description of AerChemMIP. Aerosols are a chemically reactive species (most of them are, they were created by gas phase chemistry). This Intro really must have a better inclusive discussion of greenhouse gases (including ozone), of indirect greenhouse gases (CO) and of aerosol (primary and secondary). This is at odds with the content of AerChemMIP.

L90- again, every time I read this, it sounds odd and misleading: " aerosols and reactive chemistry" are not opposites.

L93- again, this phrasing sounds wrong: " anthropogenic and reactive species" is this tow separate species or is it 'both'

L103- what does 'down to' mean? from 0.001 to 560 hPa?.

L107- typo 'is includes'

L102-L165 This information should really be in a table somewhere, not in the text. Focus on the results. In fact, most of this is already in Table 1.

L172-173- You should say 'emissions' with the NTCFs, also you should note that methane is an SLCF, which your statement seems to preclude. Also, I thought that SLCF was new preferred, but…

L174- There is a serious problem with the AerChemMIP as defined and we realized this at the time, but did not address it: viz. because of the large changes in atmospheric chemistry and oxidants between 1850 and 2014 (including the ozone depletion), it is not clear that the effect of today's NTCF emissions in today's atmosphere are anywhere close to those calculated here for the PI atmosphere. This needs to be addressed when compiling results and clearly adds to the uncertainty. The reason why it is dangerous is that it could totally misrepresent the magnitude of the response if we were to cut NTCF today.

L200- "ocean state" implies much more than SSTs – do you really mean that.

L235- I am confused here, it seems like the direct forcing would include, not ignore, the absorption and scatting by aerosols. Is this a typo.

L239- this section is very confusing as written. I am sure it is simpler that it seems but some of the writing seems incorrect. In this line surely you mean the aerosol direct radiative effect, since "radiative effects due only to aerosols" would imply both ari and aci. The notation for ERFcs,af is inconsistent between eqn and text (comma or not). Eqns 4 & 5 sum to give not ERF, so where is this missing 'surface albedo' term (ERF-ERFcsaf) and does it matter? It must certainly be counted as an aerosol ERF. [OK, I see this in Table 3 later, is the cs,af just noise?]

L254ff- This is odd, you said just above the SARF is calculated from ERF – A_trop, so of course this should give 0% difference. Are you just checking the math? Also, with SARF calculated as a residual term, and the ability to denote and sum all the A_trop being highly uncertain, SARF would not appear to be very certain. In fact the SARF term would depend on the models' ability to diagnose A_trop correctly.

L260- With BC, the long-standing problem is that some models get far too much in the stratosphere and that would cause a very large SARF.

L285- in this figure and some others, please define carefully what the shorthand for the terms means.

L296ff- This is a very good discussion of the aerosol components!

L321- This would be a much better lead off to the aerosol section and analysis, begin with the big picture before the weeds.

L343 – just put this table into the figure, it is just a summary of the bars anyway.

L354- Please, stop wasting space, Table 5 and Fig 4 has the same information. If you want to show AOD, then add it to the figure.

L370ff & Fig 5- I do not understand the purpose of this AOD scaling, it really does little to help. The figure shows the key data: ERF from parts vs ERF from all. The ERF-parts consistently over accounts for the combined ERF. This is as expected wince the cloud effects are largest in a pi atmosphere with little background aerosols. So herein lies one of the fundamental problems with the AerChemMIP that must be acknowledged and accounted for. I am not sure that scaling by AOD is any more justified than just scaling the indirect to match – An interesting question is whether the ERF-ari sum of the parts equals the whole? Keep this fudge factor simple since there is no correct way to do this.

L393- Again, this title jars a bit, SO2 and NH3 are reactive gases.

L406- How big is the error in GHGas ERF if one ignores clouds? I would think large.

Fig 6 & Table 6, can easily combine and understand the Std Dev better.

L419- Here is a case where some assessment is due as to how well these model simulations are accurate in the sense of including all the effects. As noted the N2O-O3 link is important and missing in some, and the other key link between N2O and tropospheric OH and methane (and maybe aerosols) is established but missing here.

L431-441- This is a very interesting and important discussion about the additivity of the components. I suspect that the CH4 result is similarly affected.
BTW, where is the effect of stratospheric H2o form CH4 noted or counted?

L480- Yes halocarbons, but also N2O, and N2O may be more important since it depletes ozone in the tropics (there are papers on this). I do not wee what difference stratospheric Cl will make on the total lifetime – hopefully the Stevenson paper becomes referencable. Also, you need to be careful here since the methane feedback factors ff, apply to the PI atmosphere, and that is different from the present, particularly lower CO and NOx…. . The feedback factor used for GWPs etc, is the current one, not the PI one, so these results should NOT be used to change any previous assessments.

Fig 11 is really hard to understand or see clearly, it will need a cleanup.

L525- Here is a very important statement and I am not sure that you have put together all the reasonable uncertainties or non-linear scaling. The individual components here must be adjusted to recognize that the total ERF (with all simultaneously) is much less than the sum of the components. Thus you cannot recommend the individual results without scaling and without increasing the uncertainty.

L561- Same as above. The individual values will not sum correctly and so these do NOT reflect the ERF of NOx emissions as we progress from 1850 to 2014. Thus they should not be used as part of an assessment until they are more critically evaluated and put in context.
These are all very important AerChemMIP results, and the analysis here is highly valuable, but their use in attribution and related studies should reflect the bias and uncertainties in combining nonlinear parts that were calculated separately, and in basing these all on a pi-atmosphere.