

We thank the reviewers for their constructive comments, suggestions, and corrections. In this revision, we have addressed the reviewer's concerns. The following is a one-to-one response to their questions.

Response to Comments by Anonymous Referee #2

This is a relatively straightforward analysis of the interplay of meteorological processes and atmospheric chemistry in venting out ozone and ozone precursors from Africa and reaching Africa. The manuscript is well written, the figures appropriate.

As the authors state- there is relatively little literature discussing Africa-to-Asia transport, so this is a welcome addition, despite it doesn't make use of the recommendation in the HTAP2 exercise to harmonize region definitions to allow comparability of results.

We thank the reviewer for the encouragement and for the valuable and thoughtful comments. In Fig. 1, we show the regional definitions of the HTAP2 and in our analysis so the reader can have an idea for the similarity and difference between the two definitions.

A minor remark is that I don't see terribly much added value of the trajectory analysis in figure 12.

The trajectory analysis is improved in this revision from the following aspects. (1) Trajectories in two more seasons, spring and autumn, are added for comparison between the four seasons instead of just two seasons in the last version, (2) trajectories from four more stations in Africa are added for a wider coverage and representation, (3) the mean transport pathways for the trajectories that arrive Asia are illustrated with lapse time indicated, and (4) more discussions are added in the revision on the mechanisms that control the transport of African ozone to Asia in section 3.3.

Although some attempt has been made to demonstrate the model's ability to model ozone over Africa, I think this could be done more convincingly- there is meanwhile a

host of other observations (surface, aircraft, satellite tropospheric ozone columns) that could be explored.

Thanks for the point. In this revision, we add the validation between ozonesonde observations and GEOS-Chem simulations that shows the performance of the model in simulating the seasonal and interannual variability of tropospheric ozone over Africa in the surface layer, lower, middle, and upper troposphere. The comparison at three more ozonesonde stations in India is added. Furthermore, the ozone data from the Tropospheric Emission Spectrometer (TES) satellite instrument are used to evaluate the GEOS-Chem simulation in the middle troposphere (464 hPa) globally. The detailed comparisons are shown in Figs. 2-4 and Tables 1-2 and discussed in section 2.2.

Are signals from African ozone visible in soundings over India?

The transport of air mass from African in summer is reflected in the ozonesonde data at Poona and Thiruvananthapuram in western India (in an added figure, Fig. 4). The effect of the Somali jet on western India is obvious as low ozone concentrations appear in the lower troposphere in NH summer.

The organization and discussion of methods could be somewhat more systematic.

Thanks for the suggestions. The methods are reorganized and the presentation is polished.

I suggest that the authors explore somewhat further these aspects, and recommend the manuscript to be accepted after taking these major and minor comments below into account.

Minor comments.

1. 11-30 the abstract could be somewhat more explicit in describing the regions and attribution methodology.

Thanks. The abstract is rewritten to include the regional description and attribution

methods.

l. 16 Replace boreal by NH winter. Or find better way of describing which months are discussed. Are the > and < really meant to express minima and maxima?

Thanks. We have replaced boreal winter by northern hemisphere (NH) winter in this revision. We have removed the > and <, and used certain numbers to express minima and maxima.

l. 30 I miss some statement on the relevance of this analysis. How much of the Asian ozone was produced in Africa or from African precursor emissions- where is it most important (not only vertical but also geographically).

The sentence has been rephrased for clarity. Vertically, the influence of African ozone is mainly in the middle and upper troposphere. Geographically, the imported African ozone mainly distributes over latitudes south to 40°N in Asia.

l. 35 give reference time to which this RF estimate pertains.

Thanks. The reference time is given: “It also acts as a greenhouse gas, whose global mean radiative forcing is about $0.4 \pm 0.2 \text{ W/m}^2$ for the period 1750-2011 (Myhre et al., 2013)”.

l. 46 add: as well as a range of papers in the HTAP2 (Galmarini 2016) special issue.

Thanks. Galmarini et al. (2017) and several other papers in the HTAP2 special issue are now added in the citations.

l. 53 The issue is also very connected to legislative issues related to the control of ozone and ozone exceedance in the western states of the USA, e.g. as discussed in Huang et al. (2017; already cited).

Thanks. This point is now included in Introduction.

l. 54 One reference on LRT transport between South Asia and East Chakraborty et al.

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Thanks for your recommendation. The reference is helpful to the study and it is added in the citations.

l. 73 ... makes a contribution ... how is contribution defined? Zero out of emissions? This is important because later you present a different method.

