

Responses to the Comments of the Anonymous Referee #2

We very much appreciate the constructive comments and suggestions from this reviewer. The following are our point-by-point responses to the reviewer's comments (the reviewer's comments are marked in *Italic font*).

General comments:

The paper provides a correlation of modeled particulate matter with low visibility days recorded at observation sites across South East Asia. Information is presented about the most likely source areas for biomass burning pollution for different cities and different seasons.

This is an interesting application of an alternative observation dataset for assessing the impact of biomass burning haze on the region and for validating CTM and dispersion models. However, the significant flaw in the way the results are presented is that the model is assumed to be correct and that all low visibility days that are not modeled are therefore specified to be due to other pollution contributions. The validity of this assumption is not demonstrated. It is quite possible that the model is over-estimating the biomass contribution at some sites and underestimating it at others. Fig 6 for example would suggest that the model may not be capturing up to 50% of the fire haze days, and Fig 4 would suggest that the model misses 50% of the VLVDs at Singapore. The references in the text to fire and non-fire LVD are therefore misleading. The authors need to reconsider how they interpret this data and present it in the paper.

We are fully aware of the uncertainty of our model due to factors including emissions, model resolution, and meteorological fields. The uncertainty of modeling was repeatedly indicated in the manuscript, and the additional simulations using different emission inventories and meteorological fields were all designed and conducted for the purpose of identifying, at least partially, the influences of these uncertainty factors on modeled results. Nevertheless, the reviewer's point is well taken. We have made our best effort to reiterate the model uncertainty and evaluation in the revised manuscript. In addition, we have specifically indicated in many places that the model's overestimates in visibility range (underestimates in visibility degradation) are likely due to the fact that observed visibility reflects contributions of both fire and non-fire aerosols.

We have revised the description in Section 2.3 regarding our method to attribute low visibility events to fire aerosols (such events can be induced by either fire or non-fire aerosol alone or in combination), as: "As mentioned above, a visibility of 10 km is considered an indicator for a moderate to heavy particulate pollution. Hence a visibility of 10km in observation is used as the threshold for defining the "low visibility day (VLD)" in our study. We firstly derived the observed low visibility days in every year for a given city using the GSOD visibility data. Then, we derived the modeled low visibility days following the same procedure but using modeled visibility data that were only influenced by fire aerosols. Both the observed and modeled visibilities were then used to define the fraction of low visibility days that can be caused by fire aerosols alone. It is assumed that whenever fire aerosol *alone* could cause a low visibility day to occur,

such a day would be attributed to fire aerosol caused LVD, regardless of whether other coexisting pollutants would have a sufficient intensity to cause low visibility or not. In addition to the LVD, we have also used a daily visibility of 7 km as the criterion to define the observed “very low visibility day (VLVD)”. Such heavy haze events in the region are generally caused by severe fire aerosol pollution, thus we use their occurrence specifically to evaluate the model performance”. In addition, we have revised statements of fire aerosol contribution to contain “up to” whenever necessary.

Furthermore, the descriptions of model evaluation based on model-observation comparison have been revised, two new or largely revised paragraphs in the revised manuscript are added in Section 3.1, they provide the procedure and present the uncertainty of the model in a greater detail and clarity:

“The surface observational data of $PM_{2.5}$ concentration among these four cities are only available in Singapore since 2013 from the National Environment Agency (NEA) of Singapore. We thus firstly used these data along with visibility data to evaluate model’s performance for fire-cause haze events reported in Singapore during 2013-2014 (Fig. 3). Note that the observed $PM_{2.5}$ level reflects the influences of both fire and non-fire aerosols, whereas the modeled $PM_{2.5}$ only includes the impact of fire aerosols. We find that the model still predicted clearly high $PM_{2.5}$ concentrations during most of the observed haze events, especially in June 2013, and in spring and fall seasons of 2014 (highlighted green areas), though with underestimates in particle concentration of up to 30-50%, likely due to the model’s exclusion of non-fire aerosols, coarse model resolution, overestimated rainfall, and errors in the emission inventory. Figure 4 shows observed visibility versus modeled visibility in FNL_FINN during the fire events shown in Fig. 3. Note that all these events have an observed visibility lower than or equal to 10 km, and are identified as LVDs. In capturing these fire-caused haze events, the model only missed about 22% of them, or reporting a visibility larger than 10 km in 40 out of 185 observed LVDs as marked with purple color in Fig. 4. When observed visibility is between 7 and 10 km, model results appear to align with observations rather well. For cases with visibility lower than 7 km, the model captured all the events (by reporting a visibility lower than 10 km, or LVD) although often overestimated the visibility range. These results imply that the VLVDs only count a very small fraction in VLDs and thus are episodic events. It is very likely that the size of concentrated fire plumes in VLVDs might be constantly smaller than the 36 km model resolution; therefore, the model results could not reach the peak values of $PM_{2.5}$ concentrations of these plumes.

