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Biogeosciences Discussions

Interactive comment on "African CO emissions between years 2000 and 2006 as estimated from MOPITT observations" *by* F. Chevallier et al.

F. Chevallier et al.

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We wish to thank the referee for his most detailed and helpful comments. We address all the issues he/she has raised in the following. The full reviews are copied hereafter and our responses are inserted where appropriate.

> I like the concept of this paper. Simply put, the approach assumes that any CO in the atmosphere is seen by MOPITT. A further assumption is that if one "feeds" a global model with the correct amount, time, and location of the CO emissions, and properly models transport and secondary production and losses, the model will produce a global CO distribution that is in agreement with the MOPITT CO. With these assumptions in mind, three important advanced features of this work are: (i) account for some sec-

ondary CO due to VOC oxidation, (ii) check agreement of the modeled, global CO field also by comparing to selected ground-based stations, and (iii) constrain the model OH (and thus CO losses) with methyl chloroform measurements.

However, I do see potential to improve this paper in 3 basic ways. (i) As written, the performance of the processes employed is not always clear to the average reader. (ii) A more generous assessment of uncertainty is probably appropriate. (iii) The figures and tables require work.

1. Some things that are unclear as presented. I likely represent a typical reader and I found this paper unusually difficult to understand. It is very brief and may assume a bit too much technical background on the part of the readers. It tends to describe the model with jargon instead of telling the reader how the different elements of the model affect the overall accuracy.

We hope that the revised version broadens the audience of the paper.

One example is given here and the rest follow in the specific comments later.

On line 11 of page 3851, it is stated that there are 1,870,000 variables to tweak to fit 950,000 observations. The famous mathematician John von Neumann once claimed "With four parameters I can fit an elephant. With five, I can make his trunk wiggle!" By comparison, this illustrates that a mere listing of the number of variables and observations can make the average reader wonder how well the model is constrained by the observations? It would be better to make it clear to the reader what the constraints are on varying each parameter and if the system is in fact over-determined, under-determined, none of the above, etc... ?

We added the following sentence at the end of the paragraph: "With twice as less

BGD

5, S2245–S2259, 2008

observations as variables to infer, the inversion problem is clearly underconstrained: the prior guess x_b regularizes it".

2. Thinking about the underlying assumptions in this work causes me to question the currently reported level of accuracy. Underlying assumptions in no particular order include:

2A) All the CO in the atmosphere is detected by MOPITT. The MOPITT validation paper (Emmons et al. 20004) shows a low bias in CO columns, which is an impressive result. However an examination of Figure 2 in that paper shows that the aircraft vertical profiles used for validation probed conditions where the mixed layer (ML) CO averaged about 100 ppb and did not exceed 250 ppb. In fact most of the CO produced by fires is initially stored in highly polluted mixed layers containing CO in the range of ~400-1200 ppb and mostly below the altitude of 3300 m, which is the lowest altitude that MOPITT is sensitive to. This is supported in more detail next. First, however, the fact that MOPITT has not yet been validated against situations that are typical for biomass burning regions, combined with the one-month lifetime of CO in the tropics (Mauzerall et al. 1998) does raise the possibility that some fire CO is missed entirely by MOPITT. A related point is that the selected ground-based stations are quite distant from the source regions and may not probe air masses until after much of the fire CO is gone. This implies that the available ground-based stations may not be that sensitive to fire CO and in any case; any error in the transport or secondary chemistry could throw off the estimate of fire CO. Details on the vertical profiles now follow, but the disinterested reader can skip to the point number 3 if desired.

2A.1. CO from biomass burning (BB) in Brazil. Andreae et al., (1988) shows Amazon dry season ML CO ranging from 150-600 ppb (their Fig. 4). Kaufman et al., (1992) reported ML CO ranging from ~150-600 ppb

BGD

5, S2245–S2259, 2008

(their Fig. 4). Blake et al., (1996) observed ML CO values from ~100-400. Reid et al. (1998) observed average ML CO values for flights based in central Brazil ranging from 440-760 ppb (their Table 1). Yokelson et al (2007) observed ML CO in the 200-600 ppb range over the Amazon basin (their Fig 4). They also reported higher levels near 1350 +- 1150 ppb at ground level in afternoon. Finally, a huge regional plume covering 5 degrees of latitude had CO values from 400- 1200 ppb at 2 km elevation. Most of the CO in all the Brazilian studies was confined to elevations below ~3000 m.

2A.2. CO from BB in southern Africa. The fires occur at higher ground elevation and the CO eventually extends to higher elevation. However, before noon much of the CO is trapped in temporary stable layers lower in the ML. After noon the ML is better mixed and elevated CO can extend to 6000 m with typical ML values again approaching 600 ppb (Yokelson et al., 2003 their Fig. 1).

