

## **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<sub>2.5</sub> 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<sub>2.5</sub> 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 Line 132 of the revised version.

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

We only included fire PM<sub>2.5</sub> 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<sub>2.5</sub> 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  $b_{ext}$ ”*

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<sub>2.5</sub> 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<sub>2.5</sub> 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 hygroscopic 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 hygroscopic 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 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.”

*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<sub>2.5</sub> 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<sub>2.5</sub> is 10 µg m<sup>-3</sup>).*

The sentence has been modified to “In the FNL\_FINN simulation, the seasonal mean concentration of PM<sub>2.5</sub> within the planetary boundary layer (PBL) can exceed 20 µg m<sup>-3</sup> in this region (note that the air quality standard suggested by World Health Organization is 10 µg m<sup>-3</sup> for annual mean and 25 µg m<sup>-3</sup> 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  $432 \times 148$  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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> level reflects the influences of both fire and non-fire aerosols, whereas the modeled PM<sub>2.5</sub> only includes the impact of fire aerosols. We find that the model still predicted clearly high PM<sub>2.5</sub> 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.

## 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<sub>2.5</sub>) in the study. Besides PM<sub>2.5</sub> data in Singapore, there are some PM<sub>10</sub> 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<sub>2.5</sub>. 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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> concentration in Bangkok (Table 4). The lower fire PM<sub>2.5</sub> 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<sub>2.5</sub> 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*<sub>1</sub> to *a*<sub>5</sub> are fitting coefficients given by 0.5532, -0.1034, -1.05, -1.957, 0.3406, respectively. The radius increase of wet particle (*r<sub>wet</sub>*) due to hygroscopic growth will be

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

where *r<sub>dry</sub>* is the radius of dry particle in micron.” has been added in Section 2.4 in the revised version.

(d) The fire PM<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>pw</sub> 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  $\leq 10$  km) and very low visibility days (VLVDs; observed visibility  $\leq 7$  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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> 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.



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**Biomass Burning Aerosols and the Low Visibility Events in Southeast Asia**

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30 **Abstract**

31 Fires including peatland burning in Southeast Asia have become a major concern ~~of~~  
32 ~~the~~ general public as well as governments in the region. This is because ~~that~~ aerosols  
33 emitted from such fires can cause persistent haze events under ~~favorite~~certain weather  
34 conditions in downwind locations, degrading visibility and causing human health issues.  
35 In order to improve our understanding of the spatial-temporal coverage and influence of  
36 biomass burning aerosols in Southeast Asia, we have used surface visibility and particulate  
37 matter concentration observations, ~~added~~supplemented by decadal long (~~2002~~2003 to  
38 2014) simulations using the Weather Research and Forecasting (WRF) model with a fire  
39 aerosol module, driven by high-resolution biomass burning emission inventories. We find  
40 that in the past decade, fire aerosols are responsible for nearly all the events with very low  
41 visibility (< 7km), ~~and~~. Fire aerosols alone are also responsible for a substantial fraction  
42 of the low visibility events (visibility < 10 km) in the major metropolitan areas of Southeast  
43 Asia: ~~38~~up to 39% in Bangkok, ~~35~~36% in Kuala Lumpur, and 34% in Singapore. Biomass  
44 ~~burnings~~burning in ~~Mainland~~mainland Southeast Asia account for the largest  
45 ~~contributor~~contribution to total fire-produced PM<sub>2.5</sub> in Bangkok (99.1%), while biomass  
46 burning in Sumatra is ~~the~~a major contributor to fire-produced PM<sub>2.5</sub> in Kuala Lumpur  
47 (~~49~~50%) and Singapore (41%). To examine the general situation across the region, we  
48 have further defined and derived a new integrated metric for 50 cities of the Association of  
49 Southeast Asian Nations, ~~(ASEAN)~~; i.e., ~~the~~ Haze Exposure Days (HEDs) that measures  
50 the annual exposure days of these cities to low visibility (< 10 km) caused by particulate  
51 matter pollution. It is shown that HEDs have increased steadily in the past decade across  
52 cities with both high and low populations. Fire events alone are found to be responsible

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53 for up to about half of the total HEDs. ~~Therefore, our~~Our result suggests that in order to  
54 improve the overall air quality in Southeast Asia, mitigation policies targeting ~~at~~ both  
55 biomass burning and fossil fuel burning sources need to be ~~put in effect~~implemented.

