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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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General Comments

This very interesting paper uses an inverse method with a 3D model and methyl chloroform (MC) measurements and concludes that substantial year-to-year variations occur in global-average OH concentrations (the principal MC sink) between 1980 and 2000. This conclusion was previously reached by Prinn et al (2001), but subsequently challenged by Krol and Lelieveld (2003) arguing that these variations are caused by model shortcomings and that models need, in particular, to include observationally-

based, interannually-varying meteorology to provide accurate annual OH estimates. However, this latest paper, which uses observationally-based meteorology and estimates OH on monthly time scales, yields interannual OH variations that agree remarkably well with the Prinn et al (2001) and equivalent Krol et al (2003) estimates. But neither the 2D Prinn et al (2001) or the 3D Krol et al (2003) inversion models used interannually-varying circulation. This implies that these interannual OH variations are in fact real, and from the authors sensitivity studies, their phasing in particular appears robust. However, the uncertainties in the amplitudes of the annual values from both this present paper and Prinn et al (2001) are still significant. A second important aspect of this paper is its attempt to estimate both sources and sinks at the same time. This certainly requires a very accurate and realistic high resolution circulation model since the surface sources and OH sinks are separated by only about 5 kilometers vertically. The problem is apparently made more tractable by their use of stringent prior emission error estimates, and they provide further evidence of unexpected MC emissions in recent years. Parenthetically, Prinn et al (2005) have recently shown that the substantial estimated OH variability remains even after accounting for these residual emissions, and that OH has returned by 2004 to near its 1979 levels. If the major conclusions of Bousquet et al are correct, it is an important contribution. However, there are a significant number of questions that need to be answered to better establish their conclusions and these questions are given in the following pages.

Principal Comments

(1) Calculation of source pulse responses (p1690, l 3-15; p1708, l 17-21; Appendix C). The procedure of neglecting any perturbation to chemical destruction after a 1-month (OH) or 1-year (photolysis) perturbation (or pulse) appears correct. But it appears that the 1-month source pulses also ignore chemical destruction after that month. Surely the effect of the source pulse on the global atmosphere after that month should decrease exponentially with about a 4.9 year e-folding lifetime due to the net effect of OH, photolysis and the ocean sink? In other words MC is not to be treated as a passive

ACPD
5, S515–S523, 2005

tracer in this case which the text implies.

(2) Observational uncertainties (p1692, l 6-28 and p1693, l 1-10). The authors use the standard deviations of the monthly means and then further augment them to define the total MC observational error. The use of these standard deviations (as opposed to the much smaller instrumental precision) in this way was justified by Prinn et al (2001, 2005) to account for the inability of the point measurements at the stations to exactly measure the large volume averages needed to compare with their 2D model. Specifically over the period of one month the multiple synoptic meteorology cycles enable the station to sample air from a wide variety of locations (and thus define the MC variability) in the above model grid volume. Thus this procedure already accounts for many of the model "representation" error sources (too coarse resolution, uncertain circulation, etc.) especially given the much higher resolution of the authors 3D model. Thus the further augmentation of these standard deviations used by the authors is overdoing it (it would be more reasonable if their procedure was used to augment instrument precisions only).

(3) Neglect of Oregon/California data (p1684, l 10-21 and p1699, l 6-10). Why are the AGAGE data from the Trinidad Head CA station (1995-present) ignored? Since the authors' sensitivity tests showed a significant impact when Cape Meares OR data (1979-1989) are included, then the same is probably true for Trinidad Head. The optimal solution should use both the OR and CA data.

(4) European and U.S. emissions (p1695, l 20-29). Two recent papers that estimate emissions from high frequency AGAGE and other data disagree with the two papers quoted here. First, Reimann et al (2005) use multi-year Mace Head, Ireland and Jungfraujoch, Switzerland measurements and estimate 2000-2003 European emissions of 1.8 (0.3-3.4) Gg/year or an order of magnitude less than those estimated by Krol et al (2003). Second, Li et al (2005) use multi-year Trinidad Head CA measurements, and extrapolations of estimates by Barnes et al (2003) using Harvard Forest MA observations, to estimate 2001-2002 USA emissions of 2.2 Gg/year (or about half of

ACPD

5, S515–S523, 2005

the Millet and Goldstein (2004) estimates). Are emission estimates in this paper more or less consistent with these more recent lower estimates (i.e. what are the uncertainties in your 2000-2002 global and regional emission estimates)? Also, what would you conclude about recent residual European, Asian, and USA emissions if you include the omitted Trinidad Head CA data?

(5) Source and sink error correlations (p1698, l 21-25 and p170, Table 6). It is very surprising that there are no significant error correlations between the estimated MCF surface fluxes and atmospheric sinks (OH). This surprise is because the authors use monthly mean observations and on these time scales the mixing between the surface (flux locations) and mid-troposphere (OH maxima) is quite efficient at least in the real atmosphere. Are the authors confident that vertical mixing out of their model boundary layer and into the mid-troposphere by moist convection and other subgrid-scale processes is realistic? If their model vertical mixing is too slow, then it would lead to the false conclusion that sources and sinks are uncorrelated.

