

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

Anonymous Referee #1

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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 secondary 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)

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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. 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, underdetermined, none of the above, etc. . . ?

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

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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 (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

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(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 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.

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 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?

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

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error in the Africa estimate.

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).

3. Additional comments in order of appearance.

P = page, L = line number

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

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

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

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

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

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

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.

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

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.

P3851, L21: “than” should be “as”

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.

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

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.

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?

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.

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

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.

P3856, L22: omit "an"

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?

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? Also, why not use more ground-based stations?

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

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

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?

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.

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Figure 3: The axes should be labeled Latitude and Longitude? The mean CO concentrations are probably in “parts per million” not “particles per million.” Finally, it is important to state what elevation or elevation range the monthly average was computed for.

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?

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 $\pm 30\%$, which may not be true yet. 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.

References in brief form:

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