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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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Specific comments

P20688, L8-9: The independent observations were not only satellite retrievals but also ship and aircraft measurements.
This is right, the sentence will be corrected as: "Independent data from ship and aircraft measurements and satellite retrievals are used to evaluate the realism of the results."

P20688, L10: Mention also that CO increases are confined to the northern hemisphere (whereas the southern hemisphere shows a decrease). P20688, L11: Emissions of CH4 decrease (not increase) in boreal areas.
The sentence will be modified: "The total emitted mass of CO is 30% higher after

the inversion, due to increases of fluxes up to 35% in the Northern hemisphere. The spatial distribution of emissions of CH_4 is modified by a decrease of fluxes in boreal areas up to 60%."

P20689, L15: Throughout the text, the term 'dihydrogen' is used for H2 . However, this term is hardly ever used, so I propose changing it to 'hydrogen'.
The change to 'hydrogen' will be made throughout the text and captions.

P20690, L16-20: From this description it is not completely clear how the state vector is defined. At which temporal and spatial resolution are the 3-D concentrations optimized? At which temporal and spatial resolution are the initial conditions optimized?
This was not clearly stated so we will add the following sentences: "[. . .] the initial conditions of all concentrations (see Sect. 2.2.2 for details on species). The concentrations (HCHO production and OH plus initial values) are optimized as column averages over each surface grid cell at the same 8-day temporal resolution. OH concentrations are further aggregated into four bands of latitude, as described in Bousquet et al. (2005). "

P20692, L12-13: The references to papers by Hauglustaine et al. (2004) and Folberth et al. (2005) suggest that the SACS scheme was developed and used in those studies. However, this does not seem to be the case. Therefore, I assume that SACS is actually a new scheme. If this is true, it should be explained in more detail. Reactions should be explicitly written down and assumptions for reaction rates of ensembles of reactions should be provided.
This is right, SACS has never been published before. The list of reactions will be added in this section: "The intermediate reactions not represented in Fig. 1 are regarded as very fast compared to the principal reactions above which are described in SACS as:

$$CH_4 + OH \rightarrow CH_2O + \ldots$$
$$CH_2O + h\nu + O_2 \rightarrow H_2 + CO$$
$$CH_2O + h\nu \rightarrow 2HO_2 + CO$$

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$CH_2O + OH + O_2 \rightarrow HO_2 + CO + H_2O$
$CO + OH \rightarrow CO_2 + H$
$H_2 + OH \rightarrow H_2O + H$
$CH_4 + O1D \rightarrow CH_3O_2 + OH$
$CH_4 + O1D \rightarrow H_2 + HCHO$
$CH_3CCl_3 + OH \rightarrow CH_2CCl_3 + H_2O$
In addition, H_2 deposition is parameterized as: $H_{2deposited} = v_{dep}.[H_2]$ where v_{dep} is the deposition velocity. Reaction and photolysis rates of SACS are those of LMDz-INCA (Folberth et al., 2005b). OH concentrations, the three-dimensional chemical production of HCHO by VOCs and v_{dep} are taken from a full forward simulation by LMDz-INCA.

P20694, L10-14: Chevallier et al. (2005) is a study on CO2 . Please provide details on variances assumed for the different emissions. Report also the prior assumptions for other parts of the state vector (initial conditions, 3-D concentrations). Are temporal correlations taken into account?
This was not clearly stated so we will add the sentence: " [. . .] 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. Correlation lengths of [. . .]"

P20695, L4-8: Are representativeness errors taken into account?
The data from GLOBALVIEW take into account the synoptic variations. We assumed here that they are a 'proxy' for transport errors. We will re-phrase this paragraph: "The errors on observations were set as follow. For CO and CH_4, we used yearly means of synoptic variability from GLOBALVIEW; for MCF, we used yearly or monthly values from Prinn et al. (2005) or from the NOAA; and for H_2,

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data from CSIRO or from the NOAA were used. It is assumed here that the synoptic variability is an approximation of the transport errors, which is consistent with the weaknesses of global model in reproducing continental synoptic concentrations of trace gases (Geels et al., 2007). Otherwise, if these data were not available, the uncertainties associated with the measurements were used. In all cases, a minimum uncertainty of $\pm$3 ppb for CH_4, CO and H_2 and $\pm$1.2 ppt for MCF is fixed, in order to prevent the error getting too small."