(6) Large model-observation residuals (section 3.1 and Fig. 2). Given the large number of state vector elements that were optimized, the large residuals between the observations and their optimized model values found by the authors (Fig. 2) are surprising. These can be compared to the much smaller residuals found by Prinn et al (2001, Fig. 4; 2005, Fig. A3 in Auxiliary Material). Is this indicative of a problem in the formulation of their cost function J? Specifically is too much weight given to the second term in J (involving state vector errors in P)? Referring to my point (2) above, have the authors overestimated R? Or underestimated P? Also, the residuals should be plotted (rather than the totals) in Fig. 2 to clarify their behavior.

Other Comments

p1681, l 2-3: the problem with current direct in situ or remote sensing measurements is not their inability to address the OH variation time-scales as stated here, but rather their inability to cover the regional to global spatial scales of OH variability that largely

ACPD

5, S515–S523, 2005

determine OH destruction rates for long-lived trace gases.

p1681, l 26 and p1682, l 11, 23, 29: the relevant paper is Krol & Lelieveld (2003) not Krol et al (2003).

p1681, l 26-27: neither Prinn et al (2001) or Krol & Lelieveld (2003) ignored spatial variations of OH fields (both papers estimated hemispheric as well as global OH).

p1683, l 2: what is the “point 4” referred to here?

p1684, l 15-16: The Oregon/California AGAGE stations did not “stop in 1989”. The Oregon (Cape Meares) station began in 1979 and indeed ceased in 1989, but the California (Trinidad Head) station then replaced it beginning in 1995 through to the present (see also principal comment on this above).

p1685, l 9-12: reference Prinn et al (2001) for these observed MCF variations.

p1687, l 1-2: please provide the lifetime of MCF due to stratospheric photolysis (e.g. total global MCF amount divided by total stratospheric destruction rate) and compare to that used by others (e.g. 39 years by Prinn et al, 2001).

p1688, l 12: the text should note that the uncertainty in the reaction rate constant that is neglected here, was included in the 10,000-member Monte Carlo error analysis by Prinn et al (2001, 2005). Rate constant error was the largest contributor to the error in the estimated average OH concentrations (whereas emission errors dominated uncertainty in OH trends).

p1689, l 11-12: this is not a complete description of the cost function used since it also minimizes the quadratic posterior-prior state vector difference. And for both terms the quadratic differences are weighted (by R and P respectively).

p1693, l 15-16: are these errors (and all others in the paper) 1 sigma?

p1694, l 22-24: is the cost function J computed separately for each semi-hemisphere or hemisphere, or is it global? If it is global, then could e.g. northern hemisphere

ACPD

5, S515–S523, 2005

stations unrealistically influence southern hemisphere OH estimations?

p1694, l 25 and p1695, l 1: the text should reference Prinn et al (2000) for the method used to actually remove the polluted data in computing the monthly means used by the authors. Presuming the authors model simulates at least the phasing of these pollution events correctly, then their model outputs during these events should also be removed in computing the model monthly averages. Has this been done?

p1696, l 25-26: the "no-ocean-sink" case is not credible. The real argument is between an oceanic sink without an effective long-term polar-ocean storage (Yvon-Lewis and Butler, 2002), and one with it (Wennberg et al, 2004). Prinn et al (2005) tried both and found that the inferred interannual OH variations were present using either formulation, but inferred OH was lower in the pre-1992 years and higher after that using the Wennberg et al (2004) formulation.

p1698, l 8-11: the 2D model used by Prinn et al (2001, 2005 and prior papers) has always shown these expected changes in the stratospheric sink over time (see Prinn et al, 2005, Auxiliary Material, Section 3 and Figure A1), contrary to the claim that it does not (Krol and Lelieveld, 2003).

p1701, l 20: text should note that PR also considered the geographic equator and obtained essentially the same N/S OH ratio as this paper

p1703, l 2-4: it should be emphasized that allowing a 2-s error criterion (i.e. probabilities as low as 1 in 20) is very unlikely according to the McCulloch and Midgley (2001) analysis.

p1704, l 9-12: what is meant here by "reliable"? The authors' error analysis presumably provides a reliable estimate of the uncertainties, and the Monte Carlo error analyses in Prinn et al (2001, 2005) reach the same qualitative conclusion as shown by the significant error bars accompanying their estimates. I suggest replacing "reliable" with "more accurate".

ACPD

5, S515–S523, 2005

p1708, l 2: should there also be a factor of 1/2 in front of the second term of this cost function?

p1708, l 17-24 and p1709, l 1-4: as noted in my "major comments" earlier, this extrapolation procedure appears to be wrong. The MCF pulse will decay with an e-folding time of about 4.9 years and never reach an asymptote.

p1714, l 2: add the missing co-authors to the Prinn et al (2001) paper.

p1715, Table 1: the NH, SH and NH/SH values given here should be compared to those in Krol and Lelieveld (2003) and Prinn et al (2001).

p1722, Fig. 1: why is the Trinidad Head, CA AGAGE data omitted (see earlier "major comment").

p1725, Fig. 4: what are the apparently dimensionless units for OH in the response functions in graphs (a) and (b) (percentages or fractions relative to a reference OH?).

p1727, Fig. 6: vertical axis should be "semi-hemispheric" not "global"?

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ACPD

5, S515–S523, 2005

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