Responses to the Comments of the Anonymous Referee #1

We very much appreciate the constructive comments and suggestions from this reviewer. Our point-by-point responses to the reviewer's comments are as follows (the reviewer's comments are marked in Italic font).

General comments:

The manuscript addresses an emerging issue for Southeast Asia which concerns the impact of biomass burning on air quality and visibility. The topic is highly relevant for publication in Atmospheric Chemistry and Physics, however major issues related to the form in which the work is structured and presented (i.e. a whole rewriting of the paper is needed), clarifications in methods and analyses need to be addressed. The overall work needs to be synthesized both in the text and in the selection of the figures presented (of the 13 figures included some of them duplicate information included in other ones. If the authors want to keep all of them, they should consider moving some of the figures to the Supplementary Materials).

Based on the reviewer's suggestion, the structure of the manuscript has been rearranged, especially in Section 2 and 3. In addition, Section 4 has been rewritten. Please note that, based on the other reviewer's suggestion, all analyses of model results and observations are now applied to the time period from 2003 to 2014.

Specific comments:

Language

A major rewriting of the paper is needed. Several sentences are not fluent and a grammar/ punctuation check is needed. Below are some examples:

Line 32: remove "that"

Done.

Line 33: favorite should be "favourable"

Modified to favorable.

Line 41 and other parts: please be consistent with the tense you use. ....

We have checked the tense throughout the manuscript.

Line 55: "put in effect", replace with "implemented"

Done.

Line 82: please check your references (e.g. Miriam is the first name)

Corrected.

Line 118: "the great Southeast Asia" should be replaced with something similar to "over the whole Southeast Asia". Please check also elsewhere in the paper.

Modified to "the whole Southeast Asia" throughout the manuscript.

Line 135: please rephrase

The sentence is revised to "Our focus in this study is on the fire aerosol life cycle. Therefore, we chose to use WRF-Chem with a modified chemical tracer module instead of a full chemistry package, 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." in Lines 118-122 of the revised version.

Line 168: "estimations" should be always replaced with "estimates"

Modified throughout the manuscript.

Line 172: remove "with"

Done.

Line 178: "comparing" should be "compared". Please amend this everywhere in the paper.

Modified throughout the manuscript.

Line 190-202: please rephrase and summarize. This paragraph is too repetitive and needs to be more concise.

The paragraph has been rephrased to "Generally speaking, there is a strong correlation between the seasonal variation of fire emissions and that of rainfall in all fire regions as shown in Fig. 2. Because mainland Southeast Asia (s1) and northern Australia (s5) are on the edge of the seasonal migration of the ITCZ, the correlation in these two regions is even more pronounced. On the other hand, in Sumatra (s2), Borneo (s3) and the rest of Maritime Continent (s4), while inter-seasonal variations of rainfall and fire emissions are still correlated with each other in general, fire emissions do exist in some raining seasons (Fig. 2b – d), owing to the precipitation features in multiple scales over these regions (e.g., the passage of MJO events) and underground peatland burning." in Lines 172-180 in the revised version.

Line 211: units, please replace also elsewhere Line 236: "this" is missing

Done.

Line 294: "so that" is very often used incorrectly. Please check all the occurrences. Line 343: "are occurred", should be "occurred" Line 515: "reasons" should be "seasons".

Please check also other typos.

Removed "so that" in the sentence and rephrased. Done correcting typos.

Line 518-519: Please rephrase

The sentence is removed. Section 4 has been rewritten in the revised version.

Line 571-580: this section needs to be rewritten. Sentences are too long and convoluted and several grammar errors are present.

We have rewritten Section 4.

Methods:

All the introduction regarding WRF is not needed since you are using a modified version of WRF-Chem. Also you start introducing the model and have section 2.2 describing the emissions and section 2.4 discussing again the simulations. The whole method section has to be reorganized (e.g. have one section discussing the data, one on the model and one on the methods used). Please be more concise and avoid repeating the same information in different sections.

The introduction of WRF-Chem in Section 2.1 has been condensed. We have also rearranged the structure of Section 2. Besides section 2.1, the descriptions of numerical simulations and model evaluation has been moved to Section 2.2, observation data and model derivation of visibility to Section 2.3, and the "Haze Exposure Day (HED)" definition to Section 2.4.

Line 123: please refer more precisely to your "targeted science questions"

The sentence has been revised to "In this study, we have used the Weather Research and Forecasting (WRF) model coupled with a chemistry component (WRF-Chem) version 3.6 (Grell et al., 2005). Our focus in this study is on the fire aerosol life cycle. Therefore, we chose to use WRF-Chem with a modified chemical tracer module instead of a full chemistry package, 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" in Lines 117-122 of the revised version.

Line 139: you mostly focus on visibility so please also add that.

