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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Multi-species inversion of CH_4, CO and H_2 emissions from surface measurements" *by* I. Pison et al.

I. Pison et al.

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We thank the reviewer for his/her comments. We addressed them all. Our response appears inserted within his/her text: the comments of the reviewer are in black and answers in bold.

We are very aware of the importance and of the difficulties to maintain a measurement network (LSCE/RAMCES is an experimental team of our lab). They were all contacted by e-mail to get an agreement on the use of their 2004 data before we submitted the paper to ACPD, as requested on the WDCGG website. They agreed to be acknowledged at the end of the paper. However, we forgot to include the individual references in the text. We apologize for this and we will add the references in the revised version of the paper.

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General comments

1. The multiple species inversion technique is original (yet not new see Müller and Stavrakou [2005]) yet critical pieces of information are missing in the manuscript and more importantly in the current state the scientific significance of the study is not clear. **We will add references in the introduction: "[. . .] whereas they are linked through chemical reactions and transport. At the mesoscale in the framework of air quality, Elbern et al. (2007) have studied the feasibility of the inversion of emissions of sulphur dioxide and ozone precursors. At a global scale, Stavrakou and Müller (2006) have inverted CO emissions taking into account their relation to the non-methanic volatil organic compounds (NMVOCs) through OH. In this work, we describe an inversion system that optimizes the four main reactive species of the methane oxidation chain [. . .]". Our approach is somehow original because instead of developing the complex and computationally expensive adjoint of a full chemistry model, we chose to start from a simplified chemistry model (with only the CH_4 oxidation chain) so as to be able to handle long inversions and large systems. We will add information as detailed in the answers to the specific comments. We will also re-write parts of the conclusion to make it clearer (see answer to General Comment 6 and Specific Comments F).**

2. The Pison et al. submitted manuscript does not cite adequate references and does not give proper credit in the main text to the people and institutions doing the measurements, especially the in-situ monitoring networks (AGAGE, CSIRO, JMA/MRI, LSCE, NIWA, NOAA ESRL). The whole study relies almost exclusively on these measurements and it is clear that without such long term commitments from various national laboratories to globally distributed observations, such studies would be impossible. The fact that the observational data are easily available has led many scientists and non-scientists to take these time series for granted. The truth is that keeping such networks going and improving them is an every day dedication and challenge that current

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and future research relies on heavily. Observations, just like model, outputs need to be described and referenced properly for the reader to understand the specifics of a particular dataset. The authors have acknowledged the problem and should provide the relevant information in the revised manuscript.

We will add references in the description of the measurements: " The observations used in this study are concentration measurements for the year 2004, performed at surface stations for MCF (Montzka et al., 2000), CH_4 (Dlugokencky et al., 1994; Cunnold et al., 2002; Worthy et al., 1998), H_2 (Novelli et al., 1999) and CO (Novelli et al., 1992) from various networks (NOAA: Conway et al., 1994, CSIRO: Francey et al., 1999; AGAGE: Prinn et al., 2000; NIWA: Lowe et al., 1991; JMA/MRI: Matsueda et al., 2004) available on the World Data Centre for Greenhouse Gases (WDCGG) site at http://gaw.kishou.go.jp/wdcgg/. We used instantaneous measurements and data that are 24-hour averages (named 'event' and 'daily' data in the database). Continuous measurements by the AGAGE network were averaged over 24 hours and these daily means were used. We added observations available from the RAMCES (Reseau Atmospherique de Mesure des Composés à Effet de Serre) network coordinated by LSCE (Schmidt et al., 2006)."

3. I did not find in the text a correct description of the a priori emissions used for the inversion. The specific year studied, 2004, is not covered by the EDGAR 3.2 inventory which is for 1990 and 1995. The QUANTIFY emissions used for CO on road and shipping emissions have no citation.

The description of emissions was not clear enough. The paragraph on emissions will be re-written to be clearer and Table 1 will be removed: "Two main datasets are combined to build the prior inventory:

- **version 3 of the Emission Database for Global Atmospheric Research (EDGAR 3) inventory for the year 1995 for anthropogenic emissions (Olivier and Berdowski, 2001)**

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- **version 2 of the Global Fire and Emission database (GFED-v2) (Van der Werf et al., 2006) for monthly emissions due to biomass burning for the year 2004.**

In addition, other sources are taken into account:

- **emissions of CH_4 due to wetlands and termites are based on the study by Fung et al. (1991)**
- **the biogenic emissions of VOCs are provided by the ORCHIDEE vegetation model (Lathière et al., 2005).**
- **the biogenic emissions of H_2 are based on the study by Hauglustaine and Ehhalt (2002)**
- **an oceanic source of CO, equivalent to $\approx$20 Tg per year, is considered.**

For MCF emissions, the inventory by Montzka et al. (2000) is rescaled according to an update of the study by Bousquet et al. (2006). Note that for the other species no effort is made here to adapt the 1995 EDGAR 3 inventory to the year 2004. Since global economic growth occurred since 1995 and induced a modifications of the emissions (Ohara et al., 2007), in particular in Asia, the results of the inversions performed here combine both the corrections on the bottom-up emissions and the trends over the 1995-2004 period." Note that for this pixel-based inversion, the prior structure of the fluxes, although important, is not as critical as in the "larger region" approach: the large prior errors allow the inversion to correct almost independently each pixel. The large increase over Asia is a good example of this. We all wait for EDGAR 4 that should come up in the next weeks to update prior spatial patterns anyway.
Where does the MCF emissions estimate for 2004 come from?

