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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.

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_2 columns with *a priori* and *a posteriori* emissions to the OMI NO_2 columns.
- An ozone sensitivity analysis with lightning turned off has been added to show

patterns of significant influence from lightning sources.

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

2 Reviewer 1

2.1 Specific Comments

The Authors use a posteriori CO surface emissions derived by a companion paper (Jones et al. [ACPD 2008]). I did not find in both papers a discussion on TES limits to derive surface emissions through inverse model analysis. Averaging kernels for O3 (Fig.1) in this paper and for CO in Luo et al. [JGR, 2007] paper clearly show weak sensitivity in the lower troposphere (below 750hPa). I understand model inversion should be able to limit this low sensitivity by considering transpor t from surface to the maximum of TES sensitivity (600hPa according to the Authors). However, it would have been interesting to discuss the incidence of this low surface sensitivity on the a posteriori inventory. Indeed I am concerned that this could be a problem for Africa or Australia, where persistent high pressure systems during biomass burning season tend to block emissions, implying high CO and O3 concentrations below 750hPa (e.g. Jonquiere et al., [JGR 1998] for nor thern biomass burning season over Africa; papers from SAFARI campaign and more recent studies). A good example that illustrates this, is the difficulty of TES to obser ve lower tropospheric high CO mixing ratios simulated by GEOS-Chem (continental surfaces of Fig7a and Fig 8); but also some of the high lower tropospheric O3 mixing ratio seen in SHADOZ (comparison of Fig3a and Fig4 Pretoria and Reunion Island, but also Fig 2 of Nassar et al. [JGR 2008] study). This low sensitivity to lower troposphere may suggest that a posteriori emissions inventor y is low biased and therefore, the response in O3 and precursors would be different. Some

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sensitivity test and at least discussions on this point would strengthen the manuscript.

The reviewer makes excellent and important points concerning the limitations of TES/MOPITT to constrain surface emissions. These satellites measure in the thermal infrared and therefore depend on thermal contrast in an atmospheric layer to derive the atmospheric concentrations. Consequently, they are not sensitive to surface CO or ozone directly . Therefore, this analysis is dependent on GEOS-Chem to correctly describe the vertical transport of emissions in to the free troposphere. All inversions using MOPITT and TES data are subject to this issue. The key issue is does the inversion analysis reduce the bias in the model with respect to independent data? In Jones et al. 2008 we showed that the a posteriori emissions reduced the bias in the modelled CO with respect to GMD surface data at Guam and the Seychelles. In contrast, the bias was exacerbated at Ascension. A similar overestimate in CO at Ascension was reported by Arellano et al. (2006), which they attributed to a bias in the altitude dependence of the transport in the lower troposphere.

Observations of NO_2 from the OMI instrument provides an important additional observational constraint because its wavelength ranges are more sensitive to the surface than a thermal instrument. The comparison between OMI NO_2 and GEOS-Chem with a posteriori emissions is improved relative to the climatological emissions but is still underestimated.

Nevertheless, we must also consider the possibility that the assumed relative change in CO and NO_2 is incorrect. Future research will use the OMI NO_2 directly in the inversion.

We have added the following text in the section "Signatures of lightning and surface NO_x": The assumption that NO_x sources scale with CO was tested by comparing GEOS-Chem *a posteriori* and *a priori* NO_2 columns to OMI NO_2 as shown in Figure **??**(b,c). The spatial distribution of GEOS-Chem generally agrees with OMI but does not capture the enhance NO_2 concentrations in northern Australia, which are consistent with the concentrated MODIS firecounts. The *a posteriori* derived NO_2 are in

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better agreement than the *a priori* with observations but still generally underestimate the NO_2 columns. There are several possible explanations for this discrepancy. OMI observations are more sensitive to surface concentrations whereas TES/MOPITT are more sensitive to the free troposphere. Errors in convection and boundary layer transport within GEOS-Chem could lead to an underestimate of the boundary flux of trace gases into the free troposphere. However, Jones et al. (2008) showed that the a posteriori emissions reduced the bias in the modelled CO with respect to GMD surface data at Guam. On the other hand, we assume here that we have correctly captured the relative changes in NO_2 and CO with the uniform scaling. Given that the inverse estimate did not distinguish between types of sources, e.g., biofuels or biomass burning, we could expect discrepancies between the a posteriori NO_2 distribution and the OMI observations. Nevertheless, in the absence of additional information on the different source types and solving simultaneously for NO_x and CO emissions, uniformly scaling the emissions is a reasonable approach.

