Atmos. Chem. Phys. Discuss., 8, S5878–S5882, 2008
www.atmos-chem-phys-discuss.net/8/S5878/2008/

Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on “Consistent simulation of bromine chemistry from the marine boundary layer to the stratosphere – Part 2: Bromocarbons” *by* A. Kerkweg et al.

A. Kerkweg et al.

Received and published: 12 August 2008

We thank the anonymous referee #3 for the helpful comments.

Conclusions regarding a longer CH3Br lifetime are not supported (abstract, conclusion, p. 9492). CH3Br emissions are said to be derived from observed surface concentrations and a modeling approach TNUDGE, which is not qualitatively described in the paper thus making it difficult to assess its robustness. The observed surface concentrations used to constrain emissions are minimal: AGAGE surface concentrations are available from 2 sites only –these are assumed to be representative of all longitudes and are extrapolated to all latitudes (other, more widely distributed and published data is not considered).

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

We agree that the data is poor. But we are definitely talking about more than 2 sites. Most probably this is a misunderstanding caused by an incomplete reference for the data. We will extend the citation for the data base naming it “ALE/GAGE/AGAGE database”, which is consistent with the reference (Prinn et al., 2000) and we will add the additional reference to Montzka et al. (2003) which was unfortunately missing. Furthermore we will add two sentences on TNUDGE.

A significant flux in the CH3Br budget is related to oceanic production and loss –no mention of how the ocean is handled with respect to CH3Br is considered, yet a careful study of the CH3Br lifetime would include detailed consideration of this process.

As we apply an atmospheric model no ocean model is included in our simulation and the source from the ocean and the loss to the ocean have to be parameterised. The loss is calculated via the submodel DRYDEP which calculates the loss to the surface distinguishing 4 different types of surface (’water’ or ’ocean’ is among them) and taking the reactivity and the solubility of the respective trace gas into account. Other surface losses are calculated by the submodel TNUDGE. TNUDGE simply adjusts simulated tracer mixing ratios to observed mixing ratios by prescribing the observed mixing ratio in the lowest model layer by a Newtonian relaxation technique (Kerkweg et al., 2006a). Alternatively, the ocean could only be accounted for by including seawater observations. We agree that the data base used for the nudging is relatively sparce, but to conclude that we do not take the ocean into account at all is wrong.

If oceanic loss is ignored in a simple assessment of CH3Br lifetime, the CH3Br lifetime becomes greater than 1 yr. Given these shortcomings, I conclude that this estimate of lifetime does not represent an advancement in our understanding of this important quantity.

As stated above we do not ignore the ocean loss completely. But we also agree, that a model study calculating the oceanic loss and source directly would give more insight into the lifetime of CH3Br, but we did not have that possibility for this simulation. Nevertheless, it might be an indication that the lifetime is greater than expected so far.

This is also supported by Montzka et al. (2003).

The lifetimes quoted in WMO (2006) are explicitly stated to be local lifetimes (Table 2-1 in WMO report), i.e., calculated for a specific OH and light flux. The actual lifetime you calculate in your model is dependent upon the emission distribution you use. Hence, it is not appropriate to suggest that the CHBr3 loss included in your model is larger than suggested by lifetime quoted in the WMO report (p. 9484, lines 20-30). You would need to calculate a local loss given the OH rate constant and photolysis parameters for equivalently averaged time and space to address the issue you hope to. (Text to qualify of the WMO lifetime in this regard in your Table 2 seems necessary too.)
We agree and we will rewrite the discussion about bromoform (also for other reasons, see answer to referee #1). Additionally, we will added the information about the local lifetime to Tab. 2.

CH3Br emissions are time-dependent, given the changes in industrial production that have been reported since 1998. While some accounting for this fact is included in the manuscript, it would be clearer if in Figures 17-23 years of the campaign were explicitly indicated in the caption. Also, it isn't explicitly stated that emissions were derived based on observations in the year 2000 alone, is this true?
The observations are calculated individually for each month and year. Thus the annual average shown in Figure 17 is specific for the year 2000. We will add the time information in the figure captions where it was missing (Fig. 17/18/20/21).

