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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.

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We took notice of the relevant comments of Dr Prinn about our work. In the following, we answer point by point to his concerns. The updated version of the paper will take into account most of them and will clarify some hypothesis and limitations of our study.

Principal comments

(1) Calculation of pulse response functions We do not neglect the destruction of MCF by OH after 1-month pulse. We just consider changes in MCF concentrations as a linear combination of one-month length sources and one-month length sinks (one-year length for stratospheric loss). Actually, as explained in section 2 (see equation 5 and

associated text), sources and sinks are solved independently from each other after linearising the mass conservation equation. This means that each month of the inversion, pulse sources and pulse OH sinks are run independently. For pulse source, no chemical decay is applied and for pulse OH sink, no source is applied. Accounting for OH sink while solving for the source (resp. for the source while solving for OH sink) would lead to a double counting of the sink (resp. of the source). We will rewrite part of the Appendix C to clarify this point.

(2) Observational uncertainties We agree that part of the representation error is already included in the standard deviations provided with monthly MCF observations, as too coarse resolution or uncertain transport. However, there are other causes of representation error such as data selection : observations are selected to remove polluted events which is not the case for the LMDZ model in our study, as no selection criteria is applied to model outputs, mostly because we did not use inter-annual meteorology before 1990 (see also point 6 for further discussion on this issue).

According to Tarantola (1987, p211 : §Are the residuals too large ?), Calculation of the chi-squared statistics is an objective value to check whether too much confidence is given to the observations, given the freedom defined for the parameter space (P matrix). For MCF, the parameter space is rather tightly constrained by McCulloch & Lidgley uncertainties.

Chi-squared value divided by the cost function at the minimum should be close to one. A too large value of the chi-squared means that “some violation of the hypothesis is to be feared” (Tarantola, 1987). Directly using the standard deviation provided with the monthly means, leads to chi squared larger than 1 in our inverse configuration with the smaller number of degrees of freedom (I-14). Thus we applied a scaling of the observation uncertainties to get a chi-squared close to 1 in this case. The sensitivity test in which observations uncertainties are multiplied by 0.7 (I-08), roughly corresponds to the original standard deviations. It leads to changes in OH variability of less than 5%, as noticed in the text (p1700, l9-10). We will add a sentence p1700 and also p1692

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about this issue. We will also be more precise on our hypothesis (no data selection, see point 6 below).

(3) Oregon/California data We propose to add the California data for the 1995-2000 period in the sensitivity test in which Oregon data are used (I-15). As when using CMO data only, I-15 produces the smallest OH variability of our tests with ś7.2% changes over 1980-2000 compared to ś8.5% for the mean inversion (-15% reduction in variability), but only small changes in the phasing and the trend of the OH variations (Fig. 7). We will modify section §3.7 to quote TRH impact, and also all tables and figures linked to this modification. However, we do not want to use this couple of stations in our standard case because there is a lack of data of 6 years, which is a too long period to interpolate data as for MHD or SMO (Thoning et al., 1989, see point 6 below). This interruption of data could create artificial variability when stations appear and disappear, as shown by Rodenbeck et al. (2003) for CO2. Anyhow, including or not CMO/TRH data in the standard case would not change the main conclusions of the paper that are 1/ substantial OH variability is found when letting OH totally free to change and 2/ OH variability is largely reduced when letting MCF emissions varying within ś2σ bounds.

(4) European & US Emission after 2000 When we wrote this paper, AGAGE MCF observations were not available after 1999. Thus consistently commenting for residuals emissions of MCF in the early 2000s is not easy. However, as it is an important issue, we did one inversion after 1999 using NOAA/CMDL network. As mentioned in the text (p1695, l20-), giving more freedom to MCF source after 1997, leads to an increase of the emissions of 3Gg over Europe, 4Gg over Asia and 6Gg over North America. For the 2000-2002 period, optimized MCF emissions become 7.1ś2.6 Gg/yr over Europe, 7.8ś2.8 Gg/yr over Asia and 11.8ś3.8 Gg/yr for all North & central America. For the 2000-2002 period, optimized MCF emissions are found to be 7.1ś2.6 Gg/an over Europe, 7.8ś2.8 Gg/an over Asia and 11.8ś3.8 Gg for all North and Central America. For Europe, these emissions stand between the large emissions inferred by Krol et al. (20