2A.3. Transport of CO from BB regions. The BB CO is transported away from southern Africa by two chief, fairly narrow pathways. One pathway is off the coast of Namibia and features CO at altitudes (3-5 km) that are somewhat favorable for detection by MOPITT (Fig. 1c, Yokelson et al. 2003; or, using aerosol as a proxy for CO, see Fig 12, Schmid et al., 2003). The other major pathway is the "River of Smoke", which involves the smoke exiting off the SE coast of Africa at ~1000 m where MOPITT is less sensitive (Sect. 4.2.3 and Fig 8b, Stein et al. 2003; Figs 5 and 6, Magi et al, 2003; Fig 1, Swap et al.,). Much of the CO from South American BB also exits the continent over the SE coast at low elevation (Fig. 8 Yokelson et al. 2007). Given the one-month tropical lifetime of CO many fire-impacted air masses may be significantly depleted in CO by the time they arrive at either a ground-based station or an elevation for which MOPITT has high sensitivity. Again, a strong point of this system is that it contains a treatment

BGD

5, S2245–S2259, 2008

of CO transport and secondary chemistry. On the other hand, a finite possibility remains that significant amounts of fire CO could be unseen initially and then transported to areas where it is not detected by a ground-based station used in this system nor by MOPITT.

We thank the reviewer for his extensive and well-documented comment. We added the following paragraph in the conclusion to account for it: "The CO budget in Africa is dominated by fires. Fresh tropical biomass burning plumes are usually confined in the boundary layer (e.g., Labonne et al. 2007) and may leave the African continent at low elevation (Garstang et al. 1996). By the time the plumes reach the free troposphere, they may be well depleted in CO (Mauzerall et al. 1998). As a result, biomass burning induces large gradients in the CO vertical profiles over tropical lands (e.g., Yokelson et al. 2003, Yokelson et al. 2007). Depending on the thermal contrast at the surface, the MOPITT instrument has some sensitivity to lower tropospheric CO (Deeter et al. 2007), but cannot capture these steep gradients. Further, the early overpass of the MOPITT spacecraft during daytime (1030 LT) does not favor the observation of the boundary layer, that develops mostly in the afternoon. Therefore, MOPITT provides coarse spatial information about the CO emissions."

2B) The a-priori CO emissions are known to approximately 40%. Current estimates of burned area, or hot spot number, or total biomass burned can differ between studies by factors of 2-4 or more (D. Roy, private communication, Scholes et al. 1996). There are good reasons for this. For instance, we are not sure if understory fires are detected and cooking fires are a major source that is definitely not detected from space and hard to quantify. Thus, I am surprised by the implication in Table 1 (data from Van der Werf) that the CO emissions from fires in Africa are known to 40%. This is important because the authors state on lines 12-13 of page 3856 that the analysis they use tends to pull the final value towards the initial

BGD

5, S2245–S2259, 2008

value. In fact, the final values and initial values are not significantly different according to the data in Table 1. If the model assumed a broader range of a-priori emissions and/or imperfect detection efficiency of both MOPITT and the ground-based network, could a different final result be obtained? If so, could the new result be closer to the real global situation?

We added another sensitivity test among those described in Section 3.2. In this new experiment, we multiplied our prior errors for CO by four. The results differ by less than 8 Tg in each one of the 2 regions. However, since the reviewer points at the arbitrary assignment of our prior errors, we removed the error estimate from Table 1 and inserted a warning in Section 3.1 when we show the expected uncertainty reduction: "These figures should be taken with caution since the prior errors have been assigned rather arbitrarily. Further, some systematic errors are known to affect the MOPITT level 3 products (Emmons et al. 2008) and could also bias the inversion estimate". Last, we inserted a specific paragraph in the conclusion: "The errors of the prior emissions have been empirically assigned. The inversion results would clearly benefit from a rigorous investigation of the statistical characteristics of the emission inventory errors. However, such a task will be complicated by the high variability of the CO emissions in space and in time, that makes the error distributions diverge from normality".

2C) A related point that I did not see explicitly discussed, is how error in the primary CO emissions from regions other than Africa (e.g. Brazil, Australia) could affect the error in the Africa estimate.

We added the following sentence in the conclusion: "[The results for the rest of the globe are still being analysed]. Their quality influences the present results over Africa to some extent, since they implicitly provide boundary conditions for the regional inversion problem". We prefer not to speculate on the impact.

