

Atmospheric chemistry and physics,

To Reviewer #1,

We appreciate the constructive and helpful comments provided by Reviewer #1, which helped us to improve our manuscript. We have modified to paper to address these comments and a detailed reply is given below with referee comments in italics.

Regards,

Dr. Benjamin Aouizerats

Reviewer #1 (Comments to Author) :

This study presents model simulation results about tropical biomass burning emissions affect air quality of Singapore. This study applied WRF-Chem model to simulate transport of biomass burning emissions in Jul.-Oct. 2006 and the model results are compared with measured PM10 and CO, also satellite measured AOD. The influence of biomass burning to the air quality of Singapore is also evaluated by turning "on" and "off" biomass burning emission. This work is important as it present how emissions from several hundreds km away affect air quality of a highly populous metropolitan. However, the analysis in the manuscript is weak at this point, I recommend major revision before it can be published in ACP.

1. *"This study compare WRF-Chem simulation with measured PM10 in Singapore, measured CO at a station in Sumatra. They also compared WRF-Chem results with satellite measured AOD, but it is kind of failed. The good agreements of PM10 and CO with measurements at two locations are somewhat convincing. But, they have no aerosol composition measurements at all. Good agreements of PM10 can arise from overestimating one species and underestimating the other species, or arise from overestimating primary emissions and underestimating secondary formation. The authors spent a whole section to discuss aerosol compositions in Section 3. If the authors can not provide some evidence to validate their model, it is hard to believe the results. The sentence (P11228 L21-23) "The comparison of model outputs with observations shows that the WRF-chem model set-up is capable of representing quite accurately the evolution of the aerosol concentration for the 4 months of simulation" is just too ambitiously."*

We understand and agree with the point raised by Reviewer #1 stating that the good agreement in PM10 comparison does not necessarily lead to a correct representation of the chemical composition of the aerosol particles. We have added a sentence in that direction in order to put things into perspective concerning the comparison of PM10 : "While the PM10 comparison indicated the model was able to reproduce the measurements, we cannot conclusively state that the model managed to reproduce the aerosol chemical composition because no measurement information on the exact aerosol composition was available. However, given our efforts to accurately take into consideration

the partitioning of emissions (including various Volatile Organic Compounds) as well as the use of one of the most accurate aerosol/chemistry reaction scheme available at the present time (VBS scheme), the good match between the total aerosol mass concentrations modelled and observed yields some confidence in these results."

2. *"The authors also use aerosol compositions data from model to investigate secondary formation in biomass burning plume. Many related important studies are not cited in the paper, including several aircraft BB plume observation data and also laboratory data, such as Vakkari et al., 2014; Yokelson et al. 2009; Akagi et al., 2012; Cubison et al., 2011; Capes et al., 2009; Hennigan et al., 2011. Some of the studies show that OA formation can be very significantly in BB plume. The study of Yokelson et al., 2009 saw very fast (1.4 h) of OA enhancement of a factor 2.3 in tropical BB plume evolution in Yucatan, Mexico and the study environment is highly relevant in this study. This is contrast with the authorsâ model results. Given that SOA is usually underestimated in models and very low SOC/POC ratio in this study, I would recommend the authors work more on this issue."*

We thank Reviewer #1 for pointing out this issue. we have added a short discussion on thus using the suggested literature in the manuscript :

"The results in this study show a significantly lower SOA/POA ratio in the plume than the ratio reported by several studies mainly focused over northern America (Vakkari et al., 2014; Yokelson et al. 2009; Akagi et al., 2012; Cubison et al., 2011; Capes et al., 2009; Hennigan et al., 2011.). This difference may be attributed to several reasons. First, it is well known that due to the complexity involved in the chemical reactions, almost every numerical model tend to underestimate the secondary aerosol formation (Seinfeld and Pandis, 2006). However, it is more likely that the large difference between the SOA/POA ratios reported in the previously mentioned studies and the ones presented in this work are due to the very large concentrations of primary particles emitted by peatland fires. Indeed, the fact that Indonesia has the highest density of fire emissions leads to very large emissions of both primary particles and precursory gases responsible for the formation of secondary organic aerosols. However, the formation of secondary organic aerosols is a strongly non-linear process which depends on numerous and complex processes (such as the VOC concentrations, ozone concentrations, NO_x concentrations, water vapor, aerosol internal mixing rate, etc.) (Seinfeld and Pandis, 2006; Ng et al., 2007). Therefore its formation can quickly reach its saturation mixing ratio or a threshold due to a limiting factor. In our case we believe that the partitioning between the vapor and aerosol phase has quickly reached a saturation point due to the NO_x and ozone conditions, and despite the fact the VOC needed for the formation of SOA are still abundant."

