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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of surface emissions to the zonal variability of tropical tropospheric ozone and carbon monoxide for november 2004" *by* K. W. Bowman et al.

Anonymous Referee #2

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General comments:

This manuscript aims to show that the observed tropical distributions of carbon monoxide and ozone are determined by factors including the distribution of surface emissions, weather/dynamical patterns, and injection of nitrogen oxides associated with lightning. The topic is relevant and the effort to fold available satellite observations into the analysis is valuable. However in its current state I believe there are significant flaws in the approach taken in the manuscript. In particular, significant emphasis is placed on interpretation of the differences between two chemical transport model simulations that differ in their surface emissions: one simulation with climatological emissions and

one with 2004 emissions constrained by inverse modeling using satellite CO observations. The inverse modeling work is presented in another manuscript currently in review (Jones et al., 2007). Jones et al. (2007) state that the primary reason for the differences between the climatological and 2004 emissions estimates is biomass burning. This manuscript mentions (in several places) biomass burning as a major source of emissions in the region under investigation, however does not clearly state that, fundamentally, it is differences in biomass burning emission estimates that are being evaluated.

Given this context, I really do not see the significance in this paper of so extensively showing model results with climatological emissions. It is suggested (Table 1 and its discussion) that the inverse modeling results in regional scaling of the emissions climatology. The net effect in the current study is to account for the well known (e.g., van der Werf et al., 2006, cited in the manuscript) interannual variability in biomass burning. The model results would be expected to compare poorly with observations without some attempt at such an accounting.

Compounding this analysis approach is the sampling pattern of the TES instrument. In the 13 day period analysed here, significant aliasing of synoptic structure is inherent. The plots shown as a function of longitude are actually convolved functions of longitude and time (i.e., the dates of individual orbits). The relatively narrow features in longitude shown in Figure 12 are quite likely to result from aliased sampling of plume structures (both by TES and in the model). As far as I can discern from the current manuscript and Jones el al. (2007), the inverse modeling approach has not altered the spatial or temporal (i.e., daily) distributions of biomass burning emissions within the broadly defined regions defined in Table 1. Section 5.4 presents other chemical species from the model to support a discussion of possible chemical and dynamical explanations for the remaining differences between TES observations and the model prediction. As written these processes are largely speculative and need to be supported by, for example, model statistics relating to the ozone budget (including photochemical production and

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convective mixing). More seriously, this portion of the analysis avoids the quite likely possibility that the biomass burning emissions in the model are still not sufficiently accurate for synoptic evaluation against individual orbits of TES observations.

I feel significant work is required prior to publication of this analysis. Major suggestions include the following. 1) Support the speculations made in section 5.4. One possible way is to include model derived estimates of the contributions to the budgets of ozone (and, if necessary, NOx, HNO3, PAN) due to photochemistry, convective mixing, and transport (including transport of ozone from the stratosphere). 2) Evaluate the constrained emissions with respect to an independently derived set of biomass burning emissions such as the GFED version 2 emissions (available as described in the van der Werf (2006) reference.) While these emissions are also less than perfect, they are also to some extent constrained by observations. If significant differences exist in spatial distribution or magnitude of emissions, they must be addressed. One possible approach is to conduct and analyze a model simulation using these emissions. 3) Eliminate most, if not all, of the comparison with the a priori emissions. I see little value in confirming that model simulations with inaccurate emissions leads to poor comparison with observations. Instead consider including comparison with another simulation that includes biomass burning emissions estimates valid for the period, as in item 2.

Specific Comments:

Title: I suggest the word "Structure" rather than "Variability" as variance is really not shown.

P1514 discussion of Figs 3 and 4: Fig 3 shows generally larger ozone values in the southerly band (15-30 S), you should comment on this. The 2 sonde stations that you point out as lying in this band (Pretoria, Reunion) also have larger ozone values than the other stations. They appear as outliers in Fig 4. I believe it would help to plot them separately, i.e., separate Fig 4 into 0-15S and 15S-30S bands as for Fig 3. It will be a sparse plot so some other approach may be better. Perhaps it makes sense to overplot

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the sonde data on Figure 3, in appropriate latitude bands? Also, please use the same data ranges and color scales.

P1515 L20: "could expect" - This statement is largely irrelevant unless you can quantify it somehow. The production of ozone will occur during the time following injection of NOx so is not necessarily collocated with regions experiencing lightning.