Furthermore, the LVDs in the four selected near-fire-site cities during the fire seasons from 2003 to 2014 have been identified using the daily GSOD visibility database and then compared with modeled results (Fig. 5). It is difficult to identify all the fire caused haze events beyond Singapore even in recent years. However, in Southeast Asia, severe haze events equivalent to the VLVDs in visibility degradation are known to be largely caused by fire aerosol pollution. Therefore, we used the observed VLVDs in the four selected cities to evaluate the performance of the model. We find that the modeled result displays a good performance in capturing observed VLVDs despite an overestimate in visibility range during certain events compared with the observation. The model in

general only missed about 10% or fewer VLVDs observed in the past decade (Table 3; Fig. 5). In addition, the model has reasonably captured the observed LVDs despite certain biases (Fig. 5), likely due to the fact that fire aerosol might not be the only reason responsible for the degradation of visibility during many LVDs”.

The paper would benefit from some reorganization of the sections and a reduction in the number of figures.

Based on the reviewer’s suggestion, we have reorganized the manuscript. Specifically, Section 2 and 3. Section 4 has been rewritten.

Specific Comments:

Following on from the general comments, I am concerned that no real attempt at model validation is made within this paper. An additional source of observed data, e.g. PM10 concentrations, from a minimum of one of the sites (ideally many more) is needed to demonstrate that the WRF-Chem simulations are correctly capturing the fire component. The data shown in Fig 5(a) is misleading due to the use of different scales and a more robust analysis of this data is needed earlier in the paper. In fact this data may reveal useful information about missing “background” PM from the model. There are statements on line 320 that the model is underestimating PM2.5 concentration by up to 30-50% in this comparison. This is a significant underestimation. What impact does this then have on the visibility and hence the LVD calculations? The authors also need to discuss in more detail the impacts of uncertainty on the LVD and VLVD estimates. Without this level of validation, the model results cannot be used to the level of precision that the authors present in e.g. Table 2.

We appreciate the reviewer’s comments. However, as perhaps the reviewer is well aware, observational data of aerosols in Southeast Asia are still quite limited. This is also a reason why we used surface visibility data (a proxy data of PM_{2.5}) in the study. Besides PM_{2.5} data in Singapore, there are some PM₁₀ monitoring data in Thailand and Malaysia. However, these are not the best data for visibility calculation due to a lack of knowledge of size distribution, not mentioning the sparseness of these data.

As reported in the paper, our model evaluation contains two parts: one is on modeled meteorological features and the other is on fire PM_{2.5}. Accepting the reviewer’s suggestion, the detail discussion of meteorology evaluation including precipitation and wind field is now presented in Section 2.2 of the revised version.

Regarding the underestimate of PM_{2.5} concentration by up to 30-50% compared to observation as shown in Fig. 5 (a) (new Fig. 3(a)), our response to the reviewer’s general comments along with the newly added paragraphs in 3.1 should also address this specific comment. After all, observed PM_{2.5} concentrations still reflect the contributions from other besides fire aerosols. We have added statements to indicate this fact in the revised manuscript.

We have also adjusted the scales of Fig. 5 (now Fig. 3).

I would also like to see some explanation as to why the modeled visibility distance for Bangkok in Fig 4 is significantly lower than that in the observations (and in comparison to the difference at other sites), and consequently what this means for the calculation of VLVDs.

Thanks for asking this interesting question. The reason why the modeled visibility in Bangkok is lower than observation in certain time period can be explained by Fig. 2 in the revised version and Fig. S5a in the supplementary section. We find that fire PM_{2.5} emissions in FINNv1.5 are about a factor of 2 or 3 higher than those in GFEDv4.1s in mainland Southeast Asia (s1) during fire seasons. Note that such a difference between the two emission inventories does not show in other fire sites, i.e., s2 – s4. This implies that FINNv1.5 likely overestimated the fire emissions in mainland Southeast Asia and thus this leads to a modeled visibility in our FNL_FINN lower than observation in Bangkok. We have added the discussion in Section 4 of the revised manuscript as: “Compared to FINNv1.5, fire emissions in GFEDv4.1s over mainland Southeast Asia are more than 66% lower (Fig. 2a), and this results in a 43% lower fire PM_{2.5} concentration in Bangkok (Table 4). The lower fire PM_{2.5} concentration in FNL_GFED actually produces a visibility that matches better with observations in Bangkok comparing to the result of FNL_FINN (Fig. S5a). This implies that the fire emissions in FINNv1.5 are perhaps overestimated in mainland Southeast Asia”.

