Atmos. Chem. Phys. Discuss., 5, S729–S736, 2005
www.atmos-chem-phys.org/acpd/5/S729/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Two decades of OH variability as inferred by an inversion of atmospherictransport and chemistry of methyl chloroform" *by* P. Bousquet et al.

P. Bousquet et al.

Received and published: 11 May 2005

We thank Dr KROL for his relevant remarks on our work. In the following, we answer point by point to his concerns.

Principal comments

(1) OH variations or uncertain emissions The aim of our paper is double. First, we wanted to see whether or not the large OH year-to-year variability is robust to limitations noticed in previous studies (Prinn et al., 2001 ; Krol et Lelieveld, 2003) such as 3D model, inter-annual meteorology, and joint optimisation of sources and sinks of MCF. In other words, is there still large year-to-year variability of OH radicals when ad-

dressing these issues? The answer is yes, and corresponds to section 3 of the paper Second, we wanted to question some of the hypothesis made on OH. Prior OH distribution is taken from MOZART model (Hauglustaine et al., 1998). In the standard case, we assume a ś100% error on OH concentration each month (§2.5, p1691, l26-27) and month-to-month differences of OH concentrations are constrained to be smaller than ś50% of the prior value (§2.6, p1693, l15). The latter constraint avoids getting unrealistic seasonal cycles of OH (see below) radicals, whereas the former let the mean and inter-annual to decadal variability adjust almost freely. As mention in Dr Krol's comment, a ś100% error oh OH concentrations each month is hardly compatible with what is known of atmospheric chemistry. It is a crude hypothesis. Also, the amplitude of uncertainties on MCF emissions can be questioned. But if one assumes McCulloch & Midgley (2001) uncertainties to be correct, what does happen to OH variability if more realistic prior uncertainties are used for OH ? We reduced OH prior uncertainties step by step and observed when changes in MCF emissions were not any more compatible with inventories at a 2-sigma level. The main conclusion of this exercise is that the amplitude of OH year-to-year variability is not robust whereas the phasing of changes is. This means that as long as inventory uncertainties are not reduced, atmospheric inversion can only provide a large range of OH possible variations from our ś15% case to the ś100% case. This second aim corresponds to the section 4 of the paper.

We built the paper as a progressive view of what is robust and what is not robust about OH variations, a bit on the model of Krol & Lelieveld (2003) paper. We feel this is a classical but efficient way to address things, showing first that an update of previous methods more or less gives the same answer and next, questioning the hypothesis to go one step further. To our mind, figure 10 showing the possible large reduction of OH variability is, at least, as important as figure 8 or figure 5 showing large OH fluctuations. Our ś15% case will certainly be more compatible with the CH4 cycle (for instance) than the standard inversion of section 3.

We propose to make this progression more visible in the paper (from abstract to con-

ACPD

5, S729–S736, 2005

clusion), for the reader to clearly understand both objectives and not only the fact that OH presents large year-to-year variations that are known not to be incompatible with other trace gas cycle such as CH4.

(2) Aggregation error This is a very good point. In the submitted version of the paper, we do not really justify the choice of four OH regions by the problem of aggregation error but more by the fact that we have one station per latitude band over the whole period (p1683 - l5-8). However, the issue of the aggregation error is of great importance (Kaminski et al., 2001 ; Peylin et al., 2001) and we propose to add the following paragraph in §3.7 (p1700 - l3), when discussing the sensitivity test I-14 :