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As stated in the above answer, the inventory of MCF emissions by Montzka et al., 2000 was rescaled according to an update of Bousquet et al. (2006) inversions.
The uncertainties on the prior emissions are not described properly. Only the correlation lengths used are mentioned at line 12 p 20694.
We will add this information after the general reference to Chevallier et al. (2005): "In this study, the errors are then set to 100% of the maximal flux over the inversion period for CO and CH_4, 100% of the flux for H_2, 1% of the flux for MCF (in order to constrain OH), 100% for HCHO concentrations, 10% for OH concentrations, 10% for the initial concentrations of HCHO and MCF and only 3% for CH_4 and 5% for CO. Temporal correlations are neglected."

4. The authors do not compare their a posteriori emissions with comparable published work. Please check the litterature. The discussion only presents regional relative differences compared to the prior fluxes (except in Table 4 for CH4 fluxes) which makes it impossible to compare with other studies. The numbers on Figures 5a and 5b are unreadable and are not meaningful since the a priori values are not given anywhere in the manuscript. How does the reader evaluate the results and the significance of the work when the regional results are presented with such a narrow scope?
The comparison made here in Table 4 is between the variational system and the analytical system of Bousquet et al. (2006) with the same areas and the same time period (year 2004). The idea was to avoid avoid misleading figures due to comparing different systems on different years and space distributions. We therefore propose to add comparisons of the total emitted masses of CH_4 with Bergamaschi et al. 2007 and Meirink et al., 2008 in the paragraph 'Inversion: mass budgets': "For CH_4, these results might be compared to two studies of the year 2003. Bergamaschi et al. (2007) inverted CH_4 emissions for 2003 with the TM5 model, various prior inventories and either SCIAMACHY satellite or NOAA ESRL surface measurements. With a fixed total prior emitted mass at 507.7 Tg of CH_4, their analysis for 2003 ranges from 508.8 to 554.4 Tg. In the same framework, Meirink et al. (2008) compared synthesis and 4D-var inversions. With the

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total prior emitted mass at 507.7 Tg, their analysis ranges from 523.5 to 525.2 Tg. For CO, the study by Pétron et al. (2004) inverted emissions with a sequential scheme and MOPITT satellite data from April 2000 to March 2001. From a prior total emitted mass of 1034 Tg, their inversion ranges from 1228 to 1384 Tg."
Specific comment: Bousquet et al. [2006] does not cover the year 2004 so the reader does not know what went into the Bousquet CH4 emissions estimates in Table 4 for this particular year.
We use here an update of this study that covers up to 2007: this will be clearly stated in the text. The setup of the inversions in the update is the same as for Bousquet et al. (2006) with the transport for the proper years.

5. When comparing the modeled CO to the MOPITT satellite data, the authors need to cite at least the validation papers by Emmons et al. (and Edwards et al. work). Several (forward and inverse) studies have been done using this incredible dataset, only one is cited. The uncertainty on the observations is not uniform. The quality of the satellite data is best over the continent during the day. Do the numbers in Table 5 change if you sub-sample the data and model?
We will add references to the validation papers by Emmons et al. (2004), Emmons et al. (2007), Edwards et al. (1999) and Deeter et al. (2004). We also already cite the studies by Stavrakou and Müller (2006), Pfister et al. (2004) and Pétron et al. (2004). The satellite data has actually been sampled to use only daytime measurements as described in Chevallier et al. (2008). The difference between continents and ocean is what lead to show the two hemispheres separately. This will be clearly stated in the text.

6. Table 5 presents some simple statistics on the agreement between the a posteriori modeled methane and CO and some independent observations: for CH4, data from two European aircraft sites and for CO, the MOPITT 700hPa data. Again these datasets are not referenced properly: which labs produce the data and which articles describe the analytical method and the uncertainty on the data.

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The MOPITT 700hPa data is the same as in point 5. above. The aircraft sites are operated by a team from LSCE and the data were given to us as personnal communication (general description of te RAMCES network in Schmidt et al., 2006): we will add this information in the text. In the conclusions, the authors overstate the improvement reached after the inversion. For methane, the prior was as good and for CO there is a 'significant' improvement only in the slope, not the correlation.
We will re-write this part of the conclusion: "The comparison of our analyzed fluxes with the results obtained by another inversion system by Bousquet et al. (2006) shows that they lay in the same range. The comparison of our analyzed CH_4 concentrations with independent measurements by mobile surface stations and aircraft shows no significant improvement for the background conditions (over the oceans) and a significant improvement in the vertical profile of standard deviation in the boundary layer and in the free troposphere. The comparison of CO concentrations to independent satellite data shows that the fit between simulation and measurements is significantly better in both hemispheres after the inversion, which indicates that the inverse system is able to correct for large underestimations in the prior and obtain consistent analyzed fluxes."

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 20687, 2008.