2-The resulting outcomes concerning the characterization of processes governing the zonal variability of tropospheric O3 and precursors, especially those in terms of the characterization of the tropical sources and dynamical processes influencing the so-called "zonal wave one", are not entirely new or original (Sec 3, Sec 5.2 and Sec 5.3). Therefore, I would encourage the Authors to focus on findings that are different in comparison to other existing and recent studies. In considering these comments, please pay attention to advances in tropical tropospheric O3 and its precursors, and in the characterization of processes influencing the zonal wave-one pattern, published by Moxim and Levy, [JGR, 2000], Wang et al. [JGR 2006], Sauvage et al. [JGR, 2007 and JGR 2006]. The main result of the paper concerns the influence of surface sources of emissions over Indonesia and Australia, and the authors give a nice explanation in the last par t of the paper (Sec 5.4). The Authors should better focus and discuss this impor tant result, and previous sections of the paper should encourage the investigation of this region, the influence of its sources and their uncer tainties. This last section (Sec 5.4) may also investigate the response of CO to OH.

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It was not our intention to make any new contributions to the zonal "wave-one" pattern *per se*, particularly the high ozone concentrations in the tropical Atlantic. Since TES is a new instrument, we thought it important to show that TES exhibits the zonal wave-one and is consistent with SHADOZ data.We also thought it was important to investigate the high ozone in Indonesia/Australia against the backdrop of the zonal wave-one with appropriate references to previous investigations. **We have adjusted the paper to more clearly emphasize the new results over Indonesia/Australia and de-emphasize the zonal wave-one, particularly in the abstract.** The more recent papers by Sauvage et al have already been cited in the paper. We have included the Moxim and Levy as well as Wang.

3-The methodology used to derive surface NOx emissions should be better explained and discussed as O3 mixing ratio are highly sensitive downwind of lightning NOx emissions. The brief explanation given in the paper suggests that NOx emissions are scaled uniformly according to regional factors obtained in the inversion analysis for CO. An increase in CO emissions factors does not necessar y lead to an increase in NOx emissions factors. Moreover, uniform scaling of all combustion sources should be better discussed and evaluated. The Authors could discuss this approach by comparing a posteriori NOx emission factors for individual sources (biofuels, biomass burning and anthropogenic) to existing estimates derived from a bottom up approach to check consistency.

We agree that NOx emissions need not scale with CO emissions but clearly any emissions from combustion (fossil fuel, biofuel, or biomass burning) also generate NOx and other combustion-related emissions, so we felt it is a reasonable approach to take.

However, we can compare the *a priori* and *a posteriori* NO_2 columns with OMI as a check. **We've added a comparison of GEOS-Chem prior and posterior emissions to the OMI NO2 concentrations. These show that the *a posteriori* emissions improve the comparison, but still underestimate OMI NO2.** This discrepancy could be due to an error in the relative scaling or as implied by the reviewer, an error in the

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model vertical transport, or both.

4-A posteriori emissions are derived from a short period of TES observations (Nov 4-15). It would have been more conclusive to study changes of O3 to emissions over several months. It would be interesting to have looked at the under estimation of surface sources over Indonesia and Australia during a whole season. I understand this would represent a substantive piece of work. Therefore, due to this small sample, conclusions on the O3 and precursor feedbacks to a posteriori emissions should be less affirmative, as strong seasonal and inter annual variability of emissions may be expected over the tropics.

We explained in our response to Jones et al. why we used the data from November, 2004, for this study. There were few global measurements available before and after this period in fall of 2004. Furthermore, in 2005 there was low sensitivity to CO in the TES measurements due to problems with the optical alignment. Clearly, the response of ozone to surface emissions over other time periods and scales will be different, and will be the subject of future work. We have added text to clarify that limitation.

We have added the text: "Given the relatively short time frame for the study, this analysis should be extended to seasonal and yearly time periods to see if these mechanisms are robust over longer time scales" to the conclusions

5-The abstract should be rewritten focusing on the new results derived from this paper. Page 1506 line1: I do not see investigation of dynamical processes governing the zonal variability of O3 and CO in this paper. There are a lot of assumptions in the paper concerning transpor t but these are not demonstrated (eg Page 1515 lines 1-2). The Authors should better reference other studies that have already demonstrated transpor t processes, as this can not be implied with only the analysis of ozone and precursors distributions. Another approach would be to perform transpor t analysis between region of sources and region of receptors with the GEOS Chem model. Page 1506 line 6-8: The so-called "wave-one" pattern showed by SHADOZ network has already been

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demonstrated in Thompson et al. [JGR 2003] study, and is not a new result. Please remove. On the contrary the Authors should highlight the significance of TES to see zonal wave one, which can complete SHADOZ and MOZAIC obser vations, at least in the middle and upper troposphere.