The section on "Bromocarbons as sources of reactive bromine" is interesting, but I can't see how the entries in Table 3 are consistent with Figure 24 regarding the rate of Br radicals from CH3Br and CHBr3. A fairly similar area above the tropopause looks to be shaded non-purple for both compounds (CH3Br max is even larger than CHBr3), and yet the rate of Br production from CHBr3 is stated in Table 3 as being 2.5 times higher than from CH3Br...?
Yes, you are right. Our diagnostic tracers are tracing the destruction of the halocarbons. In Fig. 24 we forgot to multiply with the stoichiometric factor (number of bromine radical

produced by the decomposition), but we did so for deriving the numbers in Table 3.

Furthermore, it seems that the graphical projection is drawn as a function of latitude, which potentially under-represents tropical latitudes. You might recheck the calculation and explain it, the averaging or summing, and weighting more fully.
You are right. At least the unit given in the figure caption is wrong and will be corrected. Anyhow, according to suggestions of referee #2 we will replace Fig. 24.

Also in this section, some imprecise/unsupported text: Line 20-25, p. 9493: “Stratospheric bromine chemistry becomes important above...” with respect to ozone chemistry or something else?
Yes, of course. We will delete the whole section following suggestions of the other referees.

Furthermore, does most of the Br from CHBr3 formed in the lower stratosphere reach the upper stratosphere or not?
Unfortunately, there was no diagnostic tool available for the simulation to derive this quantitatively.

Line 23-26, p. 9494: The sentence “In the stratosphere the photolysis of halons contributes substantially to the Br production” is not supported by any figures or discussion. On what basis is this conclusion drawn?
We will add the calculated numbers to Table 3 and discuss this issue in more detail.

Can the accuracy of the Br production rates calculated here be assessed to any degree by the total Br mixing ratios they would imply? Mixing ratios of inorganic Br are not given, but might further confirm, if they were calculated to be above 25 ppt, for example, that the contribution of CHBr3 is likely overestimated here. This is one reason for the substantial interest in these compounds, no doubt. The authors have a tool to address this important issue, and for some reason don’t follow through.
The mixing ratio of inorganic bromine is mainly below 25 ppt. But in the tropics around 10 hPa this threshold is reached. We agree that this can be an indication, that $CHBr_3$

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

is overestimated. As the mixing ratio of inorganic bromine is also influenced by the release from sea salt aerosol, we intend to discuss this within the third part of this series of articles which is dedicated to release from sea salt. Only after the analysis of the sea salt source strength for inorganic bromine, we can discuss all aspects of the inorganic bromine mixing ratios. We prefer to discuss this issue once all sources are analysed, because only this can give the full picture.

Following up on this, the text in the conclusion regarding CH3Br (lines 18-20) should include the point that the contribution to stratospheric Br calculated here is also likely overestimated.
Line 18-20 comprise a statement about CHBr3, thus we assume that you thought about bromoform. We will add the information according to the results of the sensitivity study performed due to the comment of referee #1 (see this comment and the associated final comment).

Finally, I'm not sure all the multi-panelled figures are necessary. Furthermore, they don't readily convey how any discrepancy or agreement varies with region or season (e.g., line 19, p. 9491). Some summarising of these differences might be useful. Perhaps plotting flight tracks on modelled distributions would be helpful too.
We decided to show the full set of available measurements from the PEM campaigns in an extra document within the supplement. Consequently, we reduced the number of shown profiles within the paper to 5 panels per comparison. Apart from $CHCl_2Br$ we found no differences between the different PEM-TROPICS campaigns. As these are the only data we have access to, we cannot identify any seasonal or regional differences.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 9477, 2008.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper