

Interactive comment on “Two decades of OH variability as inferred by an inversion of atmospheric transport and chemistry of methyl chloroform” by P. Bousquet et al.

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OH variability and aggregation errors.

This paper is an important contribution to the subject of inverse determination of OH from (methyl chloroform (MCF) measurements. There are two important conclusions. 1. Using the same emission estimates, similar year-to-year variations in OH are deduced compared to earlier estimates of Prinn et al. (2001, and recently 2005) and Krol et. al 2. It seems feasible to deduce information on sources (i.e. emissions) AND sinks (OH, ocean, ..) of MCF within the inversion set-up.

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Doing this kind of inversions with a 3D high resolution model is a major task. As stated, the methods developed are useful for longer lived trace gases such as CO and methane. The paper is well-structured with technical details nicely hidden in Appendices. It should be published in ACP. However, the two main points are addressed below need some attention.

OH variation or uncertain emissions?

The first point above implies that it does not seem to matter whether a 2D or 3D climatological model is used or, like here, a 3D model with time-varying meteorology. Subtle differences in deduced trends and OH levels are found but the year-to-year variations seem to be robust. One could thus conclude that the inferred OH-variation are real (and alarming!). Another plausible conclusion, that comes into play only at the end of the paper (and is mentioned in Krol and Lelieveld (2003) and Krol et al. (2003)) is that the assumed accuracy of the applied MCF emissions lower than assumed. As ever in inverse modelling, the choice of the prior information comes into play here. What do the authors use as prior information concerning the sources and sinks of OH? In section 2.5 it is stated that the uncertainties on monthly OH are set to 100% (in each of the 4 latitude bands). Month-to-month errors are assumed uncorrelated. In section 2.6 an additional constraint is set that limits the month-to-month variations in OH to 50%. On the source side, the emissions of McCulloch and Midgley (2001) are adopted with the stated "tight" uncertainties. Not surprisingly, the results (e.g. figure 9) thus clearly show that most of the model-measurement mismatch is attributed to OH. I have strong doubts on these choices and wonder why the authors have chosen this scenario as the 'base' case. Further in the paper they show that a 15% OH uncertainty indeed results in larger attribution of the model-measurement mismatch to the MCF emissions. Why do a disagree with the choices? From in-situ measurements of OH and from photochemical theory we seem to understand OH chemistry better than the adopted 100% error. Many dedicated experiments have shown that our understanding of the background photochemistry is rather good. Furthermore, from methane obser-

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vations it is clear that year-to-year variations in global OH of 100% are impossible. On the emission side the situation is reversed. Recent evidence shows that the prior uncertainty estimates in the McCulloch and Midgley paper (2001) do not hold. Even from background sites at Mace Head, Jungfraujoch (Reimann et al. 2005) and Trinidad Head (Li et al., 2005) larger emissions are observed. Sampling in polluted continental airmasses generally show even stronger signs of recent emissions (Krol et al. 2003, Barnes et al. 2003). Although the exact estimates of ongoing emissions are difficult to make, it shows that the errors in the emission model are (much) larger than stated previously. It is this unbalance in prior estimates of the sources and sinks that drives the results. In conclusion, a more realistic error estimate for sources and sinks is clearly needed.

Aggregation error

One of the main reasons that is mentioned for optimising monthly OH in four different regions is the aggregation error. The theory is that if something wrong is 'fixed' in the inversion (e.g. the seasonal OH variation), the inversion will translate this error in parameters in the state vector that are allowed to vary. However, a limit to the allowed variations should be set based on physical realistic assumptions. A simple example: Suppose that we optimise OH in all model grid-boxes on a monthly basis. One can imagine that a good fit with the measurements can always be found by varying OH wildly in the 'measurement' grid-boxes alone. Months with a modelled overestimate of MCF would require higher OH while under-estimates are solved 'solved' by lowering the OH locally for that month. A perfect fit with the measurements will always be possible. Again, it boils down to the same thing: a good prior estimate of the errors. In this respect it is worrying that the authors only show de-seasonalised variations in the (optimised) OH concentrations. Even these values show large year to year jumps (e.g. figure 6, SHT OH) which can not be understood from photochemical theory. In conclusion, I would welcome a more detailed discussion on the issue of the aggregation error.

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Minor points Page 1682, line 27: I would like to see the resulting seasonal cycle. See point above concerning aggregation error.

Page 1684, line 10: Why monthly averages and not 'daily' or weekly values? Line 19: 'Data' should be 'data'

Page 1686, line 17: This fast NH-SH exchange will have consequences for the MCF transport. If emitted MCF is transported too fast to the SH, more OH on the SH will be needed (see page 1697).

Page 1688, line 1: subscripts do not match the formula above. Line 23: It is stated above that figure 4 is a linear version of eq. 3. What does a linear version of eq. 4 mean?

Page 1690, line 6: Is there a way to show the impact of using this average response function for the 1980s. I understand that the whole calculation is rather involved but I cannot judge the impact of this choice. The same holds for the assumptions in Appendix A for formula A3. It would help to show in some way the loss in accuracy associated with the assumptions.

Page 1692, line 13. planet should be planetary

Section 2.5: see point above.

Figure 1: for SPO the minimum is larger than the mean.

Page 1694: How did you sample NWR? Sometimes there are good arguments to sample the model not at the surface but in layer 2 or 3. Line 24. This is hard to observe from figure 2.

Page 1965: line 5: 'is' should be 'if' Line 15: this reduction is quite large. Further in the paper (with OH errors set to 15%) even larger reductions are found, despite the fact that a constraint was set on the total flux. Maybe the error on the total emissions should be smaller, since this number is accurately known. It might be the timing in the

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emissions that is more uncertain.

Page 1697: line 3: If more parameters are used in the optimisation (and the prior error is reduced) it does not come as a surprise that the fit to MCF measurements becomes better!

Page 1699: varying meteorology. It would be interesting to see if the residuals with varying meteorology are smaller than with reusing meteorology. If not, I would like to see a better validation of the model, e.g. comparing with methane/CO observations at Mace Head. Using actual meteorology should be important (e.g. Bergamaschi et al., ACPD, 1007-1066, 2005).

Page 1702: Line 8: I doubt if the bounds are really realistic. Recent studies suggest that they are not (see main comment).

END OF REVIEW

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 1679, 2005.

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