

Interactive
Comment

Interactive comment on “Tropospheric bromine chemistry: implications for present and pre-industrial ozone and mercury” by J. P. Parrella et al.

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Parrella et al present very interesting results of the inclusion of bromine chemistry in two global CTMs with a strong focus on results from GEOS-CHEM. The manuscript is well written and shows very interesting results of the impact of bromine chemistry on global ozone and OH concentrations as well as mercury cycling. The impact of bromine chemistry on mercury concentrations is discussed as well and runs without anthropogenic emissions are included in the discussion. I suggest publishing the manuscript after minor modifications have been made.

Specific comments

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With a study like this there will always be some shortcomings and most of them are discussed in the manuscript but I think that a few need a bit more thorough discussion.

(i) Polar bromine explosion: It is surprising that p-TOMCAT requires the inclusion of a polar source to reproduce the satellite columns but GEOS-CHEM doesn't. This indicates that the required simplified inclusion of bromine chemistry in global models still has quite significant uncertainties. Some of these might also be "hidden" by the presentation of large scale averages. Please discuss this more.

(ii) Release of bromine from sea salt: The use of the assumption that 50 % of bromide gets released is a simplification of a rather complicated multiphase reaction cycle which should probably be acknowledged more. Release of bromine from sea salt is acidity dependent and hence one would expect latitudinal differences and also differences in modern versus pre-industrial conditions. Details of this will require a much more thorough examination which is beyond the scope of this paper but these shortcomings should be acknowledged and this caveat discussed when the pre-industrial runs are presented. Arguably anthropogenic emissions have increased the fraction of bromine depletion from sea salt which cannot be captured with the "50% release" assumption so the bromine chemistry in pre-industrial times might have been different from what is presented here.

(iii) Heterogeneous/multiphase reactions: This paper showed again that the multiphase cycling of bromine is key to maintain realistic (as compared to satellite columns) BrO mixing ratios but it should be acknowledged that the lumped treatment of R29 and R30 is a great simplification and that these reactions are in fact not occurring as listed but involve multiple steps.

(iv) Comparison with data: Only satellite observations are considered for comparisons, which is to some extent understandable for a global model study. Nevertheless, the long-term data from Cape Verde (Read et al., Nature, 2008; Mahajan et al., ACP, 2010, see their supplement) provide quite good coverage over 8 months and show large

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variability. This data set is not even mentioned in the current manuscript which should be improved. How does GEOS-CHEM fare in the tropical North Atlantic?

(v) GEOS-CHEM vs. GOME-2: GEOS-CHEM underestimates the satellite columns by about 30%. You wrote that this is within the error margin but it could also indicate that the sources of reactive bromine are stronger or lifetimes longer than assumed/calculated in the model. This is especially interesting in the light of the study by Hossaini et al (ACP, 2010) who presented detailed breakdown schemes for CHBr_3 and CH_2Br_2 . Their work suggested that the effective yield of bromine atoms is less than the maximum yield as assumed in this manuscript which would reduce the modelled bromine source.

Technical comments

p. 9670, l. 22: These uncertainties should be discussed in more detail, here or in section 3, see above.

p. 9673, l. 24ff: See discussion above. Please add more discussion about the simplifications.

p. 9675, l. 14: When referring to mol mol^{-1} please use the correct term “mixing ratio” rather than “concentration”. Please correct here and all other occurrences in the manuscript.

p. 9677, l. 7: Why do you use an asymmetric time window around the satellite overpass time?

p. 9678, l. 16-26: Are these sensitivity studies specific for the tropics? If not, please consider moving to the related discussion on p. 9677, l. 20-25.

p. 9680, l. 9-10: I can't read from figure 3 that HOBr sinks drive ozone loss, please explain.

p. 9684, l. 4-5: It might be worth to highlight this result a bit more and add that this is

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an effect of about 20%.

p. 9685, eq (2): I couldn't quite follow the derivation of this expression, please explain.

p. 9685, l. 24 – p. 9686, l. 4: This is a very interesting result but please highlight the uncertainties with your “pre-industrial” runs somewhat as the bromine release from sea salt could be significantly different, see above.

p. 9686, l. 13 – 21: These are interesting thoughts but again I feel that the caveat and uncertainties that underlie them should be mentioned.

p. 9688, l. 6-9: Again, very interesting but please mention the uncertainties/caveats.

p. 9697: The Saiz-Lopez paper is now in ACP

p. 9697/8: JPL evaluation 17 was published in 2011, not in 2010

p. 9711, caption: What do you mean by “all species are in steady state” On what time scales is this the case? Daily? Day vs night? Seasonally? Please explain.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 9665, 2012.

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