Page 1506 line 20-21: The influence of lightning NOx emissions to explain remaining discrepancies in upper tropospheric ozone is not demonstrated in the paper.

The abstract has been rewritten to focus more on the impact of surface emissions on tropospheric ozone as recommended by the reviewer. Nevertheless, dynamical processes are an important component for understanding the impact of surface emissions on ozone and distinguishing those from other processes. That is why there is a discussion of NCEP and ISCCP products that describe the high pressure system in Australia. We are only describing dynamical processes necessary to help understand the impact of surface emissions.

We have added the following text to better describe the circulation of surface emissions based on our companion paper: the pervasive high values of CO across the Indian ocean are indicative of outflow of continental emissions, which is shown are shown by CO tagged tracers for S. America, subequatorial Africa, and Indonesia/Australia in ? and is consistent with previous studies from the Southern African Fire-Atmosphere Research Initiative (SAFARI), e.g., (?) We have changed the abstract to read "Vertical ozone profiles from the Tropospheric Emission Spectrometer (TES) and ozone sonde measurements from the Southern Hemisphere Additional Ozonesondes (SHADOZ) network show elevated concentrations of ozone over Indonesia and Australia (60-70 ppb) in the lower troposphere against the backdrop of the well-known zonal "wave-one" pattern with ozone concentrations of (70-80 ppb) centered over the Atlantic ..

We removed Page 1506 line 20-21 in the abstract

6-Page 1508 lines 13-15: It is not possible to discuss ozone formation from lightning by

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just looking at LIS flash counts. See remark 14 on Section 4. Please remove or restate

The following text has been added: Comparisons with GEOS-Chem *a posteriori* emissions where lightning is turned off are used to determine its relative spatial contribution to ozone formation.

7-Page 1508 line 17: Marenco et al. [JGR 1998] paper does not concern tropical tropospheric ozone. Reference should rather be Sauvage et al. [ACP 2005]. Please restate **Done**

8-Sections 2.1 and 2.2 are already described in the companion paper. This could be shor tened. We appreciate the reviewer's concern. However, we believe each paper should be relatively stand-alone so some duplication is unavoidable.

9-Page 1511 line 12: I do not believe 600 hPa is the lower troposphere. Below 750-700hPa would be.

changed to ...are centered near 750 mb indicating...

10-Page 1513 line 10-13: Please update with recent studies on the zonal wave-one pattern **Updated.**

11-Page 1513 lines 23-25: I do not see comparison with MODIS fire counts. Please try to be more precise. I do not think additional figure would be necessar y, but the Authors should specify that they checked the consistency between MODIS and CO from TES. **Added " MODIS firecounts are elevated across northern Australia and Eastern Africa (not shown but available at http://rapidfire.sci.gsfc.nasa.gov/firemaps/) are indicative...."**

12-It would be better to merge Fig 3 and Fig 4 for easier comparison. Besides, would not it be possible to make a comparison between TES and SHADOZ for exact SHADOZ locations (shown in Fig4), rather than making an average between 0-15S? This would strengthen the manuscript, if the consecutive collocation of TES and SHADOZ profiles were to show good comparison of O3 mixing ratio.

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It is not straight-forward to directly compare SHADOZ sondes and TES observations because they are make measurements at very different times and locations. Consequently, the primary purpose for the comparison between the two is to see if they provide a similar picture rather than a strict quantitative comparison. *13- Section-4. The purpose of this section is unclear. The Authors can not demonstrate signatures of lightning on O3 distributions by just looking at lightning flash counts locations from LIS. O3 from lightning NOx emissions is not expected to be created close to emissions but downwind, and it is not obvious to conclude with this figure on lightning contribution over Indonesia/Australia compared to South America and Africa.*

We have added a new figure showing GEOS-Chem with and without lightning, which shows the relative contribution of lightning to regional ozone formation. The regional formation is consistent with the spatial distribution of LIS measurements.