3. *"P11226 L13 : How PM10 and CO are measured. How many sites do you have PM 10 data. Are they urban sites ? Please provide the information."*

We have added information concerning the measurements. As previously mentioned

in the manuscript, the PM10 data are averaged from 5 stations located over Singapore. We have added the urban qualification for more clarity.

4. *"P11247 : Fig. 6 Please provide more explicit x-axis in the figure, e.g. latitude."*

We have added the distance in the transect caption as the latitude and longitude are not linear within the transect.

5. *"What is the different between POA and OCp. Please use a consistent terminology in the paper."*

We thank Reviewer #1 for pointing out this issue. The terminology has been corrected throughout the manuscript to be more consistent.

Atmospheric chemistry and physics,
To Reviewer#2,

The authors appreciate the constructive and helpful comments provided by Reviewer#2 which helped to improve our manuscript. The paper has thus been modified to take into account the recommendations given. Below, we have copied the referee comments in italics and inserted our responses in standard font where appropriate.

Regards,

Dr. Benjamin Aouizerats

Reviewer #2 (Comments to Author) :

"Interesting, useful and timely study. Especially in light of the current efforts to develop and improve biomass burning (BB) emission estimates. Such regional and local studies are necessary to complement the global-model approach, to refine the methodology, challenge assumptions, and enhance our understanding of the complex processes contributing to the picture, which (processes) are often difficult to discern from the coarse global-scale approach to correcting the whole global emission datasets. Distinction between anthropogenic and BB contributions to general smoke pollution is also valuable in this study. The paper is well structured and the study uses appropriate analysis methods. However, backing up the analysis claims more thoroughly with a few more references or explanations would benefit the conclusions. The manuscript is recommended for publication in ACP with some revisions :"

1. *"P11223-L23 Indonesia has the highest concentration of emissions (concentration is expected per some unit : time, person, unit area. . .) - not well communicated"*

We thank Reviewer #2 for this comment and have modified this sentence to "highest density of fire emissions (up to 2000 gCm^2year^1) due to frequent fires and high fuel loads".

2. *"P11226 Section 2.2. Changing the structure of the section will improve readability. Currently the first part of the section leaves me wondering what observations were used (which network/instruments/satellites, where to get the data, references etc. . .) until they are briefly described on P11227-L7. Better familiarity with the dataset earlier in the section, before presenting the result of the comparison could set the stage for better understanding the comparison."*

We agree with Reviewer #2 and have therefore introduced the various datasets in introduction of section 2.2.

3. *"P11227-L17 Why 2-weeks average ? Could you compare instantaneous AOD but more frequently, or 2 weeks was the best signal you get for whatever reason ?"*

It was necessary to perform a moving average over a 2-week period of the satellite observations in order to present a consistent comparison of the various sensor measurements and minimize the error and noise due to the different satellite overpass time and large number of the cloud contaminated pixels. We have added a sentence in the manuscript to clarify this point.

4. *"P11228-L8 CO observational dataset introduction would be helpful, even if only named and described in 1-2 sentences. If I am familiar with the dataset & I can relate, if not and I'd like to know more, I'll pull up the paper that is appropriately referenced."*

We have added a descriptive sentence of the dataset as well as the related link to access data and further information.

5. *"Technical corrections : P11223-L22 ...is neither well understood nor quantified. P11231-L18 . . . as the number of day*s*s* for which . . P11233-L20-21 . . . the impact of biomass burning *on* (?) aerosol pollution levels . . Table 2 please provide the units of mass concentration numbers"*

We have modified the manuscript in order to take into account the corrections. Again, we would like to thank Reviewer #2 for his/her useful comments.

