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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Optimizing global CO emissions using a four-dimensional variational data assimilation system and surface network observations" *by* P. B. Hooghiemstra et al.

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We would like to thank the reviewer for this extensive review. We feel that by taking the comments into account the quality of the paper is improved. Below we answer all comments and indicate if necessary which text has been changed. As indicated in more detail below, major changes have been made to Section 4, where we added a paragraph to elaborate on the observation error. In short, we found that that the main reason for the rejection of the large amount of observational data is the coarse model resolution, in which both high and low observations are poorly reproduced. A detailed study of the model representativeness error showed that it remains extremely

challenging to define a model error representation that works well for all stations.

Furthermore we changed the discussion paragraph on separation of the sources (§4.1). The main conclusion is that the available observations constrain total CO emissions and therefore, the uncertainty reduction in the total emissions is larger than in the individual emission categories. Hence, in the posterior solution, the source categories are negatively correlated. We now more clearly emphasize that this implies a solution space in which the different source categories can hardly be separated.

Since the topic of the current paper is a thorough testing of the 4D-VAR system, we acknowledge current shortcomings and apply improvements in future work.

Answers to major comments :

1. p. 353 : Please illustrate the advantages from the use of the current assimilation system compared to other inversion techniques where the initial concentration field is not optimized. How much would the results be different if instead of optimizing the initial CO concentration field, we used a longer spin-up time (6-8 months instead of 1 month)? The differences between the two approaches should be made more apparent in the text in a way to make the results more sound and demonstrate the usefulness of this approach also in the case of CO.

First of all, increasing the spin up time (for example, to 8 months) would result in a simulation window of 22 months and hence the already computationally intensive computations would take 50% more time. Second, we did sensitivity tests to investigate the necessity of optimizing the initial CO mixing ratio field. It turned out that starting from an optimized initial CO field from a previous run or optimizing a field from a forward run with prior sources, did not have a large effect on the derived emission estimates in the months after the spin up. Finally, this approach has been applied successfully for methane inverse modeling (note that the methane lifetime is nine years) previously (e.g., Meirink et al. 2008, Bergamaschi et al. 2005, 2009, 2010) and is therefore adopted here.

In paragraph §2.2, page 350 lines 6-10, we changed the text to: "A forward model simulation with these a priori emissions has been performed for the years 2002-2005 and daily mean CO mixing ratios have been archived. The a priori initial CO mixing ratio field is taken from this archive and further optimized by including the initial 3-D field in the state vector. This approach has been adopted from previous methane 4D-VAR studies (e.g., Meirink et al. (2008b)). Also, when optimizing the initial CO mixing ratio field in this way, a long spin up time is not necessary saving up to 50% of computer-time. The approach outlined here yields similar emission estimates compared to an inversion starting from a posterior field from a previous run that is not further optimized. "

2. page 349, last paragraph : To determine the NMVOC-CO source, you subtract the monthly CH4-CO term, which is derived using climatological OH (same page, line 10), from the total CO field, derived from a full TM4 model run with OH calculated in the model. Should this subtraction make sense, the OH fields used must be the same.

We agree with the reviewer that to obtain fully consistent CH4-CO and NMVOC -CO field, the OH fields used should be the same. However, this is not the case. But one should bear in mind that the CH4-CO and NMVOC-CO fields are only used as prior (3D) emissions. We think therefore that the impact of this small inconsistency is small, since the system optimizes both chemical CO sources and can therefore account for inconsistencies in the prior estimates.

3. page 352, line 20 : Leaving out every one out of five GMD data might bias the a posteriori estimates. Note that the posterior bias after the second inversion cycle is higher in 35% of the stations compared to the first inversion cycle (Table 1). How different are the posterior emissions obtained after the first inversion cycle compared to the final result, in terms of both magnitude and spatial distribution?

This comment was also posted by the other reviewer. Therefore, we refer to our answers on comments 9, 10 and 11 in the author comment to reviewer 1.

4. page 350, first paragraph : One globally-defined factor is optimized per month for the NMVOC-CO source. This is a shortcoming in the inversion setup. In particular, it leads to very strong changes in this source, from the prior 812$\pm$40 to 574$\pm$38 Tg CO/yr in 2003 and 410$\pm$36 Tg CO/yr in 2004, which are mainly driven by the strong prior model overestimation at remote SH sites (South Pole, Syowa station, Argentina, Table 1), in regions where only the NMVOC-CO source is present. Furthermore, a difference of more than 150 Tg CO in the posterior NMVOC-CO estimates between 2003 and 2004 cannot be but an artefact, as there is, to my knowledge, no physical reason explaining such a difference. Such artefacts could be avoided, if more than one emission parameters for this source are determined by the assimilation, e.g. one per continent or big region. In that case, a strong posterior NMVOC-CO reduction would have been derived only in the SH regions but not on the global scale. This feature should be changed in the setup to yield more realistic results.