—START When solving only for 1 global OH region, 1 global MCF source region and 1 global ocean region (I-14), OH variability is increased by 11%, indicating that the impact of aggregation error is significant and tends to emphasize OH variations. Aggregation error occurs when an inverse procedure only solves for one scalar factor for an ensemble of model cells in space (making a region) and/or in time (Kaminski et al., 2001). Then if the space and/or time pattern is wrong, the inversion may scale the region source/sink for wrong reasons because of this hard constrained put on aggregated model cells. A solution to limit the aggregation error is to largely increase the number of regions solved for, and to provide soft constrains in order to regularize the inverse problem (Enguelen et al., 2002). Such soft constrains can be error correlations in the flux space (off diagonal elements of P matrix, see appendix C). Setting error correlations that are physically based is not an easy task, and several groups are currently working to produce consistent variance/covariance matrices of error based on biogeochemical models (Peylin et al., 2005, Rodenbeck et al., 2003). Anyhow, the problem is always to find a compromise between the quantity of available information in the observations and the number of degrees of freedom in the flux space. If one has only a small number of observations, it may appear that soft constrains will almost turn into hard constrains (error correlations very close to 1). In this case, it makes sense to directly use hard constrains, that is to solve for large regions. Considering 1/

ACPD
5, S729–S736, 2005

the small number of available MCF stations over the 20 years, the fact that OH sink is 3-dimensional and 3/ the very large computing time that were necessary to address inter-annual variability, we chose to solve only for 4 OH regions and 4 MCF regions, knowing that we do not fully address the issue of aggregation error. ——END

We also propose to add the following short paragraph in section 5 (conclusion) of the paper about these possible improvements.

——START A possible improvement of this work would be to solve MCF emissions and OH sink at the model resolution and to set up, at the same time, error correlations based on biogeochemical models and measurements. It would limit the aggregation error discussed in section §3.7. This will surely require the adjoint of the CTM both for transport and chemistry, and theoretical developments to estimate possible error correlations in OH fields and MCF emissions. ——END

Minor comments

P1682 - l27 : Seasonal OH is discussed in §3.6 (p1697 - l20-26). We did not included a figure because the paper is already dense, but the figure B below plots the seasonal prior and optimized OH variations. One can observe the reduction of seasonal amplitude in the northern hemisphere and the increase in the non tropical southern hemisphere. Also clearly visible are the large anomaly in seasonal cycle occurring in NHT in 1985-87 and in SHT in 1988-90, producing the large inter-annual anomalies of the figure 6 of the paper. These anomalies can be clearly questioned in regards to what is known of atmospheric chemistry. The ś15 case (red solid line) produces 60% less OH variability, associated with very small changes in the seasonal cycle compared to prior estimates, that seems much more reasonable.

We propose to add this figure and the comment above at the end of section 4, as an argument in favour of reducing uncertainties on prior OH estimates (section 4).

[Figure B, available on ftp ftp.cea.fr, see connection protocol below]

ACPD

5, S729–S736, 2005

Figure B : Monthly OH concentrations for the four regions of the standard inversion I-09 (in 105 cm-3). Prior estimate from MOZART model (dark dotted line) is plotted with optimized model for the standard case I-09 (dark solid line) and for the ś15% case (red solid line).

P1684 - l10 : We chose to invert monthly observations mainly because monthly sources and sinks were optimized. Since Peylin et al. (2002), we assume that there must be a consistency in the time scale of the observations and the time scale of the sources & sinks that are solved. For instance, optimizing monthly CO2 fluxes against annual observations has been shown to produce results that are more sensitive to inversion set up than time-consistent inversion (Peylin et al., 2002). Solving for weekly or daily OH variations being unrealistic for MCF inversions, we decided to aggregate observations on a monthly basis.

P1686 - l17 : We will add a sentence to quote your point about fast NH/SH transport in §3.8 section, where N/S ratio is discussed.

p1688 - l1 : modifications will be made in the text. “Linear” is in fact “linearized”

p1690 - l6 : We did not test the impact of using mean transport response functions for the 1980s but we did test the impact of using one specific year of transport over the whole period. We did this test with all years between 1990 and 1999, showing that the meteorology of year 1992 was producing the largest changes in optimized OH compared to standard case (see §3.7, p1699, l14-25). The optimized Chi-Squared is minimum when using inter-annual meteorology compared to recycled one. We do not think that using climatological response function would deeply modify the results but we agree that the best thin to do would be to run the 1980s source function, which is “just” a matter of computing time.

