

# ***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.***

**K. W. Bowman et al.**

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## **1 General**

We appreciate the reviewer's thoughtful comments. We have made a number of changes that have strengthened the paper and incorporated many of the reviewer's recommendations. We point out here some of the major changes to the text:

- We have added a comparison between GEOS-Chem NO<sub>2</sub> columns with *a priori* and *a posteriori* emissions to the OMI NO<sub>2</sub> columns.
- An ozone sensitivity analysis with lightning turned off has been added to show

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patterns of significant influence from lightning sources.

- Comparisons of GEOS-Chem with updated emissions are compared with the SHADOZ ozone sonde measurements.

## 2 Reviewer 2

### 2.1 General Comments

*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.*

The reviewer points out that significant interannual variability of biomass burning is well-known. The reviewer goes on to indicate that the model should be expected to compare poorly with observations without such accounting. We agree on both counts. However, actually quantifying what the contribution of surface emissions to CO and ozone for this time period is key, and is the basis of this investigation. Moreover, the framework by which one “accounts” for surface emissions is both critical and non-trivial. This is the first paper to use TES/MOPITT CO observations to constrain the emissions estimate while using TES ozone profiles to test the response of the model to those estimates. When this project was started, the GFED2 inventory had not been incorporated into GEOS-Chem. Yes, we could have later used the GFED2 emissions as our a priori, but it was not necessary to do so. This research goes beyond what has been done by van der Werf during this time period and also beyond previous top-down emissions

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studies, e.g., Arellano et al, 2006, that do not examine the response of ozone changes in emissions.

The key point of this paper is to show the response of tropospheric composition to a change in the surface emissions, and that this change improves agreement with the ozone data. By examining how NO<sub>x</sub>, CO, PAN, ozone, etc. change between the climatological mean and the a posteriori emissions, we are able to better understand what processes are driving ozone formation during this time period. We could have chosen another inventory such as van der Werf, but the resulting analysis and conclusions would be the same.

*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 et 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. Yes, the approach does not alter the temporal distribution of emissions, or the spatial distribution within the regions. Since CO is more or less a passive tracer over this time period, errors in the model's calculation of synoptic structure would show up in the residual error between Figures 6 and 7(b) or Figures 6b and 7b in Jones et al. However, we see that after the a posteriori CO emissions estimate, there is generally good agreement between the GEOS-Chem and TES/MOPITT. Consequently, there does not appear to be significant error in the emissions estimate introduced from the assumed spatio-temporal distribution of biomass burning within the satellite observational constraints.*

The structure of free tropospheric ozone, however, could be more affected by model synoptic structure error, particularly associated with lightning. This error would be

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manifested in the residual plot in Figure 13. However, based on the considerations from the previous paragraph, this error should not effect the contribution of surface emissions to ozone, which is the subject of this paper.

We have added a new figure that shows the comparison of GEOS-Chem with the SHADOZ ozone sondes. These figures show that synoptic scale events as well as other factors such as lightning emissions, may significantly reduce the accuracy of GEOS-Chem simulations, particularly in the mid to upper troposphere.

*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 convective mixing).*

The goal of Section 5.4 is to investigate the response of GEOS-Chem to changes in ozone and NO<sub>x</sub> distribution from a posteriori emission estimates, which includes processes not captured by the model. The response of the model to emission changes is not speculative, but the interpretation of the residual error does require judgement.

*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, NO<sub>x</sub>, HNO<sub>3</sub>, PAN) due to photochemistry, convective mixing, and transport (including transport of ozone from the stratosphere).*

Such an analysis would be interesting, but is well beyond the scope of the present paper. The focus here is on showing how a better accounting of emissions (based on the inversion using TES/MOPITT) leads to changes in ozone and NO<sub>2</sub> that are more consistent with TES and OMI observations of ozone and NO<sub>2</sub> over Indonesia/Australia.

*2) Evaluate the constrained emissions with respect to an independently derived set*

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*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*

Within the constraints provided by the satellites observations and based on the CO residuals discussed previously, we do not see the need to run multiple a priori emissions. The a posteriori emission magnitudes are relatively insensitive to the choice of a priori. See the companion Jones et al response for more discussion. We now compare our top-down estimates to the bottom-up GFED2 estimates. Work by P. Kasibhatla (pres. comm.) for 2000-2006 and J. Logan using the GFED2 estimates for 2005 and 2006 shows the GFED2 emissions are far from perfect; based on comparisons of GEOS-Chem simulations with MOPITT (P.K.) and TES (J.L.) CO.

*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.*

As discussed earlier, the primary intent of the comparisons with the a priori emissions is not evaluate those emissions but rather as a reference point from which the response of the atmospheric chemical state to changes about that climatology can be assessed.

## 2.2 Specific comments

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

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We will change the title accordingly.

*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 NO<sub>x</sub> so is not necessarily collocated with regions experiencing lightning.*

This text has been changed and includes a figure with the GEOS-Chem ozone plotted with and without lightning. The spatial pattern is consistent with the paper of Sauvage et al. (2007) that shows that the regional contribution of lightning to ozone over Indonesia/Australia is less than that over South America, Africa, and the south tropical Atlantic.

*P1516: As written, the OMI NO<sub>2</sub> 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.*

We have modified the figure to include a comparison with GEOS-Chem NO<sub>2</sub> column with prior and a posteriori emissions. We show that using the a posteriori emissions improves agreement with the OMI NO<sub>2</sub> data.