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56 **1 Introduction**

57 In recent decades, biomass burning has become frequent and widely spread across ~~the~~  
58 ~~mainland of Southeast Asia~~ ~~and~~ the islands of Sumatra and Borneo (Langner et al., 2007;  
59 Carlson et al., 2012; Page et al., 2002; van der Werf et al., 2010). Abundant ~~particulate~~  
60 ~~matters~~ ~~aerosols~~ emitted from such fires cause ~~the~~ haze events to occur in ~~the~~ downwind  
61 locations such as Singapore (Koe et al., 2001; Heil et al., 2007; See et al., 2006), degrading  
62 visibility and threatening ~~on~~ human health (Emmanuel, 2000; Kunii et al., 2002; Johnston  
63 et al., 2012; Mauderly and Chow, 2008). Besides causing air quality issues, ~~the~~ fire  
64 aerosols contain rich carbonaceous compounds such as black carbon (BC) (Fujii et al.,  
65 2014) and thus can reduce sunlight through both absorption and scattering. ~~Based on~~  
66 ~~satellite data and numerical simulations, Tosca et al. (2010) found that tropospheric~~  
67 ~~heating from BC absorption in the Maritime Continent (MC) is  $20.5 \pm 9.3 \text{ W m}^{-2}$ , and~~  
68 ~~the reduction of both surface net shortwave radiation and regional precipitation can~~  
69 ~~be as high as 10% due to the direct and semi direct effects of fire aerosols.~~  
70 ~~Nevertheless, indirect~~ ~~Indirect~~ effects of fire aerosols are even more complicated due to  
71 various cloud types and meteorological conditions in the ~~MC~~ ~~Maritime Continent (MC)~~  
72 (Sekiguchi et al., 2003; Lin et al., 2013; Wu et al., 2013).

73 ~~Majority~~ ~~The majority~~ of present day fires in Southeast Asia ~~occurs~~ ~~occur~~ due to  
74 human ~~interferences: oil palm plantation related~~ ~~interference such as~~ land clearing, ~~for~~  
75 ~~oil palm plantations, other causes of~~ deforestation, ~~and~~ ~~poor~~ peatland management, and  
76 burning of agriculture ~~wastes~~ ~~waste~~ (Dennis et al., 2005; ~~Miriam~~ ~~Marlier~~ et al.,  
77 ~~2015b~~ ~~2015a~~). Certain policies and regulations, ~~such as those~~ regarding, ~~e.g.,~~ migration,  
78 also affect the occurrence of burning events. ~~For example, large~~ ~~Large~~ fires have occurred

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79 since ~~the~~ 1960s in Sumatra; however, the first fire event in Kalimantan happened in the  
80 1980s (Field et al., 2009). Based on economic incentives and population growth in  
81 Southeast Asia, future land-use management will play an important role in determining the  
82 ~~coverageoccurrence~~ of fires across the region (Carlson et al., 2012; ~~MiriamMarlier~~ et al.,  
83 ~~2015a2015b~~).

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84 Besides human interventions, meteorological factors, ~~such as rainfall~~, can also  
85 influence fire initiation, intensity, and duration (Reid et al., 2012; Reid et al., 2015). ~~Of~~  
86 ~~particular importance is rainfall~~. Reid et al. (2012) investigated relationships between fire

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87 hotspot appearance and various climate variabilities as well as meteorological phenomena  
88 in different temporal scales over the MC, including: (1) ~~the~~ El Nino and Southern  
89 Oscillation (ENSO) (Rasmusson and Wallace, 1983; ~~McBride et al., 2003~~) and the Indian  
90 Ocean Dipole (IOD) (Saji et al., 1999); (2) ~~Seasonalseasonal~~ migration of the Inter-

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91 tropical Convergence Zone (ITCZ) and associated Southeast Asia monsoons (Chang et al.,  
92 2005); (3) ~~Intra~~intra-seasonal ~~variabilities such as variability associated with the~~ Madden-  
93 Julian Oscillation (MJO) (Madden and Julian, 1971; ~~Zhang, 2005~~) and the west Sumatran  
94 low (Wu and Hsu, 2009); (4) ~~Wave~~equatorial waves, mesoscale features, and tropical  
95 cyclones; and (5) ~~Convectionsconvection~~. One interesting finding is that the influence of

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96 these factors on fire events varies over different parts of the MC. For example, the fire  
97 signal in ~~aone~~ part of Kalimantan is strongly related to both the monsoons and ENSO. In  
98 contrast, fire activity in Central Sumatra is not ~~as~~ closely tied to the monsoons and ENSO  
99 but MJO ~~signal~~.

