Atmos. Chem. Phys. Discuss., 8, S11083–S11092, 2009
www.atmos-chem-phys-discuss.net/8/S11083/2009/

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 #1

Received and published: 10 February 2009

General comments

In this paper a method is presented to derive emissions of multiple species from surface measurements of those species. Compared to single-species inversions, this approach offers the potential to use many more constraints and to derive emissions in a chemically consistent way. This is certainly the direction in which we should be going, and this paper is thus a good step forward. However, I have a number of concerns:

1. The multi-species inversion approach is not new. It has been used before by e.g., Müller and Stavrakou (2005) and Elbern et al. (2007). These references (and maybe more) should be cited.
2. The multi-species inversion is compared with CO and CH_4 mono-species inver-

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Interactive Discussion
Discussion Paper

sions, but differences are found to be small. The paper would be considerably strengthened if the authors could provide a case for which the multi-species inversion does a (demonstrable) better job than a corresponding single-species inversion. The reader should be convinced that it is useful to do multi-species inversions.

3. There appear to be inconsistencies in the reported trace-gas budgets (see specific comments). These should be clarified and repaired.

4. The evalutation of the analysis results with independent observations is not very convincing (see specific comments).

Specific comments

P20688, L8-9: The independent observations were not only satellite retrievals but also ship and aircraft measurements.

P20688, L10: Mention also that CO increases are confined to the northern hemisphere (whereas the southern hemisphere shows a decrease).

P20688, L11: Emissions of CH_4 decrease (not increase) in boreal areas.

P20689, L15: Throughout the text, the term ‘dihydrogen’ is used for H_2. However, this term is hardly ever used, so I propose changing it to ‘hydrogen’.

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?

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.

P20694, L10-14: Chevallier et al. (2005) is a study on CO_2. 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?

P20695, L4-8: Are representativeness errors taken into account?

P20695, L9: How are the measurements ‘filtered’?

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.

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.

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 implementation? Has the optimization really converged after 20 iterations?

P20697, L13: Initial conditions of OH are not important, since OH is extremely short-lived. Do the authors mean ‘a priori concentrations’?

P20699, L5-9: The results for H_2 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.

P20700, L6: I had the impression that ‘up to now’ results of a multi-species inversion

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Interactive Discussion

Discussion Paper

were shown.

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?

P20700, L10-21: A figure showing differences between CO emissions from multi- and mono-species inversion would be instructive.

P20701, L9: What is ‘not globally different’? I would say the 3.4% increase in global CH_4 emissions is a considerable change.

P20702, L5: Report how many flights were performed at both locations and how they were spread over the year.

P20702, L11-15: Is this really a significant improvement?

P20702, L16-17: The slope of a linear correlation says nothing about overestimation or underestimation.

P20702, L16-25: The RMS may have improved, but why has the correlation decreased? This fact should at least be mentioned.

P20703, L5: The slope of a linear correlation says nothing about overestimation or underestimation.

P20703, L25: At which spatial and temporal scales are these uncertainty reductions valid?

P20704, L5-6: Mention that this is the CH_4 inversion by Bousquet et al.

P20704, L5-12: This paragraph is generally too positive about the comparison of the inversion with independent CH_4 observations. (i) The fit to background CH_4 observations 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.

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 CH_4 columns and MOPITT CO columns, and surface measurements simultaneously as constraints in a multi-species inversion.’

Table 1: Is ‘road transport and shipping’ a biogenic emission? Also, some CH_4 emission categories (e.g., soil uptake, termites) appear to be missing. What is ‘QUANTIFY2 EU’? What is GFED-v2? Is ‘GFED-v2’ (used for CO) biomass burning inventory the same as ‘van der Werf et al. (2006) (used for CH_4)?

Table 2: It is better to consider observations as the reference and to calculate a bias as ‘simulated minus measured’, instead of the other way round.

Table 2: Why is the median taken and not the mean?

Table 2: Is ‘correlation’ the (linear) correlation coefficient or the explained variance?

Table 2: The percentual differences appear to be defined as: (abs(posterior) - abs(prior))/abs(prior). The caption reverses posterior and prior and does not mention that absolute values are used.

Table 3: Again the percentual differences are defined as posterior minus prior, instead of the other way round.

Table 3: It would be instructive to add the initial burden and the accumulation during the year for each species. These numbers are referred to in the text.

Table 3: The chemical loss of MCF increases (implying increased OH), whereas the chemical loss of CH_4 decreases (implying decreased OH). How is this possible?

Table 4: The Bousquet et al. (2006) study considered the period 1984 to 2003. How can emissions for 2004 be quoted from this study? Furthermore, the listed prior emissions for Bousquet et al. add to 707.9 Tg. This is an extremely large number and much larger than the 472 Tg (497 Tg minus 25 Tg soil sink) reported in the supplementary material of Bousquet et al. Finally, the total optimized and prior emissions from this study, as reported in the last two columns of Table 4, are not the same as in Table 3.

Table 5, caption: Change first sentence to ‘Slope a and correlation coefficient r of linear regressions of simulated against observed CH_4 and CO concentrations for different observation types.’.

Table 5: It is much clearer to use ‘ocean’ and ‘aircraft’ instead of ‘2-D’ and ‘3-D’. (Actually, the aircraft observations are 1-D (height dimension) rather than 3-D.)

