

Interactive
Comment

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.

Anonymous Referee #2

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The aim of this paper is the optimization of global CO emissions for 2003 and 2004 and the reduction of the uncertainty of the emission estimates from individual sources. To this purpose, a 4-dim variational data assimilation system is designed, based on a version of TM5 model using simplified CO-OH chemistry and prescribed OH fields. CO concentration data from 31 GMD surface network stations are assimilated in this system. The subject of the paper is well within the scope of *Atmospheric Chemistry and Physics Journal*. The article is written in a clear way but there is much room for improvement (see “Comments on writing and misspells”). Further, the benefits from using 4d-VAR assimilation technique with respect to previously used methods for deriving CO fluxes should be put forward. The manuscript can be accepted for publication in the *At-*

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Interactive Discussion

Discussion Paper



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.
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.
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?
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 ± 40 to 574 ± 38 Tg CO/yr in 2003 and 410 ± 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

Interactive Comment

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)

[Discussion Paper](#)



Interactive
Comment

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.

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.
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.

Comments on writing and mispells :

Interactive
Comment

1. p. 345, l. 1 : replace "Synthesis" by "synthesis"
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..."
3. p. 345, l. 16 : "large amounts of observational data" should be replaced by "large observational datasets"
4. p. 345, l. 21 : "that are set on the sources" should be replaced by "assigned on the sources"
5. p. 346, l. 11 : place a comma after matrix R
6. p. 346, l. 12 : replace "weighted with" by "weighted by"
7. p. 347, l. 10 : misspelled "descend"
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"
9. p. 347, l. 25 : replace "the method converged" by "that the minimum is reached"
10. p. 349, l. 15 : please explain why a constant methane mixing ratios os imposed, instead of using the methane simulated with a full TM4 chemistry run.
11. p. 349, l. 24-25 : Could you specify the VOC sources used to drive TM4 model?
12. p. 351, l. 19 : remove "only"
13. p. 352, l. 1 : how is the value of 1.5 ppb derived?

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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
17. p. 353, l. 7 : replace "computer-time" by "computationally"
18. p. 353, l. 11 : replace "optimize emissions in a certain month m" by "optimize emissions of month m"
19. p. 353, l. 14 : "are still influenced" should be replaced by "are in reality influenced"
20. p. 353, l. 17 : "according to" should be replaced by "that is"
21. p. 353, l. 25 : remove "the" in "the future ingestion"
22. p. 354, l. 17 : "the system is behaving well", please elaborate
23. p. 355, first 4 lines : Unease to follow here, please rephrase, e.g. "Similar values are also reported...."
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."
25. p. 355, l. 26 : replace "has the ability to better exploit" by the shorter "better exploit"
26. p. 356, l. 4 : replace "station South Pole" by "South Pole station"
27. p. 356, l. 9 : replace "obtains a value of" by "equals to"

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

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

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

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

32. p. 358, l. 6 : remove "that is"

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

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)

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

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

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

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

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

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

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

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"

Interactive
Comment

Full Screen / Esc

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Interactive Discussion

Discussion Paper



45. p. 363, l. 6 : read "focuses"
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"
48. p. 365, l. 23 : read "the remote SH still underestimates MOPITT..."
49. p. 365, l. 25-26 : replace "one would expect to infer higher biomass burning emissions" by "higher inferred biomass burning are expected"
50. p. 366, l. 3 : read "emissions increase by 75 Tg"
51. p. 366, l. 4 : read "compensated by decreased"
52. p. 366, l. 15-19 : this point should be discussed earlier (e.g. at p. 359 line 1)
53. p. 366, last paragraph : overstatement - should be omitted
54. p. 367, l. 4-5 : read "...from NOAA. The posterior simulation..."
55. p. 367, l. 14 : read "...have been evaluated against non-assimilated"
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"
57. p. 368, l. 2 : replace "show that it is possible" by "illustrate the capability"
58. p. 368, l. 9 : replace "inversion. This shows that" by "inversion, indicating"
59. p. 368, l. 12 : read "study using different fire injection heights"

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