P20695, L9: How are the measurements 'filtered'?
In this case, measurements are thinned: if several observations occur in the same grid cell at the same time-step (which is not very often with 'event' and 'daily' data), we just keep the first of them (we checked that this does not induce any particular bias).

P20695, L10-12: It can be dangerous to withdraw observations which are further than three standard deviations away from the prior simulation. Is it checked whether these are single outliers or series of measurements which are consistent with each other but not with the prior model simulation? In the latter case, they should certainly not be removed from the analysis.
We indeed examinated the time-series of measured and simulated concentrations at the stations during the year. We could then verify that it was safe to discard measurements further than three standard deviations away from the prior because they were truly outliers for all measured species. It would the case for measurements only two standard deviations away for H_2 for example.

P20696, L4: Both 'Tg' and 'Mt' are used in the text. Notation should be consistent, preferably 'Tg', since I doubt whether 'Mt' is an SI-unit.
This correction will be made throughout the text and captions.

P20696, L19-20: A reduction in the norm of the gradient by 80% is very modest (not even one order of magnitude). Have the authors carefully checked the adjoint imple-

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mentation? Has the optimization really converged after 20 iterations?
We should have stated that even with a simplified chemistry scheme, the computational burden of the optimization is quite heavy and limiting: for 20 iterations with M1QN3, the total time of computation is two months on a Opteron 64 bits architecture. We noticed that with 20 iterations, the norm of the gradient that remains is mainly due to H_2. We are well aware of the limitations of the results due to this and should have stated it more clearly in the text: this will be corrected in this paragraph: "The emission fluxes of CO, CH_4, H_2 and MCF have been simultaneously inverted from January to December. After 20 iterations, the norm of the gradient is reduced by about 83%. Due to the computational burden of the inversion with the chemical scheme, it was checked that the convergence was satisfying enough on emission fluxes (particularly CO and CH_4) for a first attempt at evaluating the inverted fluxes." The parallelization of the code has now began.

P20697, L13: Initial conditions of OH are not important, since OH is extremely short-lived. Do the authors mean 'a priori concentrations'?
This is right, it will be corrected.

P20699, L5-9: The results for H2 are very confusing. Why is the analysis increment confined to such a small region (where by the way no measurements are taken!)? Is this related to the prior emissions? Please explain this in much more detail. Otherwise, Fig. 6 is better removed from the manuscript.
The Figure with its zoom is indeed misleading, here
`http://www-lsceorchidee.cea.fr/ACPD/acpd-2008-0504-f06bis.eps`
is Figure "06bis" with the whole map that should read better. What we meant to show here is the type of problem that can occur due to the optimization of net fluxes whereas H_2 has emission and deposition fluxes. Therefore, the zoom was on South America and Africa where the deposition velocities are high (more than 0.03 cm.s^{-1} on average in the year). We propose to detail this by re-writting this

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paragraph in the following way: " H_2 The decrease in H_2 emission is located in particular areas, as displayed in Figure 6. Some of these areas correspond to the surroundings of point where constraints are available as in the case of Australia with station CGO or in East China around station TAP. In the other areas, such as South America, Africa, Indonesia, China, Europe, USA, the corrections to the fluxes may be large at places: optimized fluxes are up to 10 times smaller than the prior and their signs can be opposite like in South America for one pixel (pink in Figure 6). This seems to be linked to the high deposition velocities prescribed (more than 0.05 cm.s^{-1} on average in 2004). The distinction emission and deposition fluxes in the inversion is technically feasible but would require more constraints, such as isotopic measurements."

P20700, L6: I had the impression that 'up to now' results of a multi-species inversion were shown.
This was poorly worded, the sentence will be corrected: "In order to assess the advantage of running a multi-species inversion instead of a mono-species one, two mono-species inversions are run."