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100 ~~Above climate variabilities or~~Climate variability of meteorological phenomena  
101 ~~affectaffects~~, not only biomass burning emissions but also ~~fire aerosol~~ transport of fire

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102 ~~aerosols~~ (Reid et al., 2012). ~~Seasonal~~The seasonal migration of the ITCZ and ~~the~~  
103 ~~associated~~ monsoonal circulation dominate seasonal wind flows, whereas sea ~~breeze,~~  
104 ~~typhoon, or breezes, tropical cyclones, and~~ topography determine air flow ~~in on~~ smaller  
105 spatial ~~scales or shorter~~and temporal scales, ~~– all of them~~these phenomena play  
106 significant roles in determining the transport pathway of fire aerosols (Wang et al., 2013).  
107 For example, during the intense haze episode of June 2013, ~~the a~~ long lasting  
108 ~~situation~~event with a “very unhealthy” air pollution level in Singapore, was actually  
109 caused by ~~an~~ enhanced fire aerosol transport from Sumatra to West Malaysia owing to a  
110 tropical ~~storm~~cyclone located in South China Sea. Recently, using a global chemistry  
111 transport model ~~combining~~combined with a back-trajectory tracer model, Reddington et  
112 al. (2014) attempted to attribute particulate ~~pollutions~~pollution in Singapore ~~over a short~~  
113 ~~time period of 5 years,~~ to different burning sites in surrounding regions ~~over a short time~~  
114 ~~period of 5 years.~~ The coarse 2.8-degree resolution model used in the study, however, has  
115 left many open questions.

116 In this study, we aim to examine and quantify the impact of fire aerosols on the  
117 visibility and air quality of Southeast Asia ~~in over~~ the past decade. Analyses of  
118 observational data and comprehensive regional model ~~simulations~~results have both been  
119 performed in order to improve our understanding of this issue. We firstly describe  
120 methodologies adopted in the study, followed by the results and findings from our  
121 assessment of the fire aerosol on the degradation of visibility in several selected cities and  
122 also ~~in over~~ the ~~great~~whole Southeast Asia. We then discuss the sensitivity of our findings  
123 to the use of different meteorological datasets as well as fire emission inventories. The last  
124 section summarizes and concludes our work.

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125 **2 Methodology**

126 **2.1 The model**

127 ~~In order to address the targeted science question, we have used the Weather~~  
128 ~~Research and Forecasting (WRF) model coupled with chemistry component (WRF-~~  
129 ~~Chem). The WRF model is a compressible, non hydrostatic regional meteorology~~  
130 ~~model that uses the Arakawa C grid and terrain following hydrostatic pressure~~  
131 ~~coordinates, and includes various dynamic cores and physical parameterizations for~~  
132 ~~different scientific purposes (Skamarock et al., 2008). The WRF-Chem model is a~~  
133 ~~version of the standard WRF with an additional interactively coupled model of~~  
134 ~~atmospheric chemistry. WRF-Chem simulates atmospheric evolutions of chemical~~  
135 ~~species including particulate matters concurrently with meteorological fields, using~~  
136 ~~the same grid structure, advection scheme, and physics schemes for sub-grid scale~~  
137 ~~transport as in the standard WRF model (Grell et al., 2005). In this study, we use~~  
138 ~~WRF-Chem version 3.6 with a modified chemistry tracer module instead of a full~~  
139 ~~chemistry package. This is for the purpose to focus on the fire aerosol life cycle as the~~  
140 ~~first step, without involving a much more complicated gaseous and aqueous chemical~~  
141 ~~processing calculations. This configuration also lowers the computational burden~~  
142 ~~substantially, and thus enables~~In this study, we have used the Weather Research and  
143 Forecasting (WRF) model coupled with a chemistry component (WRF-Chem) version 3.6  
144 (Grell et al., 2005). Our focus in this study is on the fire aerosol life cycle. Therefore, we  
145 chose to use WRF-Chem with a modified chemical tracer module instead of a full  
146 chemistry package, to thus model the fire PM<sub>2.5</sub> particles as tracers without involving much  
147 more complicated gaseous and aqueous chemical processing calculations but dry and wet

