Atmos. Chem. Phys. Discuss., 8, S11291–S11297, 2009
www.atmos-chem-phys-discuss.net/8/S11291/2009/

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.

Anonymous Referee #2

Received and published: 16 February 2009

Pison et al. present a global multiple species inversion for CH4, CO and H_2 sources as well as OH concentrations for 2004. The observational constraints come from surface concentrations of CH_4, CO, H_2 and MCF measured continuously or weekly by long-term monitoring laboratories at a global network of stations. The authors use the global transport model LMDZ [Hourdin and Armengaud, 1999] nudged to the ECMWF horizontal wind analyses and a simplified chemistry module SACS (and its adjoint model) derived from INCA [Hauglustaine et al., 2004]. The Bayesian variational inversion technique was first developed and used for a passive tracer CO_2 by Chevallier et al. [2005] and was adapted for the multi-species study.

With some more work to clarify several critical points listed below and to present the scientific value of the results in light of the state of the science, the article can become

an interesting contribution.

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.

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 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.

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. Where does the MCF emissions estimate for 2004 come from? The uncertainties on the prior emissions are not described properly. Only the correlation lengths used are mentioned at line 12 p 20694.

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 CH_4 fluxes) which makes

ACPD

8, S11291–S11297, 2009

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?

Specific comment: Bousquet et al. [2006] does not cover the year 2004 so the reader does not know what went into the Bousquet CH_4 emissions estimates in Table 4 for this particular year.

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?

6. Table 5 presents some simple statistics on the agreement between the a posteriori modeled methane and CO and some independent observations: for CH_4, 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. 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.

Specific comments

A. Abstract:

(a) The abstract should clearly state which observations are used. The model LMDZ is mentioned so should be the observational networks AGAGE, CSIRO, JMA/MRI, LSCE, NIWA, NOAA ESRL and the MOPITT CO data.

ACPD

8, S11291–S11297, 2009

Interactive Comment

A vague abstract has no purpose.

(b) The optimization of HCHO is only mentioned in the abstract. What has been learnt from doing it?

B. Section 1

(a) Site the networks and the number of stations today. The statement "Structured in international networks" is not correct. Each national lab manages its own network of regionally or globally distributed stations. GLOBALVIEW is a data product and should not be mentioned here.

(b) When mentioning the 1% accuracy for methane satellite data [Frankenberg, 2005], the authors should cite which instrument they are referring to.

(c) Replace the assimilation of "bio-geochemical data" with "atmospheric concentrations: Line 10 p 20689.

(d) Which MCF measurements do you use for OH concentrations optimization? AGAGE only?

C. Section 2

(a) Section 2.2.1 states that the adjoint of the transport model LMDZ were developed for this work, but they are already used in Chevallier et al. [2005]. Has the model changed since then?

(b) Plots in Figure 2 show global mean differences?

(c) Section 2.3: How do you build your prior emissions for 2004? EDGAR 3.2 is for 1990 and 1995. Table 1 should be redone. The QUANTIFY inventory for CO should be in the Anthropogenic emissions with a reference. The biomass burning is cited in 2 different ways: first GFED then van der Werf.... Both need to be there "GFED-v2 for 2004 [van der Werf et al., 2006]". What is the magnitude of the uncertainty on the a priori fluxes?

ACPD

8, S11291–S11297, 2009

Interactive Comment

(d) Section 2.4: See General Comments: the authors should describe and reference the observational data they use. I strongly suggest redoing Figure 3 to show the various networks in different colors.

(e) How do you invert for daily averages in your set-up?

(f) Replace NOAA04 scale with "NOAA 2004 calibration scale [Dlugokencky et al., 2005]".

(g) Reference the product GLOBALVIEW with GLOBALVIEW-CH4: Cooperative Atmospheric Data Integration Project - Methane. CD-ROM, NOAA ESRL, Boulder, Colorado [Also available on Internet via anonymous FTP to ftp.cmdl.noaa.gov, Path: ccg/ch4/GLOBALVIEW], 2008.
And
GLOBALVIEW-CO: Cooperative Atmospheric Data Integration Project - Carbon Monoxide. CD-ROM, NOAA ESRL, Boulder, Colorado [Also available on Internet via anonymous FTP to ftp.cmdl.noaa.gov, Path: ccg/co/GLOBALVIEW], 2008.

(h) The whole section discussing the uncertainties on the data is really vague. Where are the uncertainties provided: reference article or website. Which uncertainties are specific to each measurement? Which are upper limit estimates for the whole dataset?

D. Section 3:

(a) Is the a priori forward simulation done with SACS?

(b) Line 12 p.20696: "Note that MCF emissions are very small since early 2005, after years of large industrial use." This does not say anything about 2004 and has not quantitative information.

(c) Line 19: Add "after 20 iterations, the norm of the **cost function** gradient. . . ""

(d) What is the lag for the inversion? How far back in time will observations on day d will impact surface fluxes? Is this mentioned anywhere?

ACPD

8, S11291–S11297, 2009

(e) Table 2 is interesting but can you say something about the regional differences on how well the filter performs and what causes them?

(f) Which units should be used: Mt or Tg?

(g) How are the uncertainties on the fluxes modified by the inversion?

(h) The CO ocean source seems to be changed: Can you provide the actual numbers?

(i) Provide a table with regional total estimates for all 3 species.

(j) When citing other studies, compare your CO regional fluxes with these other studies. For example, how much CO was emitted by fired in boreal North America in Pfister et al. [2005] and Turquety et al. [2007]? How confident are you in your inversion results?

(k) The authors should explain that they do not compute the a posteriori uncertainties on the fluxes in their variational inversion set-up. The numbers given in the "error reduction" section are clearly optimistic and do not take into account transport model errors.

(l) When you do the mono-species inversion, how many iterations does it take to converge? The reader needs to understand the cost of the multi-species approach. Giving relative changes of the fluxes is not very meaningful. Please put the regional fluxes estimates in the same Table where you will out the prior and the multi-species inversion posterior. Can you do an inversion with CO + MCF and another one with CH4 + MCF. It would be interesting to see how a different OH impacts your results. Are the OH concentrations from these 2 inversions and the multi-species one presented in the paper any different?

E. Section 3.4

(a) 2004 is not covered by Bousquet et al. [2006]. Please clarify. Page 20701 Bousquet et al. [2005] is cited: mistake? The two inversions were done with

ACPD

8, S11291–S11297, 2009

Interactive Comment

the same transport model so the inversion systems are NOT completely different.

(b) See also General Comments: When using the POC, AOC and WPC data: reference the labs and PI doing this work. Same for HNG and ORL. Observational data are not “off-the-shelf” items.

(c) Fig 9, use ppb instead of ppm

(d) See if redoing the comparison with MOPITT over the continents only and for daytime changes the numbers in Table 5.

F. Conclusions

(a) The CO emission under-estimations in India and China is not only due to fossil fuel burning but also biofuel burning.

(b) Put your study in the context of what the state of the science is and what other authors have shown. Once again, relative differences between your various inversions are not useful to compare with other authors’ quantitative estimates of sources.

(c) See also General comment # 6.

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