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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 #1

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Review of the paper "Impact of surface emissions to the zonal variability of tropical tropospheric ozone and carbon monoxide for November 2004", by Bowman et al.

General comments:

The main objective of the paper is to investigate the influence of surface emissions on tropical tropospheric ozone (O3) and carbon monoxide (CO) for November 2004. TES CO observations are used to determine a posteriori estimates of CO and NOx surface emissions from a companion study (Jones et al. [ACPD 2008]), which are poorly understood over the tropics. A chemistry and transport model (GEOS-CHEM) is used to test a posteriori emissions and investigate the response in O3, CO and nitrogen

species distributions over the southern tropics. In the mean time, TES O3 is compared to the SHADOZ ozonesondes network. I find the paper interesting because there is a clear attempt to use TES observations to better constraint global model surface emissions and to help better understand vertical distributions of O3 and CO over the tropics. There is also an important result that shows surface emissions underestimation over Australia and Indonesia, even if only demonstrated for a short period (Nov 4-15). However, I have several concerns regarding the analysis and conclusions drawn from their work. The conclusions concerning zonal wave-one analysis and tropical tropospheric O3 are not entirely new, and the Authors should better emphasize the new results regarding Indonesia and Australia. The Authors should also better discuss and evaluate their a posteriori inventory and give an estimation of the resulting variability of O3 and CO distributions. I recommend that the paper is revised before publication in the ACP journal.

Specific comments:

1-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 transport 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 northern 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 observe lower tropospheric high CO mixing ratios simulated

ACPD
8, S961–S967, 2008

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 inventory is low biased and therefore, the response in O3 and precursors would be different. Some sensitivity test and at least discussions on this point would strengthen the manuscript. I did not find evaluation of TES CO and O3 vertical profiles over the tropics in the referenced papers of TES evaluation (except Nassar et al. [JGR 2008]). In the future MOZAIC measurements could be a good dataset to evaluate TES vertical profiles over the tropics, especially for CO.

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 part of the paper (Sec 5.4). The Authors should better focus and discuss this important result, and previous sections of the paper should encourage the investigation of this region, the influence of its sources and their uncertainties. This last section (Sec 5.4) may also investigate the response of CO to OH.

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 emis-

ACPD
8, S961–S967, 2008

sions. 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 necessary 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.

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.

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 transport but these are not demonstrated (eg Page 1515 lines 1-2). The Authors should better reference other studies that have already demonstrated transport processes, as this can not be implied with only the analysis of ozone and precursors distributions. Another approach would be to perform transport 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 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

ACPD
8, S961–S967, 2008

TES to see zonal wave one, which can complete SHADOZ and MOZAIC observations, 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.

6-Page 1508 lines 13-15: It is not possible to discuss ozone formation from lightning by just looking at LIS flash counts. See remark 14 on Section 4. Please remove or restate.

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.

8-Sections 2.1 and 2.2 are already described in the companion paper. This could be shortened.

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

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

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 necessary, but the Authors should specify that they checked the consistency between MODIS and CO from TES.

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. Please use same unit range for easier comparison.

ACPD
8, S961–S967, 2008

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.

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.

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

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.

ACPD
8, S961–S967, 2008

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

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.

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.

19-Page 1521 line 29-30: "importance of background meteorological conditions". This is not demonstrated. Please clarify or remove.

20-The Authors should add discussion in that section (Sec 5.4) on the feedback on CO through OH (as a dominant think for CO), as OH is modified throughout the O3 response to a posteriori NOx emissions. Is this response improve or decrease comparison of CO distributions between TES and GEOS-Chem? This would strengthen the manuscript.

Technical corrections

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).

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

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