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148 ~~depositions. Emissions of other chemical species were excluded in the simulations. This~~  
149 ~~configuration lowers the computational burden substantially, and thus allows~~ us to conduct  
150 long model integrations to determine the contributions of fire aerosol to the degradation of  
151 ~~air quality visibility~~ in the region over the past decade. ~~In WRF-Chem, the sinks of PM<sub>2.5</sub>~~  
152 ~~particles include dry deposition and wet scavenging calculated at every time step.~~ The  
153 ~~numerical~~ simulations are employed within a model domain with a horizontal resolution  
154 of 36 km, including 432 × 148 horizontal grid points (Fig. 1), and 31 vertically staggered  
155 layers ~~based on a terrain-following pressure coordinate system. The vertical~~  
156 ~~layers that~~ are stretched ~~with to have~~ a higher resolution near the surface (an average depth  
157 of ~30 m in the first model half layer). ~~Variables other than vertical velocity and~~  
158 ~~geopotential are stored at the half model layers.~~ based on a terrain-following pressure  
159 ~~coordinate system.~~ The time step is 180 seconds ~~for advection and physics calculation.~~  
160 The physics schemes included in the simulations are listed in Table 1. The initial and  
161 boundary meteorological conditions are taken from reanalysis meteorological ~~dataset data.~~  
162 In order to examine the potential influence of different reanalysis products on simulation  
163 results, we have used two such datasets: (1) the National Center for Environment Prediction  
164 FiNaL (NCEP-FNL) reanalysis data (National Centers for Environmental Prediction,  
165 2000), which has a spatial resolution of 1 degree and a temporal resolution of 6 hours; and  
166 (2) ERA-Interim, which is a global atmospheric reanalysis from European Centre for  
167 Medium-Range Weather Forecasts (ECMWF) (European Centre for Medium-Range  
168 Weather, 2009), providing 6-hourly atmospheric fields on sixty pressure levels from  
169 surface to 0.1 hPa with a horizontal resolution of approximately 80 km. Sea surface  
170 temperature is updated every 6 hours in both NCEP-FNL and ERA-Interim. All

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171 simulations used four-dimensional data assimilation (FDDA) to nudge NCEP-FNL or  
172 ERA-Interim temperature, water vapor, and zonal ~~and as well as~~ meridional wind speeds  
173 above the planetary boundary layer (PBL). This approach has ~~been~~ shown to provide  
174 realistic temperature, moisture, and wind fields in a long simulation (Stauffer and Seaman,  
175 1994).

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176 ~~In WRF-Chem, the sinks of PM<sub>2.5</sub> particles include dry deposition and wet scavenging~~  
177 ~~calculated at every time step.~~

## 178 **2.2 Biomass burning emissions**

179 ~~Two biomass burning emission inventories are also used in this study to investigate~~  
180 ~~the sensitivity of modeled fire aerosol concentration to different emission~~  
181 ~~estimations estimates.~~ The first emission inventory is the Fire INventory from NCAR  
182 version 1.5 (FINNv1.5) (Wiedinmyer et al., 2011), which classifies burnings of extra  
183 tropical forest, ~~topical~~ ~~tropical~~ forest (including peatland), savanna, and grassland. It is  
184 used in this study to provide daily, 36 km resolution PM<sub>2.5</sub> emissions. The second emission  
185 inventory is the Global Fire Emission Database ~~with~~ version 4.1 with small ~~fire~~ ~~fires~~  
186 included (GFEDv4.1s) (van der Werf et al., 2010; Randerson et al., 2012; Giglio et al.,  
187 2013). GFEDv4.1s provides PM<sub>2.5</sub> emissions with the same spatiotemporal resolution as  
188 FINNv1.5.