The sentence has been revised to "This configuration lowers the computational burden substantially, and thus allows us to conduct long model integrations to determine the contributions of fire aerosol to the degradation of visibility in the region over the past decade." in Lines 123-126 of the revised version.

Line 145: this is redundant information, please remove it.

Removed.

Line 146: The reported time step is for chemistry or physics?

We have made this clearly by stating: "The time step is 180 seconds for advection and physics calculation." in Line132 of the revised version.

Line 165: Did you only include fire emissions? Does WRF-Chem use other anthropogenic emissions?

We only included fire $PM_{2.5}$ particles in the model; therefore, emissions of other chemical species were excluded in the simulations. To make this clearer to the reader, we have added in the manuscript that: "Therefore, we chose to use WRF-Chem with a modified chemical tracer module instead of a full chemistry package, 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." in Lines 119-122 of the revised version.

Line 208: this should be rephrased by saying what you used for computing visibility.

The sentence has been rephrased to "In this study, the visibility is calculated by using the *Koschmeider equation: …*" in Line 238 of the revised version.

Line 213-216: please add a reference and rephrase

The sentence has been modified to "Based on Eq. (1), a maximum visibility under an absolutely dry and pollution-free air is about 296 km owing to Rayleigh scattering, while a visibility in the order of 10 km is considered under a moderate to heavy air pollution by particulate matter (Visscher, 2013)." in Lines 242-245 of the revised version.

Reference:
Visscher, A. D.: Air Dispersion Modeling: Foundations and Applications, First ed., John Wiley & Sons, Inc., pp. 50, 2013.

Line 222: please be more specific by explaining how you will use the GSOD data and to address which objectives

We have added the explanation and also rephrased the sentence to "The observational data of visibility from the Global Surface Summary of the Day (GSOD) (Smith et al., 2011) are used in our study to identify days under particulate pollution, i.e., haze events." in Lines 250-252 of the revised version.

Line 219: add "by increasing bext"

The sentence has been revised to "Similarly, fire aerosols, alone or mixed with other particulate pollutants, can degrade visibility by increasing b_{ext} and lead to occurrence of haze events too." in Lines 247-249 of the revised version.

Line 225: Here you introduce model simulations, but you have a section later discussing

that. You should reorganize the methods and be more clear on the objectives you are addressing. "In order to compare with observations", what do you mean? Are you referring to a model evaluation? If so please explain in the relevant section how you will perform it.

This paragraph describes the procedure of using observed visibility to evaluate modeled $PM_{2.5}$ concentrations in our study, and also the method of deriving modeled visibility based on the extinction coefficient of simulated fire aerosols as a function of particle size. We have modified the sentence to: "The observed visibility is also used to evaluate the modeled visibility and thus $PM_{2.5}$ concentration. The modeled visibility is derived based on the extinction coefficient of the fire aerosols as a function of particle size, by assuming a log-normal size distribution of accumulation mode with a standard deviation $\sigma = 2$ (Kim et al., 2008). Note that all these calculations are done for the wavelength of 550 nm unless otherwise indicated." in Lines 255-259. We have also added the details of particle hydroscopic growth calculation in Lines 264-270 of the revised version.

Line 227: is there a reference you can quote for these assumptions? Or some local measurements used to estimate those parameters?

We have cited Kim et al. (2008) and added this reference in the revised manuscript.

Reference:
Kim, D., Wang, C., Ekman, A. M. L., Barth, M. C., and Rasch, P. J.: Distribution and direct radiative forcing of carbonaceous and sulfate aerosols in an interactive size-resolving aerosol–climate model, Journal of Geophysical Research: Atmospheres, 113, D16309, 10.1029/2007jd009756, 2008.

Line 225-233: this paragraph should be clarified. It is not clear how you link the discussion on fire emission composition, hygroscopic growth, etc. with your work. If it is for general overview purposes, please add it to the introduction or remove it.

We have added more details of the visibility calculation, specifically the method to include the effect of particle hydroscopic growth in Section 2.4 of the revised version:
"To make the calculated visibility of the fire aerosols better match the reality, we have also considered hydroscopic 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 hydroscopic growth factor (*rhf*) is given by

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

where a_1 to a5 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 hydroscopic growth will be

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

where r_{dry} is the radius of dry particle in micron."

Line 238-239: again this is repetition of definitions already given. Please remove this from here and elsewhere in the manuscript.

Removed.

Line 268: what is the NCAR_FNL? You have not introduced that before. Please add a reference for all datasets used.

We thank the reviewer for pointing out this typo. We have corrected "NCAR_FNL" to "NCEP_FNL".

Line 267-272: this paragraph needs to be rewritten. Is there any difference between precipitation simulated with NCAR_FNL and FNL_FINN? Otherwise synthesise this result by comparing the simulations run with FNL and ERA. What does it mean "both results appear to be higher"? Please rephrase.