Table 5: It would be instructive to add the number of observations for each observation type, and to provide also bias and standard deviation. This would make it possible to speak of over- and underestimations, about which the slope of the linear regression tells provides no information.

Fig. 1: As stated before, the SACS scheme is not well described. For example: is OH prescribed or calculated interactively (if yes, how?)? What are the values of the j- and k- constants? How are VOCs included?

Fig. 1: The caption says that all variables in italics are optimized. Does this mean that VOCs are optimized? Is deposition optimized for H_2 (and how?), but not for MCF? How is OH optimized? As a 3-D field or with a general scaling factor? And with which temporal resolution?

Fig. 2: Was the standard deviation based on (instantaneous) tracer distributions at the end of January 2004? Or on monthly means? The latter would not be fair, since

concentrations from the beginning of the month (which are the same because of the initialization) would be mixed into the statistics.

Fig. 2: The differences between INCA and SACS are very large for CH_2O. How can this be explained? Would this be a problem if CH_2O observations were added in later studies?

Fig. 2: From the text I got the impression that OH is fixed in SACS. But then the MCF differences between INCA and SACS should be zero, which is not the case. Thus, please provide more details on how OH is treated in SACS?

Fig. 4: Why are values for slopes and correlation coefficients so different from Table 2? This is confusing.

Fig. 5c: Is panel c ok? The large grey area, extending even over the oceans, seems like an error.

Fig. 5a and c: Increases in emissions by more than a factor 20 on a pixel basis (and 229% over a big region) are extremely large. I wonder what prior errors settings have been used to accommodate emission increments of this magnitude.

Fig. 6: What is the meaning of white?

Fig. 7: Why not make this figure in color, like all the others? This would improve clarity. Also, I recommend adding a panel with a similar plot for CO emissions.

Fig. 8: Replace '2-D- and 3-D-mobile' by 'ship' and 'aircraft', and explain what the colors mean.

Fig. 9: It is more logical to plot bias and standard deviation, since these constitute two independent components of the error. I suspect the authors have avoided to plot the bias, since the bias appears to have increased significantly at ORL.

Fig. 9: Why are the profiles so different between the two stations? I.e.: why is the correspondence between model and observations near the surface so much better in

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ORL than in HNG?

Technical comments

Here is a list of comments as a guideline for the authors to improve the presentation of their study.

P20689, L20: Place ‘HCHO’ between brackets or write ‘(CH_4, CO, HCHO (formaldehyde), and H_2)’.

P20690, L17: Replace ‘space’ by ‘spatial’.

P20690, L18: Write out ‘3-D’ at first occurence.

P20691, L10: ‘LMDZ’ is sometimes written with lowercase ‘z’ and sometimes with capital ‘Z’. Be consistent.

P20692, L16-18: ‘Wet lands’ should be ‘Wetlands’. Also, remove ‘the’ before the source names. Also, anaerobic processes do not take place in all of those sources.

P20692, L25-27: What is ‘combustion of fossil energies’?

P20693, L10: Remove ‘de’.

P20694, L27: Replace ‘location’ by ‘locations’.

P20695, L6: What is ‘or the NOAA’?

P20695, L9: ‘data’ should be ‘datum’ (or ‘measurement’).

P20696, L4: Add ‘much’ between ‘accumulate’ and ‘in’.

P20697, L11: Remove comma after ‘inversion’.

P20698, L19 and 22: Replace ‘increase of’ by ‘increase by’.

P20700, L10: Replace ‘After the mono-species inversion’ by something like ‘In the CO mono-species inversion’.

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P20700, L19-21: What are ‘points’?

P20702, L5-6: ‘tri’ should be ‘three’. Or use ‘3-D’, once this abbreviation has been introduced earlier in the text.

P20702, L21-22: Replace ‘RMS’ by ‘RMSs’ or ‘RMS values’.

P20702, L26: Start new sentence at ‘we’.

P20704, L6: Replace ‘lay’ by ‘lie’.

P20704, L11: Replace ‘measures’ by ‘measurements’.

Table 1: Add semicolons between different emission sources on one line.

Table 2: Use consistently two digits for correlations.

Table 3: Don’t remove the last digit if it is a zero (3 cases).

Table 4: Bousquet et al. was published in 2006 instead of 2005.

Table 4: Again, don’t remove the last digit if it is a zero.

Fig. 4, caption: Replace ‘as functions of’ by ‘compared with’.

Fig. 4, caption: Replace last sentence by: ‘Slopes a and correlation coefficients r of linear regressions [mod]=a[obs]+b are also indicated’.

Fig. 5c: Do not use grey in the color scale.

Fig. 5, caption: Add ‘for CO and CH_4’ in first sentence.

Fig. 9, figure and caption: ‘rms’ should be capitalized.

References

J.-F. Müller and T. Stavrakou (2005), Inversion of CO and NOx emissions using the adjoint of the IMAGES model, Atmos. Chem. Phys, 5, 1157-1186.

H. Elbern, A. Strunk, H. Schmidt, and O. Talagrand (2007), Emission rate and chemical state estimation by 4-dimensional variational inversion, Atmos. Chem. Phys., 7, 3749-3769.

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

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