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Gg/yr) and the small emissions inferred by Reimann et al. (0.3-3.4 Gg/yr). There is no agreement so far on European residual MCF emissions inferred from atmospheric observations. For North America, Millet et al. (2004) find US emissions of 5.1 Gg in 2001 and 3.7 Gg in 2002 based on MCF data analysis, which is about twice the Li et al (2005) estimates (2.2Gg/yr for the 2001-2002 period) based on TRH measurements. Our estimate for North and Central America MCF emissions (11.8ś3.8 Gg), even if covering a larger area than US territory only, appears to be much larger. One possible reason could be an underestimation of other emissions (North Asia and Southern Hemisphere) as out global MCF inferred emissions of around 20ś5.5 Gg+/yr is more constrained by the inversion than the regional estimates, because of the small number of MCF stations. It would be interesting to include recent AGAGE data in our inversion, when available, to see their impact on European and North American MCF emissions. This comment will be added to the text in section §3.2, p1695.

(5) Sources & sinks error correlations This is a very good point. The LMDZ model is basically a GCM nudged on ECMWF analysis for u and v. It has been used by a variety of groups for climate and chemistry studies. For atmospheric chemistry, validations can be found in Hauglustaine et al., 2004. LMDZ is known to have a fast inter-hemispheric mixing (about 1 yr), in the lower range of TRANSCOM inter-comparison project (Law et al., 1996). Transport in the PBL is rather diffusive and thus limits so far the ability of the model to reproduce all the sub-daily variability of tracer concentrations for land sites located close to sources (especially night-time accumulations at the surface). This limitation is common to most of the global CTM. This issue does not concern MCF stations that are mainly remote or coastal. The convection scheme is from Tiedke et al. (1989), which is widely used in the atmospheric transport community. So we are rather confident that the vertical transport in the model is not too slow (see also Hauglustaine et al. 2004, Hourdin et al., 1999).

However, we agree that the fact that annual errors on optimized sources and errors on optimized sinks appear to be uncorrelated is unexpected. We investigated further

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this issue by calculating the full variance/covariance matrix, on a monthly basis (Pa matrix in appendix B). We do find small error correlations above 0.2 (in absolute values) between sources and sinks on a monthly basis (e.g. European source correlated with Northern Hemisphere North OH sink), but only for some summer months of some years. When calculating error correlations on a yearly basis, the overall correlation is at largely reduced (maximum -0.1 to -0.15) because most of the months give small error correlations. Thus they do not appear in table 6.

Moreover, the highest monthly error correlations between sources and sinks (around -0.25) stands between MCF regions (mostly Europe) for some summer months and OH sink for the current and the previous months (0 to 3 months before). Such a time lag can also be observed in table 2, in which the "MONTH" column gives the month of the maximum of the response function at one station. One can observe, for instance, that for MHD, the impact of the MCF emissions is maximum during the month of emission, whereas the impact of NHN OH sink is maximum 2 months after the OH pulse (in the low-to-mid troposphere). The situation is more equilibrated in the tropics where MCF emissions and OH sink have a maximum impact at about the same number of months after the pulse occur. This can be explained by two opposed factors : the faster vertical transport in the tropics and the larger distance of surface stations to MCF sources in the tropics compared to northern mid-latitude stations. In other words, errors on MCF sources and sinks are more correlated in the tropics and/or for summer months, consistently with what can be expected from transport features. But the overall, yearly error correlations between sources and sinks remain smaller than 0.2 in absolute value (-0.2 in reality), because response functions are significantly shifted in time and in shape. We propose to develop this comment in §3.6, p1698.