We agree with the reviewer that the difference in NMVOC-CO of 150 Tg CO from 2003 to 2004 is not driven by a physical reason. It is in fact a consequence of the setup of the NMVOC-CO source as described in the revised section §4.1. However, the compensation effects described in that paragraph would not be solved by using one parameter per continental region. This is a direct consequence of the fact that the observations only constrain total CO emissions. Hence, the uncertainty reduction for the sum of the emission categories is larger than the uncertainty reduction for the individual source categories, which is expressed as a negative correlation between the posterior estimates. Therefore, compensation of one source, for the changes in another source will remain, as any separation of sources is prescribed in the prior. In a follow up study we are currently optimizing the NMVOC-CO source as a 2D emission field, but also in this set-up compensating emission increments are observed.

Answers to specific comments :

1. p. 377, Table 2 : Rearrange the table so as same columns do not appear twice (i.e. prior anthropogenic emissions 2003 and 2004 or prior natural emissions). In

addition, global totals never match the sum of individual regions : for prior natural emissions the global total is equal to 115 Tg CO whereas the sum is by 16% lower, but for anthropogenic emissions the global total amounts to 531 Tg CO whereas the sum is somewhat higher (532 Tg CO). Is there a reason for this? Please correct or explain. Also the isoprene emission inventory used and the global isoprene source should be mentioned.

We changed the table so that columns do not appear twice. We also added some footnotes to explain the inventories used in our setup, including the isoprene emission inventory and the global isoprene and monoterpenes source. The emission estimates for the continents are derived by placing a rectangular box around the continents and summing the emission estimates from the grid boxes falling in this box. So it is possible for some continents, that small parts of land with emissions are not taken into account. For the global totals, these emissions are added, explaining the difference.

2. p. 378, Table 3 : Make a more complete intercomparison table including more studies and emissions by category when available - especially for Asia (Bergamaschi et al., 2000, Arellano et al. 2004, Stavrakou et al. 2006, Kopacz et al. 2009). To ease readability, you might want to add table footnotes to specify details, e.g. prior emissions for different studies, data used to constrain the emissions. In that case, Section 3.4 should be lightened and contain more qualitative discussion.

The aim of Table 3 is to compare our emission estimates with other studies inverting CO emissions for specificly 2004. This has been added to the text for more clarity. All studies proposed by the reviewer (Bergamaschi et al., 2000, Arellano et al. 2004, Stavrakou et al. 2006, Kopacz et al. 2009) inverted for the year 2001 or earlier, which makes a direct comparison difficult. We therefore stick to comparison with the current studies only. However, followed the suggestion of the reviewer to add table footnotes to ease readability of this table.

Answers to comments on writing and mispells:

1. p. 345, l. 1 : replace “Synthesis" by “synthesis"

changed

2. p. 345, l. 9-12 : “of the underlying CTM, thereby...Adjoint inversions are in particular suited" should be rephrased as e.g. “...of the underlying CTM, through an iterative approach used to minimize the mismatch between model and observations. Adjoint inversions reduce the risk of aggregation errors and are in particular suited..."

changed

3. p. 345, l. 16 : “large amounts of observational data" should be replaced by “large observational datasets"

changed

4. p. 345, l. 21 : “that are set on the sources" should be replaced by “assigned on the sources"

changed to "assigned to the sources"

5. p. 346, l. 11 : place a comma after matrix R

changed

6. p. 346, l. 12 : replace “weighted with" by “weighted by"

changed

7. p. 347, l. 10 : mispelled “descend"

This text block has been changed, the word descend is removed.

8. p. 347, l. 22-24 : “it is not possible...preconditioner L" should be replaced by “the preconditioner is too large to be stored. The approach of Meirink et al. (2008b) is therefore adopted to reduce the required storage"

changed

9. p. 347, l. 25 : replace “the method converged" by “that the minimum is reached"

changed to "the minimum of the cost function reached"

10. p. 349, l. 15 : please explain why a constant methane mixing ratios is imposed, instead of using the methane simulated with a full TM4 chemistry run.

We chose the very simple CH4 = 1800 ppb, because we assume that this source of CO is relatively well known (compared to for example the NMVOC-CO source). Also, we quantify possible under- and over estimates we make compared to using more realistic CH4 fields. In a follow-up study, CH4 values derived from a similar 4D-Var study will be used.

11. p. 349, l. 24-25 : Could you specify the VOC sources used to drive TM4 model?

The emission inventories driving the TM4 model are reported in Myriokefalitakis et al. (2008, ACP). Emissions are taken from the POET/GEIA databases. This information has been added to Table 2.

12. p. 351, l. 19 : remove “only"

We keep the word "only" to emphasize that later on we want to assimilate other (satellite) datasets also.

13. p. 352, l. 1 : how is the value of 1.5 ppb derived?

We added a reference to Novelli et al. (1998).

14. p. 352, l. 3 : use citet for Bergamaschi et al. 2005 citation

15. p. 352, first paragraph : Please give the formulas used to derive the model error in the vertical and horizontal direction, and provide the resulting error estimates.