Atmospheric chemistry and physics,

To J. Reid,

We appreciate the constructive and helpful comments provided by J. Reid, which helped us to improve our manuscript. We have addressed some of his concerns. We have spend considerable time to extend the spatial domain of the study but failed to do this within the allocated time due to technical restrictions. However, we have partly addressed the concerns by adding HYPLIT model results as well as adding more perspectives on uncertainties. A detailed response is given below with the referee comments in italics.

Regards,

Dr. Benjamin Aouizerats

J. Reid (Comments to Author) :

"We have a few comments on this paper, which as an outside poster the authors can take or leave. I held off on sending these in as I was waiting for the official reviewers. But as their comments have not come in, we thought we better jot these down. Our group has performed substantial research on the observability and predictability of atmospheric constituents in the region and have some input which we hope the authors find useful. The topic that they address is an important one, with significant scientific as well as political implications and is certainly suitable for ACP. The inclusion of anthropogenic emission simultaneously with burning does make it distinct from other studies and is a useful contribution to the community. However, there appear to be problems with the analysis presented in the manuscript that the authors need to take into account."

1. *"The title gives a bit of a false impression. Really this paper is a case study on the 2006 burning season and its impact on Singapore. The title "Importance of transboundary transport of biomass burning emissions to regional air quality in Southeast Asia" implies an effort much bigger than what is presented. If they are looking at the partition between anthropogenic pollution and biomass burning in Sumatra for the biggest biomass burning event of the EOS era, they should simply say that"*

While we understand the concerns about the title not pointing that this work is based on a case study, it does reflect the main purpose of this work : a better description of the interactions between biomass burning and anthropogenic emissions at regional scale. We have changed the title to : "Importance of transboundary transport of biomass burning emissions to regional air quality in Southeast Asia during a high fire event" to indicate our work is based on a case study.

2. *"Their domain is Sumatra and the Malay Peninsula. Borneo is absent, as is Java. We dont agree at their supposition that these islands can be ignored. From Wang et*

al., 2013 (cited in the paper), transport across the Java Sea from Borneo is clearly occurring-just look at the satellite images."

While satellite images may show transport occurring from Borneo to Singapore, such analyses provide mostly information about the column concentrations and do not necessarily yield information on what is going on at the surface in which we are interested mostly. Although we were not able to rerun the model with an extended domain including Borneo, we provided supplementary material with the HYSPLIT dispersion model results including deposition process which clearly shows that the 3-D dispersion of mass emitted in Borneo does not reach Singapore at the surface level during our period of interest. Clearly the concern of Reid is valid and we have added a section in the manuscript discussing the origin of the air mass reaching Singapore during the second half of October.

3. *"Their statement that easterly winds for the Oct 2006 were light and variable is at odds with the Singapore RAOB site ([http://weather.uwyo.edu/...](http://weather.uwyo.edu/)) which shows consistent easterly PBL winds of 5-10 knots. Surface winds alone are not an adequate representation of regional transport. Further, based on the analysis of Atwood et al., (2013) there is likely a reservoir of smoke aloft being entrained into the PBL-something that models often represent poorly. Similarly, there have been many who have hypothesized (including the co-authors) that Jakarta is an important source for Singapore. Thus, I think there needs to be discussion on this point. A general Printer-friendly Version analysis describing the meteorology of Borneo and Java transport can be found in Reid et al., 2012, Atwood et al., 2013, and Xian et al., 2013."*

Concerning the wind fields, we had compared the WRF-Chem results with observations from the National university of Singapore (<https://inetapps.nus.edu.sg/fas/geog/ajxdirList.aspx>) which show for the month of October 5-meter winds oscillating from 0 to 5 m.s-1 with associated directions from NE to SSW. This is in good agreement with our model results mentioned in the manuscript.

We agree that there is the possibility of the presence of an elevated residual layer of aerosols which may act as a reservoir for the PBL. However, we believe that WRF-Chem under the configuration we used is one of the most accurate models available to reproduce the reality as closely as possible.