P20700, L5-9: Regarding the mono-species inversions I have the following questions. Is OH optimized? Which observations are used (only from species optimized?) Why are initial conditions for all species optimized?
In the mono-species inversion, OH concentrations are not optimized: as in the study by Bousquet et al. (2005), the OH fields from the MOZART model are prescribed. The observations used are only from the optimized species. The initial conditions were left to be adjusted by the optimization for equilibrium with OH but this has no actual consequence on the inversion. To be clearer the sentence will be modified: "In these inversions, only the emission fluxes of either CO or CH_4 are optimized with either CO or CH_4 observations as constraints (the state vector therefore does not include OH concentrations)."

P20700, L10-21: A Figure showing differences between CO emissions from multi- and

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mono-species inversion would be instructive.
We will add Figure "07bis" showing the difference between mono- and multi-species optimized CO emission masses (in Tg) per pixel for 2004 (`http://www-lsceorchidee.cea.fr/ACPD/acpd-2008-0504-f07bis.eps`) with the following comment: "[...] the difference between optimized fluxes are significant in particular regions. The difference between optimized masses obtained with the mono- and multi-species inversions are displayed in Figure 7bis at a pixel scale. The mono-species inversion lead to larger emitted masses than the multi-species case in regions North America boreal (+23 percentage points), USA (+6 percentage points), Europe (+6 percentage points) and Eurasia boreal (+33 percentage points). In the region of the Indian peninsula, the mono-species emitted masses are lower (-6 percentage points) than the multi-species results.

P20701, L9: What is 'not globally different'? I would say the 3.4% increase in global CH4 emissions is a considerable change.
'Not globally different' understates the difference for methane indeed. Actually, the study by Bousquet shows a global inter-annual variability of approximately 10% so 3.4% is significant even though it is not so large. We will rephrase this sentence: "The results obtained with the multi-species system are not globally different from the results of a mono-species inversion for CO; for CH_4, the difference between the two methods amounts to less than 50% of the inter-annual variability ($\approx$10% in mass in Bousquet et al. (2006))."

P20702, L5: Report how many flights were performed at both locations and how they were spread over the year.
We will add this information: "[...]202 three-dimensional observations are given by aircraft measurements made in Hungary (HNG, 12 flights in the year, none in February and March, 2 in April and December, 1 for the other months) and in France (ORL, 25 flights in the year, 1 in January, March and April, 2 in June and August, none in December, 3 for the other months) [...]"

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P20702, L11-15: Is this really a signifiant improvement?
Our sentence was poorly written. We will change it to: "The fit between simulation and mobile surface data (2D-data), which is already quite good as a first-guess, is not significantly changed by the inversion (the slope is increased from 0.97 to 0.98 for the same correlation coefficient)."

P20702, L16-17: The slope of a linear correlation says nothing about overestimation or underestimation.
Indeed we did not indicate all the statistics. We will add information as stated in our answer to the question on Table 5 hereafter.

P20702, L16-25: The RMS may have improved, but why has the correlation decreased? This fact should at least be mentioned.
There was a typo in this Table, the figure are actually 0.68 for the first-guess and 0.69 for the analysis: this will be corrected.

P20703, L5: The slope of a linear correlation says nothing about overestimation or underestimation.
See the answer two paragraphs above.

P20703, L25: At which spatial and temporal scales are these uncertainty reductions valid?
The uncertainty reductions are computed at the same space and time scales than the optimized fluxes i.e. the pixel size and eight-day periods.

P20704, L5-6: Mention that this is the CH4 inversion by Bousquet et al.
This will be corrected: "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."

P20704, L5-12: This paragraph is generally too positive about the comparison of the inversion with independent CH4 observations. (i) The fit to background CH4 obser-

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vations is not significantly improved by the inversion (same correlation coefficient; only 0.01 increase in the slope). (ii) The RMS of the deviation from observed vertical profiles may have improved, but the correlation coefficient has decreased. (iii) For CO, there is indeed improved agreement with MOPITT in terms of the slopes of the linear fits, but the correlation coefficient shows hardly any improvement. Also, this improvement is a logical consequence of the dramatic underestimation of CO concentrations in the prior simulation.

We propose to re-formulate this paragraph: "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."

P20704, L13-15: SCIAMACHY methane columns have not been used for validation in this study. Suggest to reword the sentence to something like 'The next step is now to use satellite observations, such as SCIAMACHY CH4 columns and MOPITT CO columns, and surface measurements simultaneously as constraints in a multi-species inversion'.

This will be corrected as indicated.

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