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189 A plume rise algorithm for fire emissions was implemented in WRF-Chem by Grell  
190 et al. (2011) to estimate fire injection height. This algorithm, however, often derives an  
191 injection height for tropical peat fire that is too high ~~comparing~~ ~~compared~~ to the estimated  
192 value based on remote sensing retrievals (Tosca et al., 2011). Therefore, we have limited  
193 the plume injection height of peat fire ~~within~~ ~~by a ceiling of~~ 700 m ~~above the ground~~ in this

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194 study based on Tosca et al. (2011). ~~The vertical distribution of emitted aerosols is~~  
195 ~~calculated using the plume model.~~ This modification has clearly improved the modeled  
196 surface PM<sub>2.5</sub> concentration ~~comparing when compared~~ to observations in Singapore.

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197 In order to distinguish the spatial-temporal coverage and influence of biomass burning  
198 aerosols from different regions in Southeast Asia and nearby northern Australia, we have  
199 created five tracers to represent fire aerosols respectively from ~~Mainland mainland~~  
200 Southeast Asia (s1), Sumatra and Java islands (s2), Borneo (s3), the rest of the Maritime  
201 Continent (s4), and northern Australia (s5) as illustrated in Fig. 1. The major fire season  
202 in ~~Mainland mainland~~ Southeast Asia (s1) is from February to April. In ~~the~~ other four  
203 regions (s2-s5), it is from August to October.

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204 Generally speaking, there ~~are is a~~ strong ~~correlation between the~~ seasonal  
205 ~~variations variation~~ of fire emissions ~~coordinating with those and that~~ of rainfall in all fire  
206 regions as shown in Fig. 2. Because ~~Mainland mainland~~ Southeast Asia (s1) and northern  
207 Australia (s5) are on the edge of ~~the~~ seasonal migration of the ITCZ, ~~seasonal variations~~  
208 ~~of rainfall the correlation~~ in these two regions ~~are is~~ even more pronounced. ~~On the other~~  
209 ~~hand, in~~ Sumatra (s2), Borneo (s3), ~~), and the rest of the~~ Maritime Continent (s4) ~~are all~~  
210 ~~influenced by similar meteorological regimes, i.e., seasonal migration of the ITCZ.~~  
211 ~~However, the passage of MJO events adds more intra-season variability of rainfall and~~  
212 ~~fire emissions in these three regions. Therefore, the seasonal variations of rainfall~~  
213 ~~and fire emissions in s2, s3, and s4 are not as apparent as in the s1 and s5 regions (Fig.~~  
214 ~~2b–d), owing to the influences of multiple scales of precipitation features over these~~  
215 ~~areas. Nevertheless, while~~ inter-seasonal variations of rainfall and fire emissions are  
216 still ~~highly~~ correlated with each other in ~~general, however, fire emissions do exist in some~~

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217 raining seasons (Fig. 2b – d), owing to the precipitation features in multiple scales over  
218 these regions (e.g., the passage of MJO events) and underground peatland burning.

## 219 **2.2 Numerical simulations and model evaluation**

220 Our simulations cover a time period slightly longer than a decade from 2003 to 2014  
221 based on available biomass burning emission estimates. The simulation of each year  
222 started on 1 November of the previous year and lasted for 14 months. The first two months  
223 were used for spin-up.

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224 Three sets of decadal long simulations have been conducted. The first simulation used  
225 NCEP-FNL reanalysis data and the FINNv1.5 fire emission inventory. This simulation is  
226 hereafter referred to as FNL\_FINN and is discussed as the base simulation. In order to  
227 examine the influence of different meteorological inputs on fire aerosol life cycle, the  
228 second simulation was conducted using the same FINNv1.5 fire emission inventory as in  
229 FNL\_FINN but different reanalysis dataset, the ERA-Interim, and is referred to as  
230 ERA\_FINN. In addition, to investigate the variability of fire aerosol concentration brought  
231 by the use of different estimates of fire emissions, the third simulation, FNL\_GFED, was  
232 driven by the same NCEP-FNL meteorological input as in FNL\_FINN but with a different  
233 fire emission inventory, the GFEDv4.1s. Note that the simulation period from 2003 to  
234 2014 of all these simulations was solely decided based on the temporal coverage of  
235 GFEDv4.1s.