The decision that the “other pollution contribution %” is “100% minus Fire pollution contribution %” is not appropriate for the analysis that is then presented. Statements such as those on line 336-338 and line 345-347 do not hold up. The authors need to present a justification for why the reader should assume that the model data is correct. Even so, all interpretation of non-fire LVD should probably be removed.

Our analysis only implies that “by considering fire aerosol alone” how many LVDs can be attributed to fire particulate pollution. We actually emphasized this point in many places of the original manuscript. The reviewer’s point is well taken. To further avoid the misunderstanding, we have made it even more clearly in the revised manuscript by: (1) laying out more details about our judgment making, (2) clarifying that other cases are those that cannot be explained by fire aerosol alone, and (3) adding “up to” in the statements when necessary when referring to fire aerosol contribution. In addition, we have made our best effort to indicate that all these implications do not need to assume a perfect model to achieve.

To aid the discussion of the changing number of LVDs further explanation of certain statements is needed. For example, Line 366-368, why is Kuching different to Singapore? Could this be because Kuching is within a fire area?

We appreciate the reviewer’s suggestion. We have stated “Kuching is in the coastal area of Borneo so Kuching is directly affected by Borneo fire events (s3)“, and also “Because of its geographic location, Kuching is affected heavily by local fire events during the fire season (Fig. 7d). Fire aerosols can often degrade the visibility to below 7 km and can even reach 2 km (Fig. 3d)” in the revised version.

More information and explanation on the model set-up and analysis approach are needed to help the reader understand what has been done. Including (a) in section 2, further explanation about the “chemistry tracer module” is required – is there any chemistry at all? It doesn’t appear so, so this is a bit misleading. It would be better to say “chemical tracer module” and be clear that the pollutants are being modeled as tracers only. The lines on p8 (163-164) describing the deposition processes could usefully be moved to this earlier point in the text. An explanation for why the domain extends so far west would also be helpful. (b) p9 line 180 – the authors need to clarify whether emissions have been injected at just 700 m or from the surface to 700 m. Is this asl or agl? (c) More detail (ideally the equations used) is needed as to how the hygroscopic growth is calculated on p11 line 232 and how this relates to the visibility calculation. Also where has the environmental relative humidity data that is used come from? This is fundamental part of the model data processing, and will introduced it’s own uncertainties, but is rushed over (d) There is currently no information on how the model output has been produced for each site, so this needs to be added. For example, is it based on the modeled concentration in the lowest WRF-Chem layer for the grid box corresponding to each observation site? (e) A brief explanation as to how the runs have been conducted to identify the different source sectors is needed. Did these use labeled tracers?

(a) The sentence has been changed to “to thus model the fire PM_{2.5} particles as tracers without involving much more complicated gaseous and aqueous chemical processing calculations but dry and wet depositions.” We have also moved the description of deposition calculation to this place in Lines 120-122 of the revised version.

(b) We have changed the sentence to: “Therefore, we have limited the plume injection height of peat fire by a ceiling of 700 m above the ground in this study based on Tosca et al. (2011). The vertical distribution of emitted aerosols is calculated using the plume model.” in Lines 160-162 of the revised version.

(c) We have added the calculation of hygroscopic growth factor and the radius increase adjustment after hygroscopic growth in Eq. (2) and (3) in the revised version. The data of relative humidity for the hygroscopic growth calculation are from the model results.

“We also consider hygroscopic growth of sulfate fraction of these mixed particles in the calculation based on the modeled relative humidity (*RH*). Based on Kiehl et al. (2000), the hygroscopic growth factor (*rhf*) is given by

$$rhf = 1.0 + \exp \left(a_1 + \frac{a_2}{RH+a_3} + \frac{a_4}{RH+a_5} \right), \quad (2)$$

where *a*₁ to *a*₅ are fitting coefficients given by 0.5532, -0.1034, -1.05, -1.957, 0.3406, respectively. The radius increase of wet particle (*r_{wet}*) due to hygroscopic growth will be

$$r_{wet} = r_{dry}^{rhf}, \quad (3)$$

where *r_{dry}* is the radius of dry particle in micron.” has been added in Section 2.4 in the revised version.