We use TRMM observed precipitation to evaluate modeled rainfall in FNL_FINN and ERA-FINN. We have rewritten this paragraph. We have also added more discussions of the spatial and temporal correlations of monthly rainfall between model and observation in different seasons in Section 2.2 of the revised version.

Line 301: LVDs and VLVDs have already been defined so avoid repetitions.

Removed.

Line 332: how can you distinguish the events caused by fires? Is it because your simulations do not include other anthropogenic emissions? Otherwise please explain how you conducted your analyses.

We have revised the related descriptions. Firstly, we have emphasized that many LVDs could be induced by non-fire aerosols, therefore, modeled underestimate of $PM_{2.5}$ concentration and visibility degradation is expected. On the other hand, we used the VLVDs to specifically check the model performance because these events are known to be mainly induced by fire aerosols.

In Section 2.3 of the revised version, a largely revised paragraph now reads 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."

Line 349-362: please rephrase to remove repetitions.

We have modified the paragraph to: "The percentage of LVDs in Singapore has been rapidly increasing since 2012 (Fig. 6c). During the simulation period, this increase appears to be mostly from anthropogenic pollution other than fires, especially in 2012 and 2013. In monthly variation, similar to Kuala Lumpur, two peaks of fire aerosol influence appear in February-March and in September-October, respectively (Fig. 6g). In February and March, the trans-boundary transport of fire aerosols come from mainland Southeast Asia (s1), while in the summer monsoon season fire aerosols come from both Sumatra (s2) and Borneo (s3) (Fig. 7c). Except for the severe haze events in June 2013, VLVDs basically occur in September and October (i.e., 92%) due to both Sumatra and Borneo fires. In general, 34% of LVDs in Singapore are caused by fire aerosols in the FNL_FINN simulation and the rest by local and long-range transported pollutants (Table 3). Nevertheless, fire aerosol is still the major reason for the episodic severe haze conditions." in Lines 375-386 of the revised version.

Results

Line 374-384: this part should be moved to the methods. You need to define earlier how you will conduct your analyses. Also using LVD in equation 3 might be more appropriate than C(i).

We have moved this part to Section 2.4, the "Haze Exposure Day (HED)". We prefer to keep C(i) instead of LVD because LVD is defined as a day with visibility equal or lower than 10 km. However, C(i) represents the annual LVDs which means the sum of LVDs for each year.

Line 432: here it would be also interesting to compare with the WHO limits (i.e. the limit for annual mean $PM_{2.5}$ is 10 $\mu g\ m^{-3}$).

The sentence has been modified to "In the FNL_FINN simulation, the seasonal mean concentration of $PM_{2.5}$ within the planetary boundary layer (PBL) can exceed 20 $\mu g\ m^{-3}$ in this region (note that the air quality standard suggested by World Health Origination is 10 $\mu g\ m^{-3}$ for annual mean and 25 $\mu g\ m^{-3}$ for 24-h mean)." in Lines 430-433 of the revised version.

Line 590: Section 4 should be rewritten. The way results are presented is too repetitive and convoluted. It would be also easier for the reader to have some clear sentences summarizing the skills of different models/emissions.

Section 4 has been rewritten. The revisions are well marked in the version showing tracking results.

Figures

Thirteen figures are really too many especially since most of them have several panels. Please select the most critical ones to summarize your findings and move the others to the supplementary material. Also some figures duplicate content shown in other, so either delete them or move to the supplements.

The point has been well taken. We have moved Fig. 3, 10 and 13 in the original version to the supplementary and have removed Fig. 2 and 11.

Figure 1: the number of vertical levels cannot be inferred from the figure, so please remove this part of the sentence from the caption. Also, the letters A-D are not easily readable. Please choose different colors.

We have changed the caption to "Figure 1. Model domain used for simulations. The domain has 432 × 148 grid points with a horizontal resolution of 36 km. Five fire source regions marked in different colors and labeled as s1, s2, s3, s4 and s5, represent mainland Southeast Asia (s1), Sumatra and Java islands (s2), Borneo (s3), the rest of Maritime Continent (s4), and northern Australia (s5). A, B, C and D indicate the location of four selected cities: Bangkok (A), Kuala Lumpur (B), Singapore (C) and Kuching (D)."

We have enlarged the font size of the letters of A-D.

Figure 2: $PM_{2.5}$ on the y-axis is not as subscript 2.5. It would be easier for the reader to have the whole name of the regions on top of each panel.

Figure 2 has been removed.

Figure 3: is this the yearly average of the daily means? The units can be put after "precipitation".

The figure shows daily precipitation in 2006 only. We have added the units after "precipitation" as the reviewer suggested. This figure has been moved to the supplementary as Fig. S1.