BGD

5, S2245–S2259, 2008

Interactive Comment

2D) Secondary CO from VOC

Including CH4 and HCHO oxidation is important, but a few points could be considered. CH4 is the main species added and work by Chatfield et al (1996) suggested that the main source of CH4 in southern Africa was not fires, but transport from the NH. Now that cooking fires in the region are known to produce more CH4 than previously thought (Bertschi et al., 2003); fires may be the main regional source of CH4. It is unclear and hopefully this complex situation is treated reasonably well in the author.s global model. Also the NMOC are present in much greater amounts than CH4. For example in a recent, comprehensive study of tropical forest fire emissions the CH4/CO mass ratio was ~0.055 while NMOC/CO mass ratio was ~0.46 (Yokelson et al. 2008). The NMOC are much more reactive and may affect the CO levels more on an annual basis. The chemistry of the NMOC is not well-known yet (Trentmann et al., 2005) and about half the VOC emitted globally are still unidentified so there is uncertainty due to this. Finally, not discussed is the affect of any error in the assumed global emissions of VOC by vegetation (~1000 Tg/yr) or BB (~500 Tg/yr) (Yokelson et al. 2008). In fact, I found no explicit mention in the paper of biogenic emissions, which are a huge global source of CO. As a regional issue, large areas of southern Africa are covered by Mopane trees, which are "high emitters" (Greenberg et al., 2003).

Indeed we did not mention the emission inventories for species other than CO, or those from biogenic sources. We added the following sentence in section 2.5: "Prior emissions for the other species, like non-methane volatile organic compounds, or from biogenic origin, follow the inventory gathered for the full chemistry-transport model LMDZ-INCA (Folberth et al. 2006)". The uncertainty in the chemistry justifies our generous observation error (end of section 2.3)

BGD

5, S2245–S2259, 2008

3. Additional comments in order of appearance. P = page, L = line number

P3846, L2: add "'s" to "Earth"?

We did it.

P3846, L21: "economical" should be "economic"

We did it.

P3846, L22: "until now" can be omitted or replaced with "so far".

We did it.

P3847, L7: "high-peaking channels" is jargon

We removed it.

P3848, L5: The atmospheric lifetime of CO in the tropics is closer to one month in some work (Mauzerall et al., 1998).

We corrected it.

P3848, L15-24: Is the impact of these simplifications on system accuracy discussed anywhere?

It is the topic of a specific paper (Pison et al. 2008, ACPD, in press).

BGD

5, S2245–S2259, 2008

P3849, L5-16: It is not clear in this section where the basic biomass burning emissions inventory and speciation come from. It is also not clear here if biogenic emissions are considered. Nor is it clear if HCHO from isoprene oxidation is included in the model.

The sources are those used for the full LMDZ-INCA model and include biogenic sources and isoprene (see above).

P3849, L23: Are the info sources unbiased? Or is the MOPITT CO perhaps a lower limit? Would that affect the error estimate?

We added a sentence about MOPITT biases at the end of section 2.3 "MOPITT biases are neglected even though there is evidence of some systematic errors in the products (Emmons et al. 2008)", at the end of section 3.1 "Further, some systematic errors are known to affect the MOPITT products (Emmons et al. 2008) and could also bias the inversion estimate" and in section 3.2 "Note that some drift in the MOPITT product errors may affect the interannual variations of the inverted emissions (Emmons et al. 2008)."

P3850, L18-23:
Is the model run at 3 h resolution and then averaged to 8-day resolution to compare to the daily "morning MOPITT" averaged to 8-day resolution? The process is not clear here.

The last sentence of the paragraph has been rewritten to clarify this point. It now reads: "Emissions within any of the eight-day periods are interpolated in time from the state vector x to a 30-minute resolution before being transported in LMDZ-SACS."

P3851, L21: "than" should be "as"

BGD
5, S2245–S2259, 2008

Interactive Comment

We did it.

P3852, L10: The 1030 LT MOPITT overpass is before the mixed layer is developed to its full eventual height. The 2230 LT overpass is after the mixed layer has collapsed. Neither is optimum for detecting ML CO below 3300 m.

We added the following sentence in the conclusion: "Further, the early overpass of the MOPITT spacecraft during daytime (1030 LT) does not favour the observation of the boundary layer, that develops mostly in the afternoon".

P3852, L20: I think "estimates" might be truer than "quantifies"

We replaced the word as suggested.

P3853, L1-4: Good! You are comparing the model only to the lower altitude part of the 1030 LT MOPITT information if I understand correctly.

Yes.

P3853, L9-10: Please check my math. I calculate that 5000 MOPITT spots at 484 km2 per spot accounts for less than 1% of the Earth.s surface. Though a lot of scenes are used it seems to say that there is not global coverage? I could be misunderstanding what is meant by a MOPITT spot? It is a good idea to specify here about how much of the global atmosphere is used to compare the model to MOPITT. I am guessing a larger percentage coverage would yield a more accurate result?