(6) Large model-observation residuals This is also a very good point. In their 2D model, Prinn et al. (2001, 2005) seem to get smaller residuals (observation minus optimized modelled concentrations) than ours. As suggested, we propose to include a new figure after the fit to observations (Fig. 2) with residuals at the four GAGE/AGAGE sites

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of our standard case (as in Prinn et al., 2001 and Krol & Lelieveld, 2003) and the associated text below (the figure cannot be put online but will be provided on request to pbousquet@cea.fr) :

————— START Figure A below plots observed minus optimized concentrations on a monthly basis (as in Krol et Lelieveld, 2003, Fig. 5) together with a 12-month running mean of the diferrence (as in Prinn et al., 2001, Fig. 4). For most stations, monthly residuals usually remain below ś5 ppt, whereas smoothed residuals stay within ś3 ppt. At MHD, large mismatch is observed in 1990-92, when MCF concentrations are maximum). Also, no data were available at MHD during the 1984-86 period. Thus, available measurements before and after this period were interpolated using Thoning et al. (1989) procedure. These concentrations were associated with much larger uncertainties, producing a poor fit of the optimized model. The same issue occurs at SMO in 1989-91. Residuals at RPB and SMO show apositive long-term mean in the 1980s, balanced by negative mean residuals at CGO during the same period. This feature possibly indicates a too fast inter-hemispheric mixing that leads to an underestimation of the modelled concentrations in the northern hemisphere and to an overestimation of the modelled concentrations in the southern hemisphere. Such “bias” in the residuals is also visible in Krol & Lelieveld (2003, Fig. 5) at RPB or CMO. It is reduced from the mid 1990s when the difference in MCF content between the northern hemisphere and the southern hemisphere becomes very small. Another hypothesis to explain this bias is the fact that we do not use inter-annual meteorology for transport in the 1980s, as mentioned in the text (p1690; l5). The inversion does not completely correct for this because of substantial observational errors. Reducing observational error, would reduce this bias but at the prize of a correcting a possible transport problem by additional OH variations.

As mentioned before, no selection criteria is applied to model outputs, mostly because we did not use inter-annual meteorological forcings before 1990. In 3D inversions, this could be critical point. It is still widely discussed between modellers and experimental-

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ists. In order to test the influence of this limitation on OH variability, we did a sensitivity test at MHD by using data with or without polluted events. We chose MHD because this is the station where polluted data and non-polluted data are the most different. The change in the fit is significant (Fig. A): the optimized model is overestimating the variability when using MHD non-polluted data (especially in 1990-92) and is largely underestimating it when using polluted MHD data (especially in the 1980s, when no inter-annual meteorology is used). However, as can be seen in Fig. 7 of the paper, the impact on OH variability is rather small which means that the issue of data selection is not critical here, just increasing a bit residuals of fig. XX. The residuals that are obtained in this study could be reduced by using inter-annual meteorology for the whole period and by selecting model outputs properly. ——END

It can be surprising that a 3D model still produces rather large residuals (but not that much larger than previous studies). First, each month, 4 OH and 4 MCF scaling factors are optimized against 4 to 10 observations. OH factors are rather free to adjust (ś100% error). However, MCF factors are rather tightly constrained by McCulloch & Midgley inventory uncertainties (ś2 to ś25%). Thus the number of degrees of freedom is mostly reduced to the 4 OH scalors, which is not sufficient to get a perfect fit to the observations, especially in the mid latitudes of the northern hemisphere. In other word, we do not have that many degrees of freedom. Second, Prinn et al. (2001, Fig 4) does not show residuals for each of the three individual northern stations (MHD, CMO and TRH) separately but only the aggregation of the 3 sites (they use a 2D model), which probably smoothes the differences compared to our 3D approach. One can observe monthly differences of the order of 8-10 ppt at MHD in 1980 and 1990 (for instance) between optimized model and MHD observations, on fig. 1 of Prinn et al. (2001). Third, shifting from 2D to 3D modelling and from annual optimisation to monthly optimisation, should imply more realism but also makes the model more sensitive to transport errors and data selection issue for instance. In Krol & Lelieveld (2003), residuals of the same order of magnitude than in our study are found using a 3D model (see their figure 5). Also, as we do not properly select model outputs as observations, we probably gen-

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erate additional residuals, especially at MHD. Finally, the fast north to south transport in LMDZ may create a bias that increases the residuals on the long-term (see above). Overall, the different sensitivity tests performed in this study showed that the amplitude and the phasing of the optimized OH were not deeply modified when changing the network or the uncertainties or the non selected data, as long as OH prior uncertainties are kept loose at ś100% (Fig. 5). The main changes occur when uncertainties on OH variations are reduced to get closer to MCF uncertainties.

