

Interactive comment on “Halogen chemistry reduces tropospheric O₃ radiative forcing” by T. Sherwen et al.

Anonymous Referee #2

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Review of “Halogen chemistry reduces tropospheric O₃ radiative forcing” by T. Sherwen et al.

General Comments

This modelling study nicely illustrates that including tropospheric halogen chemistry has important consequences for ozone changes since pre-industrial, and likely means that the radiative forcing from tropospheric ozone is smaller than previously thought. The scientific approach is broadly sound, although a few caveats and extra discussion points should be added (see specific comments below). In particular: whether some representation of halogen losses may have been included (inadvertently) previously in the ozone deposition to sea-water term (and whether there is a danger of double counting if this is not adjusted for); and the approximation of calculating O₃ RF from

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a column change. These potentially add some extra uncertainties, but don't change the fundamental conclusions reached here. Somewhat unbelievably, the authors fail to include a Conclusions section. I think such a section must be added for completeness. If these revisions are included, then this paper should be acceptable for ACP.

Specific Comments

P1 I10 from -> associated with decreases in

P1 I22 delete comma

P3 I19 What about anthropogenic CO emissions? Biogenic VOC? Soil NO_x? Lightning NO_x?

P3 I20 Reducing biomass burning emissions to 10% of their present-day values may be what Wang and Jacob did in 1998. However, more recent studies (e.g. Knorr et al., 2016; doi:10.5194/bg-13-267-2016) suggest that biomass burning (at least wild-fires) in pre-industrial times may well have been higher than present day values (less fire suppression; more vegetation). I am not suggesting you redesign and rerun your experiments, but mention any implications of uncertainty in the evolution of fire emissions.

P3 I20 If you insist on using the (I believe more 'correct') way of expressing trace gas mixing ratios (i.e. pmol/mol, etc.), do get the prefixes correct. I think that pre-industrial methane was 700 ppb, or 700 nmol/mol.

P3 I29 ...to be zero in the pre-industrial.

P3 I30 delete no

P4 I6 Do you really mean Br emissions increase 'due to increased iodine driven sea-salt cycling'? Or do you mean I (or halogen) emissions increase?

P4 I7 An increased stratospheric influx of Br isn't strictly an 'emissions' increase.

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P4 I10 Same point as above, not all 'emissions'.

P4 I15 15.2 days

P4 I16 shorten

P4 I16 4.5 days

P4 I20 in -> is

P4 I23 Fig. 2 doesn't show the change in O₃ since pre-industrial – I'm not sure the figure reference is helpful.

P4 I27 Strictly, Table 2 shows Ox budgets (at least that's what the caption says). I think define Ox (O₃ + NO₂), and include the extra minor terms in Table 2 (NO₂ deposition, NO₂ burden, NO₂+OH sink(?)), or at least clarify the (subtle) differences between an O₃ budget and an Ox budget.

P5 I3 that -> than

Much of the ozone difference due to halogens over the oceans is near the surface (I think – you don't actually show a vertical profile of the ozone change). I wonder if some models previously inadvertently 'accounted' for this within the O₃ deposition velocity over (sea) water? This may mean that the O₃ deposition velocity over water needs reducing when halogens are included, or you risk the models double-counting this effect. It is obviously better to include it explicitly as a halogen chemistry process, but clearly we don't want to double count it. Is this possibility worth discussing?

P5 I10 tern -> turn

P5 I13 NB, using a column ozone change to calculate radiative forcing adds the approximation that you assume the vertical profile of the ozone change is the same as that in the original full calculation. If most/all of the ozone change is originating at lower levels, the real radiative forcing will be less, as the RF depends upon the temperature of the layer of the atmosphere where the ozone changes, as ozone changes in colder

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layers generate a larger RF. (This is why ozone changes in the very cold tropical upper troposphere generate the largest RFs.) As your O₃ changes are mainly near the surface (?), this suggests that this approximate way of calculating the RF will return a higher value than if the full radiative calculations were performed. I think this should at least be mentioned.

P5 I24 Related to the above point, I think the O₃ RF from the stratospheric flux changes are likely underestimated, and those originating nearer the surface over-estimated, simply by using the O₃ column approach.

P5 I25 in -> is

P6 I7 radiative forcing

A Conclusions section should be added. Isn't a Conclusions section almost mandatory for a scientific paper? You certainly have some, so include one! This seems a rather inexplicable omission.

P12 Figure 1A. The caption states this is a flux, but the units are labelled as pmol/mol. A flux must have units of mol or kg per unit time.

P13 Figure 3. The vertical axes need units (km?)

P14 Figure 4 caption. Zonal mean. Increase -> increased. The last 2 sentences have 5 "due to" s!

P15 Figure 5 Units of ozone should be nmol/mol?

P18 Table 2. How is the troposphere defined? (i.e., by PV, T-gradient, or O₃ concentration?). As mentioned earlier, clarify if you mean O₃ or Ox.

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