(d) The fire PM_{2.5} concentration presented in the paper is averaged within the PBL for the grid box corresponding to each observation site. This information has been added in the caption of Fig. 7 and 9.

(e) Yes, we labeled tracers from each source region when we created fire emission in WRF-Chem inputs. This is actually described in the emissions section, Section 2.1.

The use of two different time periods for the analysis of the results for the FINN data vs. the GFED data introduces differences in the outputs, which could be misinterpreted. It makes Table 3 particularly complicated to interpret. I would recommend that throughout the paper the authors only present data for the same period for all 3 model simulations (i.e. 2003-2014) to avoid introducing additional uncertainty and confusion in their results and analysis.

The reviewer's suggestion is well taken. All discussion and data in the revised manuscript are now presented from 2003 to 2014.

I would also recommend that Table 3 is modified to present the total number of days in the 12 year period rather than an annual average, as the latter significantly distorts the true year to year variability and introduces false precision.

We believe the reviewer's comment applies to Table 2 not Table 3 in the original version. Actually, the percentage values used in current Tables (i.e., mean LVDs/365 x 100%) serve the same purpose to describe the haze situation in any given year as suggested by the reviewer. The standard deviation shows year to year variation.

The language needs some improvement particularly in the abstract and the introduction. The use of "particulate matters" rather than "matter" is somewhat unconventional.

We thank the reviewer's comment and we have tried our best to polish the language of the manuscript.

The discussion of the role of precipitation jumps around the sections, so the authors are encouraged to see if this could be pulled together into one, shorter overview section. Some of the text regarding the precipitation in section 2.4 needs further explanation. For example on line 275 more detail and/or a citation is needed for the FDDA grid nudging. The use of mean monthly rainfall to compare the models and observations (lines 269-274) seems strange given that the authors have nicely demonstrated the large annual variation in rainfall timing and magnitude across the region. It would be useful to explore whether the models are better in some seasons than others in this region? On Line 281 the authors mention the temporal correlation, but also need to state over what averaging period this is, e.g. is this based on daily, weekly, monthly mean or total ppt data? Figure 3 is particularly hard to interpret. Difference plots would be more useful here, but this figure is a candidate for removal.

The reviewer's point is well taken. We have added the discussion about the evaluation of simulated rainfall and wind field and moved them all to Section 2.2. We have also added Table 2 in the revised version to present the spatial and temporal correlation of monthly rainfall between model and observation in different season.

The original Fig. 3 has been moved to the supplementary.

Section 4 would benefit from a broader discussion of the NWP datasets, for example there is currently no discussion of the wind fields, which are of higher order relevance than the precipitation, particularly for the source area identification. I also find it slightly surprising that given that the LBCs are a long way from Sumatra that WRF develops such a discrepancy in precipitation over the central region of the domain in the different runs. Is there a similar difference in the winds, which would therefore impact the transport? Has any verification of the WRF wind data been conducted? This section would benefit from being merged with the other sections on meteorology.

We have added a discussion of the surface wind difference in Section 2.2 along with related figures (Fig. S2 and S3) in the supplementary. Figure S2 and S3 show the surface wind of reanalysis data of FNL and ERA in the summer and winter monsoon seasons and the difference between FNL_FINN and ERA_FINN modeled winds. In responding to the reviewer's suggestion, we have also added discussions of the mesoscale wind pattern change in Section 2.2 besides rainfall evaluation. The discussion about the impacts of different meteorology inputs on modeled PM_{2.5} concentration and LVDs are presented in Section 4 of the revised manuscript.

The attempt by the authors to use the data to assess the impact of the haze on populations in SE Asia is to be commended, but the approach taken is needlessly complicated. The units of the HED metrics are unclear and the dominance of population size on the HED_{pw} metric needs more careful explanation. What the results are showing are that the total number of LVDs in the region (based on observations at 50 cities) has increased over the analysis period. This conclusion could be reached without the HED and is easier to explain and understand for the reader. As explained previously the statements in this section about non-fire pollution are not justified by the approach.

Haze Exposure Day (HED) can be defined by the population weighted or arithmetic mean over the included cities. The latter perhaps is the format suggested by the reviewer. As shown in the paper, we have provided results of both. The population weighted exposure is commonly used in health and policy analyses because it clearly indicates the impact correlated to population distribution. The meanings of both types of HED have been described along with their definition. The reviewer's point is well taken and we have made our best effort to clarify the implication of our results relating to fire aerosols.

The manuscript would benefit from fewer figures and I am not sure the supplementary material adds anything. The line thickness in many of the line graphs means that the bottom lines are often hidden, this is always a problem with this sort of graph, but a reduction in the line thickness would be beneficial.