BGD

5, S2245–S2259, 2008

The MOPITT data are sampled at the model resolution, as described in the second sentence starting from the end of the paragraph. The model grid covers the globe with about 7000 boxes. Keeping more data may deteriorate the results because of correlations in the observation errors (as defined in the next paragraph in the paper). We expanded the sentence in the following way: "This screening keeps between 2500 and 5000 MOPITT spots per day in the inversion system, to be compared with the 6900 $3.75^\circ \times 2.5^\circ$ boxes of the LMDZ-SACS global horizontal grid".

P3854, L21-22: again, biomass burning and fossil fuel are explicitly mentioned here, but are biogenic emissions considered? It is my understanding that they are an important part of the global CO cycle.

We added the information (see above).

P3855, L4-8: It may possible to clarify this sentence.

The sentence has been simplified and now reads: "The correlations of the prior errors have been assigned for all variables following what had been done for CO2 by (Chevallier et al. 2007): temporal correlations are neglected and spatial correlation are defined by a relatively short e-folding length (500 km) over land and a larger one (1000 km) over the ocean".

P3856, L1-3 and elsewhere: Providing each year's monthly average is a little confusing when most of the biomass burning occurs in ~4 months.

We replaced the monthly figures by annual ones.

P3856, L22: omit "an"

BGD

5, S2245–S2259, 2008

We did it.

P3857, L13-14: It says maybe the system cannot separate primary and secondary CO. What is the implication of that? What are the relative sizes of those terms in your model?

We completed the paragraph with the following sentences: "In other words, errors in the atmospheric production may alter the inversion increments. However, the atmospheric production in LMDZ-SACS amounts to about 10 Tg per month in NHA and to about 5 Tg per month in SHA, which is already smaller than the emission values during the fire seasons (Fig. 4). The errors in the modeled atmospheric production are expected to be even smaller".

Section 3.3: The optimum fit to MOPITT also improved the fit to the surface stations, which is good. Was there also an optimum fit to the surface stations? If so, how did that effect the agreement with MOPITT?

The optimum fit to the surface stations is shown in Pison et al. (2008). It brings the model closer to MOPITT indeed.

Also, why not use more ground-based stations?

In the present paper, we used the unfiltered data that were available on international databases. More unfiltered data exist (at least from station CPT) and we will ask the PIs for them in the future.

P3858, L3 and L16: the ability to treat reactive gases gets much more attention in the conclusions relative to its contribution to the body of the

BGD

5, S2245–S2259, 2008

paper. Perhaps, it should be mentioned here briefly and mostly as a source of future applications.

The last paragraph answers one of the points raised by the second reviewer, so we left it. However, since we added two new paragraphs in the section, the last one has less importance in proportion now.

P3858, L4: was M1QN3 mentioned in paper? Should it be explained here?

M1QN3 is the minimizer. It is defined in section 2.2

Table 1: Tg CO/ month varies over the course of the year. It needs to be clear what is shown in the table entries. Is it the average Tg CO/month for a 12 or 13 month period?

We are grateful to the reviewer for having spotted the mistake in the legend. The values are given per year and not per month.

Table 2: Is model minus observation always positive or is the absolute value shown? Are the entries the mean difference averaged over 7 years? A reader should be able to look at the table and know what is being presented; or be directed to text that explains the entries.

The signs were missing. We added them and specified the period (7 years). Note that the signs are all negative.

Figure 3: The axes should be labeled Latitude and Longitude?

BGD

5, S2245–S2259, 2008

We added the information in the legend.

The mean CO concentrations are probably in "parts per million" not "particles per million".

We did it.

Finally, it is important to state what elevation or elevation range the monthly average was computed for.

We added the information (700hPa).

Figure 4: For the top row: the spatial and temporal units for the CO average need to be given. For the bottom row: the time period needs to be indicated in the y- axis label. E.g. is it Tg CO/month? Per 8 day period?

The temporal resolution is the month. We added the missing information.

Summary:
Without answering all the difficult questions I have posed, the paper could still be improved with a more generous assessment of error that is described in a more broadly accessible (less technical) fashion. At least a list of the main underlying assumptions and the areas where more research is needed would help. Otherwise, the reader is left with the impression that we know the CO emissions from Africa to +-30%, which may not be true yet.

We hope to have improved the quality of the presentation with the above- detailed modifications.

BGD

5, S2245–S2259, 2008

Similarly, I am uncomfortable with the assertion in the abstract (lines 14-15), and elsewhere, that this work proves that "quantitative" fire CO emissions can be derived from MOPITT.

We have removed the assertion.

Interactive comment on Biogeosciences Discuss., 5, 3845, 2008.