Concerning the contribution of Jakarta to the PM levels in Singapore, we understand and agree that under certain circumstances, it can be an important source. In this study, we focused on the transport from Sumatra to Singapore, and while Java is not in the domain (just as Borneo), their respective contribution to the total PM concentration is still accounted for by including PM and gases injections from the boundaries of the domain based on MOZART model reanalyses. It is considerably lower than the contribution from fires in Sumatra during that time period.

4. *"This then leads to the verification data being a bit at odds with a simple review of satellite imagery. Their simulations suggest no fire influence just when we expect Borneo influence to be most important (Fig. 7)."*

Indeed, while most studies focusing on this region using on satellite data tend to attribute the total contribution of PM level in Singapore to fires, the goal of this study is also to point out the complexity of modelling aerosol particle evolution (transport as well as physical and chemical processes) and we think that high-resolution models are needed to accurately represent the processes involved and solve them online. When we interpreted our results, we were equally surprised that the coinciding poor air quality in Singapore and heavy burning in Kalimantan during the second half of October may have actually just been coincidence. But based on the good correlation of PM10 values between our model and measurements as well as the HYSPLIT results in the supplementary material we can be relatively confident in our conclusions.

5. *"Regarding verification data with satellite AOT data, we would like to point out the analysis in Reid et al., 2013, which clearly demonstrates that satellite AOT products underestimate true AOT values. Thus, the difference is even bigger than reported. This is due to two things. First, high AOTs are often flagged as cloud. Second, for moderate values of AOT, the assumption of single scattering albedo is far too high. While I appreciate there is little verification data out there for the 2006 event, I think the fact that the AOTs for some of the most significant events could be underestimated by more than 50% or more should be noted. Even though as the authors note that AOT is not a criterion pollutant, the fact that they have good results at the surface yet cant constrain total mass loadings has implications for the source function and the transport"*

We have added some discussion in the revised manuscript noting the uncertainties in the AOT values and that the AOT values may be underestimated, "Moreover, as shown by Reid et al. (2013), AOD measurements in this region are often underestimated by up to 50%". We agree that comparing model results only at the surface is not sufficient to fully validate the source function as well as the transport (and the aerosol physics-chemistry) of PM. We know our study is not the last word on this but we feel we have made progress by comparing to both surface CO and PM10 and column AOD values. Future research is needed to reconcile why the surface observations were matched reasonably well but the AOD not. To date, most studies focusing on only AOD indicated GFED-type bottom-up emissions were too low. We show this may be cutting corners ; if we had done this (that way matching AOD) we would have overestimated the surface measurements. Clearly, reconciling this mismatch is an important research area.

6. *"For their source function, the authors should review Hyer et al., 2013 (as well as the commentary in Reid et al., 2009 and 2013). The fact of the matter is we donât know source functions to better than integer factors. Hyer showed that the GFED method favors larger fires. With all methods, we know there are countless small fires undetected by either burn scar or thermal anomaly. This should at least be mentioned."*

The manuscript has been updated to show some perspectives on the fire detection, and the method used by GFED3 to correct the omission of small fires. Moreover, we have compared the results from GFED3 (which includes a "boost" based on fire persistence to account for omitting fires in deforestation areas) and GFED4 where

small fires are more accurately accounted for and for our region and time of study : http://www.globalfiredata.org/_plots/timeseries_0.pdf.

It appears that the differences are relatively small.

7. *"Bottom line for us is that it is good to get the industrial pollution into the picture, and the authors can do everyone a great service by generating a solid simulation which they and regional researchers can mine. Qualitative verification of the model outputs against the observed meteorology is also a necessary step. Some discussion of satellite and model uncertainties needs to be incorporated in the study. I would strongly suggest rerunning with a larger domain. You will be glad you did (as will the community). That larger domain could then be used to apply this analysis for all of the major cities in the region, not just Singapore."*

We had hoped to update our results with a larger domain. Unfortunately we did not succeed in this within the allotted time but were strengthened in our conclusions thanks to the HYSPLIT results. We have validated our results with multiple sources of information (trace gases, PM10 measurement, AOD, and a quantitative comparison against meteorology) and have added more discussion on uncertainties. Clearly, more can and will be done in the future and our results are, just like most other studies, just a step in the -hopefully- right direction.