We thank the reviewer's suggestion. We have moved the Fig. 3, 10 and 13 in the original version to the supplementary and have removed Fig. 2 and 11. All y-axes in the figures have been set to start from zero in the revised version.

Technical Corrections

P2 line 45 – 99.1% is over stating the precision here. I would suggest using only 99%

which is in line with the precision of other numbers given in the abstract.

Modified.

P4 line 66-73 – The discussion of radiative impact isn't relevant to the rest of this work, so seems unnecessary. Recommend deleting these lines.

We have shortened the discussion of radiative impact of fire aerosols in the Introduction.

Line 325-327 – it would be more helpful to the reader if these percentages were expressed as a number of days. The language at the end of this sentence could also be improved.

The sentence has been modified to “We find that the annual mean LVDs in Bangkok has increased from 47% (172 days per year) in the first 5-year period of the simulation (2003-2007) to 74% (272 days per year) in the last 5-year period (2010-2014). The LVDs caused by fire aerosols has increased as well (Fig. 6a).” in Lines 352-355 of the revised version.

Line 237 – Is the total population figure here correct? It is not clear if this the combined total, or if each city has more than 2 million?

There is no population figure presented in the paper. We are not sure to which figure the reviewer was referred. The population information of 50 ASEAN cities has been added in the supplementary (Table S1) in the revised version.

Table 2 – The table would benefit from explanation that the VLD and VLVD for FNL_FINN and ERA_FINN are identical as they are based on observations, and that the data for FNL_GFED is different as it covers a shorter time period. However see comments regarding making the time period consistent.

The caption of Table 3 in the revised version has been changed to “Annual mean low visibility days (LVDs; observed visibility ≤ 10 km) and very low visibility days (VLVDs; observed visibility ≤ 7 km) per year in Bangkok, Kuala Lumpur, Singapore and Kuching during 2003-2014 are presented in the second column. Parentheses show the percentage of year. The third and fourth columns show the percentage contributions along with standard deviations of fire and non-fire (other) pollutions for total low visibility days.”

Table 2 - The FNL_FINN LVD line for Singapore does not add up to 100%.

In the revised version, the data have been changed to 36% and 64% based on the analysis from 2003 to 2014.

In Table 3, the caption states that “parentheses show the fire aerosol fraction in total PM2.5” – this is very unclear and confusing. It could be taken to imply that the model also contains non-fire PM2.5, but I don't think this is the case. I think the table would be more informative and cleaner if all of the parentheses data were removed.

We would like to keep the information of the percentage of fire aerosol contribution from each source region in the table. We have modified the caption to “Parentheses show the percentage of fire PM_{2.5} contribution originating from each source region.” to clarify the meaning in the parentheses.

Figure 2 – it would be useful to highlight in the caption that all of the plots have different axes scales.

Highlighted as suggested. Figure 2 has been removed to reduce the number of figures in the manuscript.

Figure 5 – the use of different axis scales in (a) is very misleading. Both data sets should be presented with the same scale and starting from 0. Where is the data that gives the green areas from? This data could usefully contribute to the discussion in the text and the validation of the model.

We now use the same scales starting from zero. The haze events highlighted in green are manually selected based on observed PM_{2.5} concentration and visibility. A detailed discussion has been added in Section 3.1.

Figure 6 – A better way to present this data would be to have the green data as the GSOD observed LVDs and the red data as the modeled fire LVDs. This would be a more robust comparison of model vs. observations and start to address issues in the comments above.

We very much appreciate the reviewer’s suggestion. However, since the observations actually contain both fire and non-fire contributions, therefore, we believe the current column charts present the results rather well. In this figure, each column presents the observed LVDs in each year or month. For example, in Fig. 6a of the revised version, column 2003 shows 40% observed LVDs (green + red), which includes 10% fire LVDs (red) and 30% other LVDs (green).

Figure 7 – the S1 and S5 line colors are too similar in my copy, so can one of these be changed please.

Changed the s5 line color to orange.

Figure 9 – Need to specify that these are “fire” concentrations in the caption. In this and Fig 10, the purple contours on the right hand plots prevent the underlying colors from being seen and are so small that they are unreadable, so recommend that these are removed.

We have modified the caption to contain “fire PM_{2.5} concentration”. We have also removed the contour lines in Fig. 9 (f) – (g) and Fig. S4 (f) – (g).

Figure 11 – To ensure that there is no unintentional bias, the plot would be better if it depicted data for only 2003-2014 for all of the data sources.

We have removed this figure in the revised version.