We have clarified this term in Introduction. The contribution of the source regions to the receptor region can be presented as absolute or fractional contribution. The former refers to the concentrations of imported ozone in a unit of ppbv, while the latter is the ratio of the imported ozone to the total ozone in a grid, a layer, or a region. We have also rephrased this sentence to make it clear.

l. 117 which resolution is used for GEOS-CHEM; what was the underlying resolution of the assimilation product. Importantly for this paper, how is convection parameterized, is there any evaluation over Africa of these processes. Interhemispheric mixing and similar: refer to any relevant application of the model that demonstrates it is fit-for-purpose for this study. I realize that these are discussed later, but I would have expected these descriptions here.

Thanks for your comment. The detailed descriptions of GEOS-Chem have been moved to an earlier part section 2.1. GEOS-4 uses the schemes developed by Zhang and McFarlane (1995) for deep convection and by Hack (1994) for shallow convection. GEOS-4 meteorology is found to be characterized with stronger deep convection in tropics than GEOS-5 (Liu et al., 2010; Zhang et al., 2011). Liu et al. (2010) and Zhang et al. (2011) have shown good agreement of GEOS-Chem simulations driven by GEOS-4 with satellite observations in tropical troposphere. Choi et al. (2017) compared the simulations of the Global Modeling Initiative (GMI) chemistry and transport model (CTM) driven by three meteorological data sets (fvGCM for 1995, GEOS-4 for 2005, MERRA for 2005) with ozonesonde and TES observations. They found that ozone simulated by GEOS-4 has the highest correlation with the observations. The information about GEOS-4 is also described in Zhu et al. (2017b).

We have not performed direct evaluation on the convection parameterization schemes. Nevertheless, these previous studies and the good validation results over Africa from this study provide us confidence on the model performance. We have revised the manuscript to explain the model's strength and limitations.

1. 125- If I understand correctly the authors merge the EDGAR3.2 global inventory with regional ones. Which period? How do these inventories compare with e.g. the HTAP2 inventory for 2008/2010 in this special issue, or EDGAR4.2 products (for time series).

We do the full chemistry simulation in GEOS-Chem to generate ozone production and loss data in 2005 with the emission data from the global EDGAR 3.2 inventory for 2000, the INTEX-B Asia emissions inventory for 2006, the NEI05 inventory in North America for 2005, the EMEP inventory in Europe for 2000, the BRAVO inventory in Mexico for 1999, and the CAC inventory in Canada for 2005. This part has been described more in section 2.1.

The comparison of the inventories used in the study with the HTAP2 inventory for 2008 is shown in the supplement file for CO and for NO_x. Compared to HTAP2 inventory for 2008, the CO emissions used in GEOS-Chem is higher in North America, Europe and East China, and lower in Africa and Southeast Asia throughout the year. The NO_x emissions are higher in North America and Europe, and lower in South Asia.

The contribution of anthropogenic emissions to African ozone is smaller than that of other emissions. Aghedo et al. (2007) estimated that anthropogenic emissions emitted in Africa account for approximately 11% (4.7Tg/42.8Tg) of the African emissions influencing the global tropospheric ozone burden. Therefore, the slight difference in the anthropogenic NO_x and CO emissions over Africa between the two may have little impact on our analysis.

1. 135 What is the global lightning source strength and specific for Africa. How does this compare to other studies.

Lightning NO_x emission used in the study is shown by annual mean and in each

season in the supplement file. The annual global lightning NO_x source amount is $5.97 \text{ Tg N yr}^{-1}$, comparable to $6 \pm 2 \text{ Tg N yr}^{-1}$ in Martin et al. (2007) and 6.3 Tg N yr^{-1} in Miyazaki et al. (2014). Miyazaki et al. (2014) estimated the annual global lightning NO_x emission by assimilating observations of NO_2 , HNO_3 , and CO measured by OMI, MLS, TES, and MOPITT into the global chemical transport model CHASER. The annual lightning emission is $1.72 \text{ Tg N month}^{-1}$ in Africa, $0.80 \text{ Tg N month}^{-1}$ in NHAF, and $0.79 \text{ Tg N month}^{-1}$ in SHAF, shown in the supplement file.

Reference: Martin, R. V., Sauvage, B., Folkins, I., Sioris, C. E., Boone, C., Bernath, P., and Ziemke, J.: Space-based constraints on the production of nitric oxide by lightning, *J. Geophys. Res.*, 112, D09309, doi:10.1029/2006JD007831, 2007.