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236 Precipitation and wind are two key factors in determining the transport and scavenging  
237 of fire aerosols. They are also the variables we use to evaluate the model's performance in  
238 simulating meteorological features. The WRF simulation driven by NCEP-FNL reanalysis  
239 data, the FNL\_FINN run, produced a monthly mean precipitation of  $6.80 \pm 0.55$  mm day<sup>-1</sup>

240 over the modeled domain for the period from 2003 to 2014, very close to the value of  
241 6.30±0.43 mm day<sup>-1</sup> produced in another simulation driven by ERA-Interim, the  
242 ERA\_FINN run. However, the average rainfall in both runs appears to be higher than the  
243 monthly mean of 4.71±0.37 mm day<sup>-1</sup> from the satellite-retrieved precipitation of the  
244 Tropical Rainfall Measuring Mission (TRMM) 3B43 (V7) dataset (Huffman et al., 2007).  
245 Based on the sensitivity tests for FDDA grid nudging, the wet bias in both experiments  
246 mainly comes from water vapor nudging. Figure S1a – c are the Hovmöller plots of daily  
247 TRMM, FNL FINN, and ERA FINN precipitation in 2006, respectively. Compared to  
248 the satellite-retrieved data, both FNL FINN and ERA FINN have produced more light  
249 rain events, and this appears to be the reason behind the model precipitation bias. Despite  
250 the model overestimate in average total precipitation, the temporal correlation of monthly  
251 rainfall between FNL FINN and TRMM is 0.68 and the spatial correlation is 0.85 during  
252 2003-2014 (Table 2). For ERA FINN, the temporal correlation with TRMM is 0.90, while  
253 the spatial correlation is 0.85. In the summer monsoon season (i.e., May, June and July),  
254 both runs show the highest temporal correlations with observation but the lowest in the  
255 spatial correlations. The comparisons show that simulated rainfall generally agrees with  
256 the observation in space and time, especially when ERA-Interim reanalysis is used (i.e., in  
257 ERA FINN). ~~these three~~

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258 The representative wind pattern in Southeast Asia is the monsoon wind flow. In the  
259 winter monsoon season (i.e., February, March and April), mean surface winds are from  
260 northeast in the Northern Hemisphere and turn to the northwesterly once past the Equator  
261 (Fig. S2a). On the other hand, the wind directions are reversed in the summer monsoon  
262 season (i.e., August, September and October) (Fig. S2b). We use the wind data from

NCEP-FNL and ERA-Interim reanalysis to evaluate model simulated winds. We find that both runs overestimated the u component (stronger easterly) in South China Sea (Fig. S3a and c) in the winter monsoon season, and overestimated the v component (stronger southerly) in Java Sea in the summer monsoon season (Fig. S3b and d). These regions are the entrances of monsoon wind flow into the MC. In general, model has well captured the general wind flows in Southeast Asia during both monsoon seasons but overestimated about 1 m sec<sup>-1</sup> in wind speed in some regions (see additional discussion in Section 4), likely due to terrain effect and model resolution limitation.

**2.3 Observational data and model derivation of visibility**

The definition of “visibility” is the farthest distance at which one can see a large, black object against a bright background at the horizon (Seinfeld and Pandis, 2006). There are several factors to determine determining visibility, but in this study here we mainly consider the absorption and scattering of light by gases and aerosol particles, excluding fog or misty days. One of In this study, the most widely used equations, visibility is calculated by using the Koschmeider equation, is given by:

$$VIS = 3.912 / b_{ext}, \tag{1}$$

where VIS is visibility with a unit in meter and  $b_{ext}$  is the extinction coefficient with a unit of m<sup>-1</sup>. Visibility-Excluding fog, visibility degradation is most readily observed from the impact of particulate pollution besides fog. 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 on in the order of 10 km is considered as under a moderately moderate, to heavily polluted heavy air pollution by particulate matters. matter (Visscher, 2013). Abnormal and persistent low visibility situations are also referred to as “haze” events.

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286 ~~Urban air pollutions~~ Air pollution sources such as fossil fuel burning, can cause low  
287 visibility and haze ~~event~~ events to occur. Similarly, fire aerosols, alone or mixed with other  
288 particulate pollutants, can degrade visibility by increasing  $b_{ext}$  and lead to occurrence of  
289 haze events too.