Figure 5: From panel (a) it is clear that the model highly underestimates observations and a scaling factor is needed. This has to be commented in the text. Could you also start both the y- axes from 0? A scatter plot might also help in quantifying the underestimation or please provide some more statistics for model evaluation.

We have changed Fig. 5 (a) and (b) (the new Fig. 3 (a) and (b)) to let the y-axes start from 0. We have accepted the reviewer's suggestion to add a new scatter plot, Fig. 4, in the revised version to show observed visibility versus modeled visibility in FNL_FINN during known fire events. We have also added discussion of this new figure as:

"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, or 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, or can be 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 different 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 LVDs 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".

Figure 6. What do you mean with "variation"? How did you compute it? Please also report the meaning of the color coding in the caption.

The caption has been changed to "Figure 6. (a) – (d) The percentage of LVDs per year derived using from GSOD visibility observations in Bangkok, Kuala Lumpur, Singapore, and Kuching, respectively. (e) – (h) The percentage of LVDs averaged over 2003-2014, derived using GSOD visibility observations in Bangkok, Kuala Lumpur, Singapore, and Kuching, respectively. Each bar presents the observed LVDs in each year or month. Red color shows the partition of fire-caused LVDs (captured by model) while green color presents non-fire LVDs (observed – modeled)."

Figure 7: Please define "variation" or rephrase. Please do the same for all other figures presenting that wording.

The caption has been changed to "Figure 7. The mean fire $PM_{2.5}$ concentrations attributed to different emission regions (s1 - s5) in: (a) Bangkok, (b) Kuala Lumpur, (c) Singapore and (d) Kuching, are all derived from FNL_FINN simulation and averaged over the period of 2003-2014."

Figure 8: (a) please rephrase saying that the size of the circles indicates the number of days and the colors refer to specific population weights. (b) Please add units on y-axes and mention in the caption the use of different scales.

Added units and days on y-axis. The caption has been changed to "Figure 8. (a) The mean low visibility days (circles) per year from 2003 to 2014 in 50 ASEAN cities. The size of the circles indicates the number of days. The colors refer to population-weighted fraction in the total Haze Exposure Days (HED). (b) Annual population-weighted HED (HED_{pw}) and arithmetic mean HED (HED_{ar}). Fire-caused HED are labeled as $fHED_{pw}$ and $fHED_{ar}$. Units are in days. Note that the y-axes are in different scales."

Figure 9: region s1-s5 are not reported on the panels, so please remove them from the caption and simplify the caption as well. Also it is not clear why you report the results

separately by region instead of on one single figure. Figure 9 is essentially identical to Figure 10 averaging on a different period, so you can have just a four panels figure with on each panel a map showing different seasons and the 5 regions together and two panels with the same for wet scavenging. Otherwise you need to move one of the two figures to the supplements.

We have removed s1-s5 in the caption and removed lines in (f)-(g). We actually have moved Fig. 10 to the supplementary.

Figure 11: this is again a repetition of Figure 7. Either you condense the information in one figure or move some of the material to the supplements. It is very hard to keep in mind so many similar figures and your key message is not delivered effectively.

The reviewer's suggestion has been well taken. We have removed the Fig. 11 in the revised version.

Figure 12: Why do you have y-axes with negative numbers? You are displaying PM concentrations and precipitation, so your minimum value should be zero. This figure again contains information already presented (Figure 11, 7, 13), so please try and condense the figures or move them to the supplements. The captions of all figures should be also more informative on the message you want to deliver to the reader.

We have changed all the y-axes scales to start from 0. We have also removed the original Fig. 2. This discussed figure (i.e., original Fig. 12) now becomes Fig. 2 in the revised version.

Reference:

Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled "online" chemistry within the WRF model, Atmospheric Environment, 39, 10.1016/j.atmosenv.2005.04.027, 2005.

Kiehl, J. T., Schneider, T. L., Rasch, P. J., Barth, M. C., and Wong, J.: Radiative forcing due to sulfate aerosols from simulations with the National Center for Atmospheric Research Community Climate Model, Version 3, Journal of Geophysical Research: Atmospheres, 105, 1441-1457, 10.1029/1999JD900495, 2000.

Kim, D., Wang, C., Ekman, A. M. L., Barth, M. C., and Rasch, P. J.: Distribution and direct radiative forcing of carbonaceous and sulfate aerosols in an interactive size-resolving aerosol–climate model, Journal of Geophysical Research: Atmospheres, 113, D16309, 10.1029/2007jd009756, 2008.

Smith, A., Lott, N., and Vose, R.: The Integrated Surface Database: Recent Developments and Partnerships, Bulletin of the American Meteorological Society, 92, 704-708, doi:10.1175/2011BAMS3015.1, 2011.

Visscher, A. D.: Air Dispersion Modeling: Foundations and Applications, First ed., John Wiley & Sons, Inc., pp. 50, 2013.