P1516: As written, the OMI NO2 data really are not contributing to this analysis. If you disagree, please quantify and clarify the contribution. If you agree, Figure 6 and its discussion may be dispensed with.

P1517: As discussed in the general comments, the model results with climatological emissions are not relevant for comparison with the observations. Regarding Figures 7 and 9, it would be more useful to show the model results with your best estimate of emissions, the same model results as viewed by TES, and the difference field between TES and the model as viewed by TES (as is done for ozone in Fig 12(b); why not do the same for CO?). If you really feel the difference field is relevant (Fig 7b) then state its value, but in any case show the model result with best emissions rather than with climatological emissions.

P1517 L17: Emissions associated with what sources, specifically? Biomass burning or all sources combined?

P1517 last paragraph: You should make comment with respect to the TES sensitivity. For example, much of the surface change is not likely to be apparent in the TES observations. To what extent is TES sensitive to the free tropospheric changes?

P1518 L12: "NOx emissions were scaled" - Was this a linear scaling? Such scaling is probably justifiable if you are assuming that the emissions and change in emissions is due to biomass burning. If there are other emissions sources, the contributions will not scale linearly. Also, what about emissions of other relevant species, such as hydrocarbons and aerosol (which presumably would alter the photochemistry as well)?

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P1518 Figs 12 and 13: Please be clear how you are calculating these increments (model minus observation, or the other way around). I think I have sorted it, but really shouldn't have to make the effort. You comment on the model "differences" but please try to be a bit more specific. Without directly stating such (unless I have missed it) it seems your point is that the model with climatological emissions underestimates free tropospheric ozone and that this underestimate is somewhat reduced through use of the improved emissions. I would also suggest that these figures be plotted as (model minus TES) so that the observation is the point of reference and, for example, model low biases will appear with a negative sign.

P1519 L5: What do you mean by "background processes"? Can you give examples?

P1519 L6: Other possibilities include that the emissions estimates of other species are deficient, including hydrocarbons and aerosols; biases associated with convective redistribution (as the vertical profiles of ozone and CO are different); and biases associated with the transport of stratospheric ozone.

P1519 L15: The statement "can not be explained by surface emissions" is not substantiated. You have only shown they are not sensitive to your choices, i.e., scaling of the climatological emissions, which is certainly not a comprehensive evaluation of emissions. This statement is also made in the Conclusion (P1563 L5).

P1520 L1:It will also be sensitive to photolysis and therefore the presence of cloud (and potentially of aerosol as well).

P1520 L22: This is speculative, as the concentrations in the upper troposphere are a combined response to photochemistry following direct injection aloft and convective redistribution over regions with surface emissions. In-situ ozone production can not be assumed.

P1520 L25: This argument is consistent with the point made with respect to L22. You may want to consider showing diagnostics of something such as convective mass flux

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to support an argument of longitudinally preferred convective redistribution.

P1520 L26-29: You need to support this statement either with your own figures or appropriate citation.

P1521 L1: "100ppt" is not evident in Fig 14b. Perhaps 50 ppt?

P1521 L8-13: Again, this statement is not supported by something shown in the paper or cited in the literature.

P1521 Final paragraph: Again, this argument seems conjecture. To really demonstrate this you would need to show and evaluate the tendencies controlling the ozone (and quite likely the nitrogen) budget, including photochemistry, convective redistribution, and large scale transport.

P1522 L5: "investigated the processes controlling the zonal distribution" - I do not believe that you have done this. You have simply shown the changes resulting from scaling the emissions, and speculated on the causes of remaining inconsistencies between the model prediction and observations.

Figures: please use the same ranges/scales on the ozone figures, e.g., 0-140 on figures 3(b) and 10(a).

Technical Comments:

P1507 L25: "measurements, and chemistry"

P1507 L26: delete "will"

P1510 L13: correct "differences... is"

P 1513 L21: do you mean "extending eastward into"

P1513 L24: "continental biomass burning emission sources"

P1514 L5: "averaged longitudinally in 15"

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P1514 L7: "high in mid-tropospheric ozone"

P1514 L23: "elevated mid-tropospheric ozone"

P1517 L26-27: "over the Indian Ocean" is repeated

P1520 L4: "200 ppb" - do you mean 200 hPa?

P1520 L7: "150S" - do you mean 150 West?

P1520 L8: "principle" - I believe you mean "principal".

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 1505, 2008.

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