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290 The observational data of visibility from the Global Surface Summary of the Day  
291 (GSOD) (Smith et al., 2011) are used in our study, as to identify days under particulate  
292 pollution, i.e., haze events. The GSOD is derived from the Integrated Surface Hourly (ISH)  
293 dataset and archived at the National Climatic Data Center (NCDC). The daily visibility in  
294 the dataset is available from 1973 to the present.

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295 ~~In order to compare with observations, we also calculate the~~ The observed  
296 visibility using is also used to evaluate the modeled fire aerosol data, visibility and thus  
297 PM<sub>2.5</sub> concentration. The modeled visibility is derived based on the extinction coefficient  
298 ~~of these~~ the fire aerosols as ~~functions~~ a function of particle size ~~f,~~ by assuming a log-normal  
299 size distribution of accumulation mode ~~with a standard deviation  $\sigma = 2$ ,~~ the complex  
300 refractive index of the particles, and a wavelength of 550 nm of the incident light. As  
301 fire plumes contain both sulfur compounds and carbonaceous aerosols, we assume  
302 the fire aerosols are aged internal mixtures with black carbon as core and sulfate as  
303 shell (Kim et al., 2008). ~~We also consider hygroscopic growth of sulfate fraction of~~  
304 these mixed particles in the calculation based on environmental relative humidity.

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305 Note that all these calculations are done for the wavelength of 550 nm unless  
306 otherwise indicated. As fire plumes contain both sulfur compounds and carbonaceous  
307 aerosols, we assume the fire aerosols are aged internal mixtures with black carbon as the  
308 core and sulfate as the shell (Kim et al., 2008). To make the calculated visibility of the fire

309 aerosols better match the reality, we have also considered hygroscopic growth of sulfate  
310 fraction of these mixed particles in the calculation based on the modeled relative humidity  
311 (RH). Based on Kiehl et al. (2000), the hygroscopic growth factor (rhf) is given by

312 
$$rhf = 1.0 + \exp\left(a_1 + \frac{a_2}{RH+a_3} + \frac{a_4}{RH+a_5}\right) \quad (2)$$

313 where  $a_1$  to  $a_5$  are fitting coefficients given by 0.5532, -0.1034, -1.05, -1.957, 0.3406,  
314 respectively. The radius increase of wet particle ( $r_{wet}$ ) due to hygroscopic growth will be

315 
$$r_{wet} = r_{dry}^{rhf} \quad (3)$$

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

317 As mentioned above, a visibility of 10 km is considered ~~as under moderately to~~  
318 ~~heavily~~an indicator for a moderate to heavy particulate pollution ~~so that this quantity.~~

319 Hence a visibility of 10km in observation is used as the threshold for ~~deriving~~defining the  
320 “low visibility day (VLD)” in our study. ~~In analysis, we derived~~We firstly derived the  
321 ~~observed~~ low visibility days in every year for a given city using the GSOD visibility data.

322 ~~Such day is identified when the daily averaged visibility in the observation site is~~  
323 ~~lower or equal to 10 km.~~ Then, we derived the modeled low visibility days ~~in~~following

324 the same procedure, but using modeled visibility data that were only influenced by fire  
325 aerosols. Both the observed and modeled visibilities were then used to define the fraction

326 of low visibility days ~~that can be~~ caused by fire aerosols ~~alone~~. It is assumed that whenever  
327 fire aerosol ~~alone~~, could cause a low visibility day to occur, such a day would be attributed

328 to fire aerosol caused LVD, regardless ~~of~~ whether other coexisting pollutants would have  
329 ~~ana sufficient~~ intensity to cause low visibility or not. ~~We~~In addition to the LVD, we have

330 also used a daily visibility of 7 km as the criterion to define the ~~observed~~ “very low  
331 visibility day (VLVD)”. Such heavy haze events in the region are generally caused by

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332 severe fire aerosol pollution, thus we use their occurrence specifically to evaluate the model  
333 performance.