What is the goal of using OMI NO2? The Authors seem to use NO2 columns to localize surface emissions. It is quite well known that there are important surface emissions and lightning NOx emissions over the Tropics. I think there is no need to include these figures. The Authors could refer to existing studies with monthly means of NO2 from GOME, SCIAMACHY or OMI (eg, Martin et al. [JGR 2004 and 2006]; Richter et al. [2002]; Boersma et al JGR [2007 and 2008]), and of lightning flashes (Christian et al. [JGR 2003]). An estimation of surface sources influence on tropospheric NO2 columns over Indonesia/Australia compared to South America/ Indonesia would strengthen the manuscript.

We have changed the figure to include OMI NO2 columns along with GEOS-Chem NO2 columns for both *a priori* and *a posteriori* emissions. The purpose of providing OMI data is to map out the distribution of NO2 during this time frame and to show that our assumption that NO2 emissions scale with CO emissions leads to an increase in agreement with the OMI NO2 data.

14- Section 5.1: Would it be possible for the Authors to give some precision of the light-

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ning parameterization used in their simulation? This could have important incidence on O3 distributions seen by the model, as demonstrated by Sauvage et al. [ACP 2007, JGR 2007] and by Hudman et al. [JGR 2007]. What is the intensity of this source in the model? This could be mentioned in a table on annual average, with the intensity of the individual surface NOx sources a posteriori (or individual emission factors of surface sources a posteriori).

The lightning parameterization and magnitudes are based on the standard version of GEOS-Chem for v7-04, which has been described by Martin 2002 and used by Hudman et al, 2007. Sauvage et al. (2007) discuss the limitations of the lightning parameterization in this version of the model. The global source of NOx from lightning in the model was 4.7 TgN/a

15- Page 1519 lines 3-6. The Authors can not assume that "assumptions used in the emissions are incorrect" and investigate in their paper response to changes in ozone and precursors from their a posteriori emissions. Please restate. Investigation of a posteriori NOx emissions factors for individual sources would help understand consistency of their a posteriori emission inventory.

sentence has been removed. The comparison with OMI NO2 columns provides a test on the scaling of NO2 with CO. The observations that we have in place can not really distinguish between individual NO2 emission sources. However, based on the difference in NO2 between GEOS-Chem and OMI, the model agreement is improved but the response is still underestimated.

16-page 1521 lines 6-8. Please add reference (eg Liu et al. [JGR 1987]; Kunhikrishnan and Lawrence [GRL 2004]) **Done**

17- page 1521 line 8-9 "Over Indonesia the dominant sink... (PAN)". Please comment figure 15 at this line if previous statement is deduced from Fig15. This text has been changed to read **PAN increases over all three continents but is most significant over sub-equatorial Africa (>150 ppt at 200 hPa) and Indone- sia (≈200 ppt at**

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600 hPa). Clearly, there is a significantly different response in GEOS-Chem over Indonesia where ozone, CO, NOx, and PAN increase whereas in sub-equatorial Africa and South America ozone, CO, and PAN increase but NOx decreases.

18-Page 1521 line 9-11 "whereas over ...(due to higher levels of OH in these regions)" Is the assumption on HNO3 is demonstrated in this paper or is this a supposition? Please clarify. See response to 17.

19-Page 1521 line 29-30: "importance of background meteorological conditions". This is not demonstrated. Please clarify or remove. The importance of meteorology on the response was shown through a discussion of NCEP winds, ISCCP cloud optical depths, the tagged-CO concentrations between regions in the Jones et al companion paper, and the underlying meteorology used to drive GEOS-Chem. We have not tried to disentangle the relative contribution of meteorology versus in-situ chemistry for each emission source but is clear that meteorological conditions play a crucial in the zonal ozone response.

1-Please improve the quality of figures and try to merge figures which are discussed at the same time in the text (eg comparison of Fig.8 and Fig.9a; comparison of Fig.10a and Fig. 11a ...). O3 and CO unit range should be the same for easier interpretation (e.g between Fig3 and Fig4, between Fig 7a, Fig 8 and Fig.9... and see previous remarks in the specific correction section).

We appreciate the reviewer's comment concerning the organization of the plots. We have attempted as best as possible to merge the figures while trying to keep the plots legible. That has forced us to keep most plots limited to 2 subplots. For example, we had at one point Fig 8 and 9 merged by the plot became either too big or too illegible.

We have made the scale for the GEOS-Chem and TES ozone and CO to have the same scale in order to facilitate comparisons.

2-page 1520 line 4: "and around 200ppb" Please correct Done.

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