P1692, Figure1, : required modification will be done

P1694 : NWR was not sampled at the surface of the model but at the model level

ACPD

5, S729–S736, 2005

corresponding to its altitude. However, this procedure is not fully correct because the model topography is underestimating the real topography, which leads to extract NWR a bit too high in altitude. This is probably the main reason why the optimized model still understimates the NWR concentrations. L24 : We will precise that this is for the 1990-92 period and will refer to the new figure showing the residuals (see answer to Dr Prinn).

P1695 : We agree that the reduction of 1.1% of the integrated source is significant. Indeed, we had yearly total MCF emissions as an additional constrain (see §2.6, p1693) but not the integrated total (1979-2000). This choice has an impact on the sensitivity study on OH prior error made in section 4 because nudging the integrated MCF total emissions to the a priori value would shift upward the optimized minus prior MCF emissions of figure 10. Thus, prior errors smaller than ś15% on OH would be compatible with inventory uncertainties. However, we chose not to add another additional constrain, having difficulties to set up an objective uncertainty on this integrated MCF total emission.

P1697 : the word “obviously” will be added.

P1699: The use of recycled meteorology produces significantly higher residuals. This sentence will be added in the text.

P1702 - l8 : see principal point (1)

References

Engelen, R.J., A.S. Denning, K.R. Gurney, and T. Modelers, On error estimation in atmospheric CO2 inversions, J. Geophys. Res.-atmos., 107 (D22), -, 2002.

Kaminski, T., P.J. Rayner, M. Heimann, and I.G. Enting, On aggregation errors in atmospheric transport inversions, J. Geophys. Res.-atmos., 106 (D5), 4703-4715, 2001.

Krol, M., and J. Lelieveld, Can the variability in tropospheric OH be deduced from measurements of 1,1,1-trichloroethane (methyl chloroform)?, J. Geophys. Res.-atmos.,

ACPD

5, S729–S736, 2005

108 (D3), 4125, 2003.

McCulloch, A., and P.M. Midgley, The history of methyl chloroform emissions: 1951-2000, Atmos. Environ., 35 (31), 5311-5319, 2001.

Peylin P., P. Peylin, P. J. Rayner, P. Bousquet, C. Carouge, F. Hourdin, P. Heinrich, P. Ciais, AEROCARB contributors, Daily CO2 flux estimates over Europe from continuous atmospheric measurements: 1, inverse methodology, Atmospheric Chemistry and Physics Discussions, 5, 1647-1678, 2005.

Peylin, P., D. Baker, J. Sarmiento, P. Ciais, and P. Bousquet, Influence of transport uncertainty on annual mean and seasonal inversions of atmospheric CO2 data, J. Geophys. Res.-atmos., 107 (D19), -, 2002.

Peylin, P., P. Bousquet, and P. Ciais, Inverse modeling of atmospheric carbon dioxide fluxes - Response (vol 294, pg U1, 2001), Science, 294 (5550), 2292-2292, 2001.

Prinn, R.G., J. Huang, R.F. Weiss, D.M. Cunnold, P.J. Fraser, P.G. Simmonds, A. McCulloch, C. Harth, P. Salameh, S. O'Doherty, R.H.J. Wang, L. Porter, and B.R. Miller, Evidence for substantial variations of atmospheric hydroxyl radicals in the past two decades (vol 292, pg 1882, 2001), Science, 293 (5532), 1048-1048, 2001.

Rodenbeck, C., S. Houweling, M. Gloor, and M. Heimann, CO2 flux history 1982-2001 inferred from atmospheric data using a global inversion of atmospheric transport, Atmos Chem Phys, 3, 1919-1964, 2003.

##

Connection protocol top get figure B :

ftp ftp.cea.fr

login : anonymous

pass : email address

ACPD

5, S729–S736, 2005

cd incoming/y2k01/ACPD

get OH_seas.pdf

##

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