1. 137 briefly describe what is the ‘standard’ tagged ozone method. Pro’s and con’s-limitations. Comes now later

The tagged ozone method tags ozone by the region where ozone is generated. The method was first proposed by Wang et al. (1998) and then developed and used by a number of studies on ozone transport (Fiore et al., 2002; Sudo and Akimoto, 2007; Zhang et al., 2008; Liu et al., 2011; Sekiya and Sudo, 2012, 2014; Zhu et al., 2017b). It is done by the following two steps. First, the production rates and loss frequencies of odd oxygen ($O_x = O_3 + \text{NO}_2 + 2\text{NO}_3 + 3\text{N}_2\text{O}_5 + \text{HNO}_3 + \text{HNO}_4 + \text{PAN} + \text{PMN} + \text{PPN}$) were generated and archived from a full chemistry simulation before the tagged simulation. Since ozone accounts for most of O_x , we refer to ozone instead of O_x for clarity. Second, GEOS-Chem is run again in the tagged ozone mode using the ozone production and loss data, with separate tracers for the ozone produced from each specific source region. The tagged ozone tracer method can assess the contributions of ozone produced in Africa to other regions. The advantages and limitations of the tagged ozone simulation are discussed in section 2.1.

1. 138 I expected this description earlier.

Thanks. The description has been moved into an earlier part of the section.

l. 140- 143: better include with the GEOSCHEM description.

Thanks. This part has been included with the GEOS-Chem description.

l. 150- what is the reason for not simply taking the ‘african mask’- instead two blocks. I suggest adding a simple figure, showing these masks on top of a map (perhaps along with the HTAP2 definition of Africa).

Thanks for the suggestion. Figure 1 is added to show the definitions of the source and receptor regions in this study. The definition of Africa in HTAP2 is also presented to show the similarity and difference between the two definitions. The reason for using the blocks to define regions is because this is the way that GEOS-Chem uses for the tagged ozone simulation mode.

l. 155 I think most papers that I know keep (anthropogenic) emissions constant- but that is not necessarily the same as keeping the production terms constant. What could be the impact?

In this study, the interannual variation is driven by meteorology only with the fixed daily ozone production rate and loss frequency in 2005. We have generated ozone production rate and loss frequency in other two separate years. Using these two sets of daily ozone production and loss frequency, two additional time series of 20-year simulations can be generated. The three time series show consistent interannual variation in transport of African ozone to the Asian troposphere. This sensitivity test suggests that our results on the interannual variation are robust. The differences in the three sets of ozone production and loss rates reflect partially the differences in emissions in the three years. It is likely that with fixed emissions, the differences in the three sets of ozone production and loss rates will be smaller.

l. 175- what about sfc observations, tropospheric residual from satellite. The comparison is fairly superficial

Thanks. In the revision, we have included more comparisons between the GEOS-Chem simulation and ozonesonde observation by adding 2 figures and two

tables. The seasonal and interannual variation GEOS-Chem simulations have been further compared with ozonesonde data in the surface layer, lower, middle, and upper troposphere at the African sites. Three ozonesonde stations at India are selected for the comparison. Figs. 2 and 3 and Tables 1 and 2 are added in section 2.2. The correlation coefficient (r), bias in percentage, and root-mean-square error (RMSE) between the simulations and the ozonesonde data are presented. The comparison in global distribution with the TES satellite ozone data are shown in the supplement file.

l. 201 Imported ozone=>try to describe more exactly what it is. Region-average abundance of imported ozone (imported ozone could be also the flux through the western border, for instance).

Thanks for the point. We define ozone generated in Africa under the tropopause as African ozone. Following Holloway et al. (2008), "Imported ozone" is used to refer ozone that is distributed over the receptor region. In the paper, when we discuss African ozone over Asia, we use "imported African ozone" to differentiate it from the total ozone concentrations in Asia. We now have clarified this term in Introduction.

p. 295 /figure 6: I guess if the units are molec/cm²/s this pertains to the integrated amount over a model layer; otherwise it should rather be per cm³?

Thanks for the point. Yes, in the last version, the unit pertains to the integrated NO_x amount over a model layer. We have converted the unit into NO_x emissions per cubic meter per second. Therefore, the lightning NO_x emissions are expressed as molec/m³/s in this revision.