[FIGURE A available on ftp.cea.fr, see connection protocol at the end]

Figure A : MCF residuals : Differences between MCF observations and optimized MCF concentrations : 12-month running mean (solid lines) and monthly differences (dotted and dashed lines) at 4 ALE/GAGE/AGAGE stations (in ppt). Observational uncertainties are in red. “Chi” stands for the contribution of the station to the Chi-squared statistics (in %). For Mace Head, purple lines represent the residuals that are obtained when polluted data are used at MHD (inversion I-11).

Other comments

P1681 - l2-3 : we will add the spatial issue in the text.

P1681, p1682 : references will be changed

P1682 - l26-27 : the reference to the lack of OH spatial variations in Prinn et al. and Krol et al. wil be removed.

P1683 - l2 : point 4 refers to §2.4 about inverse procedure.

P1684 - l15/16. We will remove the sentence and add TRH in I-15 (see principal comment 3)

P1685 - l15-16: reference will be added

P1687 - l1-2 : lifetime of MCF against stratospheric loss is not calculated directly by our 3D model as we do not save tropospheric-stratopsheric MCF flux. We will try to

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re-run the CTM to save this MCF flux and provide in the lifetime in §3.5

P1688 - l12 : we will add the sentence : We do not optimize the reaction rate, kx,t. This is a limitation as Prinn et al. (2001) showed through a monte carlo experiment that constant rate was the main contributor to the error in mean OH concentrations.

P1689 - l11-12 : we will add the sentence : The optimization relies on a classical Bayesian formulation where one minimizes a quadratic cost function J(α) between modeled and observed concentrations from one hand, and prior estimates and optimized estimates from the other hand, each term being weighted by variance/covariance matrices.

P1693 - l15-16 : yes, all errors are 1-sigma in the paper

P1694 - l22-24 : J cost function is global. Optimizing separated cost function would lead to an inconsistent picture of MCF cycle between hemisphere. One can think of the north-to-south gradient. However, it is true that if one station has a too large contribution to the Chi-squared (that is about twice the cost function at the minimum), then it would have a non realistic impact on some remote sources and sinks. This is why we check the contribution to the Chi-squared.

P1694 - l25 : we will add the Prinn et al. (2000) reference. As mentioned in principal comments (2) and (6), we do not select our model at MHD but included a sensitivity test with and without the polluted data. As suggested by Dr Prinn, LMDZ catches well the phase of these events but underestimates their amplitude. As we use the adjoint of LMDZ model to calculate source response function, a data selection procedure is not as simple to set up as in classical forward modelling. The sensitivity test showed that it has only a small influence on the inferred OH variability.

P1696 - l25-26 : We agree with your argument but our purpose with the no-ocean case was only to estimate what would be the maximum impact of ocean sink. We propose to add the following sentence : “This crude sensitivity test could be refined by testing

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another ocean sink representation such as Yvon-Lewis & Butler, (2002) as in Prinn et al (2005). ”

P1698 - l8-11 : We will refer to Prinn et al 2005 for stratospheric loss.

P1701 - l20 : reference PR will be added

P1704 : l9-12 : By reliable, we meant “more robust”. We would prefer to change reliable into “more robust”.

P1708 : you are right, a bracket is missing in the formula

P1708 - p1709 : see principal comment (1)

P1714 : co authors will be added. P1715 - table 1 : we will add KR and PR values in table 1, and add a comment in section 3.8

P1722 - fig 1 : we will add TRH in the text

p1725 - Fig. 4 : OH response functions are expressed in ppt, as mention on the Y axis and in the caption of the figure. It represents the impact on MCF concentrations of the OH sink at RPB for the different months of one year (Fig 4a) or for the different years at the same month (Fig. 4b). OH response functions are positive because the monthly scaling factors that are optimized for OH are negative. Caption will be clarified

P1727 - Fig 6 : caption will be modified.

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