16. p. 352, l. 10-11 : this information is already in Section 2.1 and can be omitted

Answer to comments 14-16: §2.4 and §2.5 have been changed. In §4.2 a detailed discussion of the observation error is included.

17. p. 353, l. 7 : replace “computer-time" by “computationally"

changed

18. p. 353, l. 11 : replace “optimize emissions in a certain month m" by “optimize emissions of month m"

changed

19. p. 353, l. 14 : “are still influenced" should be replaced by “are in reality influenced"

We disagree with the reviewer here. We assume there is a correlation in time between the emissions in this setup. However, in reality there won’t be a correlation backwards in time. That is, emissions in month m+3 will definitely not influence emissions in month m. We removed the word "still".

20. p. 353, l. 17 : “according to" should be replaced by “that is"

changed

21. p. 353, l. 25 : remove “the" in “the future ingestion"

changed

22. p. 354, l. 17 : “the system is behaving well", please elaborate

This is described in more detail at the end of that section.

23. p. 355, first 4 lines : Unease to follow here, please rephrase, e.g. “Similar values are also reported...."

changed

24. p. 355, l. 16-18 can be replaced by : “This is attributed to an underestimation of anthropogenic emissions in the EDGAR inventory, which was compiled for the year 2005."

changed. However, the EDGAR used here was compiled for the year 1995!

25. p. 355, l. 26 : replace “has the ability to better exploit" by the shorter “better exploit"

changed

26. p. 356, l. 4 : replace “station South Pole" by “South Pole station"

changed

27. p. 356, l. 9 : replace “obtains a value of" by “equals to"

changed

28. p. 356, l. 12 : replace “shows values" by “is"

changed

29. p. 356, l. 20 : why put “only" here?

Because for fine-scale emission tuning, we should iterate on much further.

30. p. 357, l. 4 and 5 : remove e.g. from parentheses

changed

31. p. 357, l. 12 : “inter annual" should read “interannual"

changed

32. p. 358, l. 6 : remove “that is"

changed

33. p. 358, l. 10 : replace “with altitudes" by “at altitudes"

changed

34. p. 359, l. 1 : remove “surprisingly" as this is already reported in Kopacz et al. 2010 and mentioned later in the manuscript (at page 366)

We agree with the reviewer and changed the text here.

35. p. 359, l. 4 : remove sentence “Over the continents..."

changed

36. p. 359, l. 15 : replace “showed" by “suggested"

changed

37. p. 360, l. 9 : “anthropogenic emissions over the United States" is only fossil fuel emisisons

changed

38. p. 360, l. 11 : “This value was further decreased..."

changed

39. p. 360, l. 13 : remove “only" and “the"

We do not remove "only" and "the", because "only" is to emphasize that they did not invert for the whole year of 2004. "the" refers to the satellite instruments.

40. p. 360, l. 14 : read “and presented results as yearly totals"

changed

41. p. 360, l. 18 : read “are by 25% lower"

changed

42. p. 362, l. 4 : read “are dominant"

43. p. 362, l. 7 : read “month-to-month"

44. p. 362, l. 9 : read “North and South America"

Answer to comments 42-44: We changed the text, these words have been removed.

45. p. 363, l. 6 : read “focuses"

changed

46. p. 364, l. 5 : replace “with 67 Tg CO" by “by 67 Tg CO"

47. p. 364, l. 13 : read “should improve with the assimilation"

Answer to comments 46,47: these sentences are removed.

48. p. 365, l. 23 : read “the remote SH still underestimates MOPITT..."

changed

49. p. 365, l. 25-26 : replace “one would expect to infer higher biomass burning emissions" by “higher inferred biomass burning are expected"

changed

50. p. 366, l. 3 : read “emissions increase by 75 Tg"

changed

51. p. 366, l. 4 : read “compensated by decreased"

changed

52. p. 366, l. 15-19 : this point should be discussed earlier (e.g. at p. 359 line 1)

In the revised manuscript we discuss this point now in §3.3 where we validate our results with MOPITT. Here we changed the text to: " However, the agreement with MOPITT CO on the SH total columns does not improve. As stated before, it seems that the surface observations and MOPITT CO total columns over the remote SH are not consistently modeled. "

53. p. 366, last paragraph : overstatement - should be omitted

We removed this paragraph in the revised paper.

54. p.367, l. 4-5 : read “...from NOAA. The posterior simulation..."

changed

55. p.367, l. 14 : read “...have been evaluated against non-assimilated"

changed

56. p. 367, l. 21-22 : read “deteriorates from a 6% negative bias in the a priori to a 40% negative bias in the a posteriori solution, due to an emission decrease suggested by SH surface observations"

changed, note that the numbers have changed due to an error in the computations we found. See also our answer to reviwer 1, comment 16.

57. p. 368, l. 2 : replace “show that it is possible" by “illustrate the capability"

changed

58. p. 368, l. 9 : replace “inversion. This shows that" by “inversion, indicating"

This sentence has been reformulated.

59. p. 368, l. 12 : read “study using different fire injection heights"

changed

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 341, 2011.