*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.*

We thought it valuable to compare the the satellite observations with the CO produced from climatological emissions as a reference point for examining the impact of the change in emissions. So this interest in this paper is not so much the absolute value

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of CO but the change is CO between the *a priori* and *a posteriori* emissions. The companion paper Jones et al goes into more detail about CO distribution including the *a posteriori* model fields.

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

All combustion sources are combined for CO: biomass burning, fossil fuel and biofuel. We assume that biomass burning is the dominant source of direction emissions of CO in the tropics The scaling of NO<sub>x</sub> with CO occurs afterwards when we look at the ozone. See the companion Jones et al paper for more details.

*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?*

The sensitivity of TES to changes in the free troposphere and the surface for CO is discussed at great length in the companion Jones et al paper. Estimates of CO at the surface will not be strongly constrained TES or MOPITT, but the free troposphere will because that is where TES has maximum sensitivity. To the extent that GEOS-Chem simulates transport of emissions out of the boundary layer, then these observations constrain the emissions as well. Please see our response to reviewer 1, specific comment #1.

*P1518 L12: "NO<sub>x</sub> 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)?* Based on the MODIS firecounts, it appears that biomass burning is the dominant contributor to surface emissions, particularly over Indonesia. The additional comparison of NO<sub>x</sub> *a priori* and *a posteriori* emissions suggest that there may be an underestimate of NO<sub>x</sub>

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emissions. However, the main point is the implications of this assumption to the ozone distribution.

We've changed the text, however, to clarify the phrasing to: **The ozone distribution from GEOS-Chem was also calculated based on the revised emissions where all emissions, including NO<sub>x</sub> and hydrocarbons (but not aerosols), were scaled with the CO a posteriori emission estimates.**

*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.*

The ordering of the increments was not correctly stated in the text. We have changed the text and added some additional comments to clarify how the mean differences are calculated. An example of the figure text now reads: **Mean difference between (a) TES ozone observations and GEOS-Chem with a priori emissions and (b) TES ozone observations and GEOS-Chem with a posteriori emissions and from 15S to the equator. Mean differences are calculated between the TES observations and model predictions sampled along orbit track within 15° × 15° bins**

*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.*

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These lines have been removed from the text.

*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).*

In P1519 L16-17 we indicated that the reason for the residual differences could be due to the assumed relative distribution of CO, NO<sub>x</sub> and other ozone precursors., which is your primary objection. We have changed the text, and now say that the residual difference may not be related to surface emissions, but may also reflect deficiencies in model transport, lightning NO<sub>x</sub>, etc.

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

This text has been removed.

*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. Our statement was not sufficiently clear. We were simply stating that the additional NO<sub>x</sub> sources are responsible for the zonal variation between the tropical Atlantic and Indonesia/Australia. The text now reads*

**Associated with these higher concentrations of NO<sub>x</sub>, model simulations with also produces more ozone (Figure ??) over South America and sub-equatorial Africa than over Indonesia/Australia.**

*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 to support an argument of longitudinally preferred convective redistribution. The difference between the model simulation with a priori and a posteriori emissions shows*

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the model prediction of the impact of convective redistribution since that is the primary mechanism of moving emissions from the surface to the free troposphere.

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

This text has been deleted.

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

Changed.

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

The text has been reworked: PAN increases over all three continents but is most significant over sub-equatorial Africa ( $>150$  ppt at 200 hPa ) and Indonesia (  $\approx 200$  ppt at 600 hPa). Clearly, there is a significantly different response in GEOS-Chem over Indonesia where ozone, CO, NO<sub>x</sub>, and PAN increase whereas in sub-equatorial Africa and South America ozone, CO, and PAN increase but NO<sub>x</sub> decreases.

*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.*

This text has been reworked. As pointed out by the reviewer, the response of free tropospheric ozone to surface emissions is based on the combined effects of photochemistry, convective redistribution, transport, etc. This analysis shows that combined response through the model simulation constrained by satellite observations. So the "tendency" of free tropospheric ozone to the surface emissions is not based on conjecture. We do not need to decompose that tendency into its constituent terms if our primary goal is to look at the response to the surface emissions.

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*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.*

The text has been changed to read:

**We have investigated the impact of surface emissions on the zonal structure of tropical tropospheric ozone with a focus on the sensitivity of that distribution to changes in surface emissions between South America, sub-equatorial Africa, and Indonesia/Australia for November, 2004.**

*Figures: please use the same ranges/scales on the ozone figures, e.g., 0-140 on figures 3(b) and 10(a). Figures 3 and 11 and 12 now have the same scale.*

## 2.3 Technical comments

*P1507 L25: "measurements, and chemistry"*

changed to

**satellite data, sonde measurements, along with chemistry and transport modeling**

*P1507 L26: delete "will"*

Done

*P1510 L13: correct "differences... is"*

Done

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

Yes. Done.

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*P1513 L24: "continental biomass burning emission sources"*

Done

*P1514 L5: "averaged longitudinally in 15"*

Since they are averaged both longitudinally and latitudinally, we've changed the phrase to

**where the TES observations have been averaged in  $15^{\circ} \times 15^{\circ}$  bins**

*P1514 L7: "high in mid-tropospheric ozone"*

Done

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

Removed

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

Yes. Corrected

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

Yes. Corrected.

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

Yes. Corrected.

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