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#### 334 **2.4 The “Haze Exposure Day (HED)”**

335 We have derived a metric, the Haze Exposure Day (HED), to measure the exposure of  
336 the whole Southeast Asia, represented by 50 cities of the Association of Southeast Asian  
337 Nations (ASEAN), to low visibility events. HED can be defined in a population weighted  
338 format for the analyzed 50 cities, indicating the relative exposure of the populations in  
339 these cities to the low visibility events caused by particulate pollution;

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$$340 \quad HED_{pw} = \sum_{i=1}^N C_{pw}(i) \quad (4)$$

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341 where

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$$342 \quad C_{pw}(i) = \text{pop}(i) \cdot C(i) / \sum_{i=1}^N \text{pop}(i) \quad (5)$$

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343 is the population-weighted fraction of the total Haze Exposure Days.  $N$  equals to the total  
344 number of cities (50),  $i$  is the index for the 50 analyzed cities,  $\text{pop}(i)$  is the population for  
345 a given city (Table S1), and  $C(i)$  represents the annual LVDs for that city calculated from  
346 the GSOD dataset. Note that we assume that the population of each city stays constant  
347 throughout the analyzed period. Another assumption of  $HED_{pw}$  is that everyone in a given  
348 city would be equally exposed to the particulate pollution.

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349 In addition, HED can be also defined in an arithmetic mean format, assuming each city  
350 weights equally regardless of its population. Its value hence emphasizes on the relative  
351 exposure of each area within the analyzed region:

$$352 \quad HED_{ar} = \sum_{i=1}^N C(i) / N \quad (6)$$



353 ~~Both  $HED_{pw}$  and  $HED_{ar}$  can be also calculated using fire-caused LVDs to define the~~  
354 ~~absolute and relative contributions of fire aerosols to the total low visibility events in the~~  
355 ~~region. We will label the fire-caused HED as  $fHED_{pw}$  and  $fHED_{ar}$  thereafter.~~

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#### 356 ~~2.4 Numerical simulations~~

357 ~~Our simulations cover a time period slightly longer than a decade from 2002 to~~  
358 ~~2014 based on availability of biomass burning emission estimations. The simulation~~  
359 ~~of each year started on 1 November of the previous year and lasted for 14 months. The~~  
360 ~~first two months are used for spin-up.~~

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361 ~~Three sets of decadal long simulations have been conducted. The first simulation~~  
362 ~~used reanalysis data of NCEP-FNL and fire emission inventory of FINNv1.5. This~~  
363 ~~simulation is hereafter referred to as FNL\_FINN and discussed as the base simulation.~~  
364 ~~In order to examine the influence of different meteorological inputs on fire aerosol~~  
365 ~~life cycle, the second simulation was conducted using the same FINNv1.5 fire~~  
366 ~~emission inventory as in FNL\_FINN but a different reanalysis data of ERA\_Interim,~~  
367 ~~referring to as ERA\_FINN. In addition, to investigate the variability of fire aerosol~~  
368 ~~concentration brought by the use of different estimations of fire emissions, the third~~  
369 ~~simulation, FNL\_GFED, was driven by the same NCEP-FNL meteorological input as in~~  
370 ~~FNL\_FINN but a different fire emission inventory, the widely used GFEDv4.1s. Since~~  
371 ~~the daily emission of GFEDv4.1s is only available after 2003, the period of the~~  
372 ~~FNL\_GFED simulation is from 2003 to 2014.~~

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373 ~~Precipitation is one of the key factors in determining the transport and~~  
374 ~~scavenging of fire aerosols. WRF simulation driven by NCAR\_FNL reanalysis data, or~~  
375 ~~the FNL\_FINN run, produced a monthly mean precipitation of  $6.81 \pm 0.55 \text{ mm day}^{-1}$~~

376 over the modeled domain for the period from 2002 to 2014, very close to the value of  
377  $6.29 \pm 0.43$  mm day<sup>-1</sup> produced in another simulation driven by ERA-Interim, or the  
378 ERA-FINN run. Comparing to the monthly mean of  $4.69 \pm 0.38$  mm day<sup>-1</sup> from the  
379 satellite retrieved precipitation in the Tropical Rainfall Measuring Mission (TRMM)  
380 3B43 (V7) dataset (Huffman et al., 2007), however, both results appear to be higher.  
381 ~~Based on the sensitivity tests for FDDA grid nudging, the wet bias in both experiments~~  
382 ~~mainly comes from water vapor nudging.~~ Figure 3a – c are the Hovmöller plot of daily  
383 TRMM, FNL-FINN, and ERA-FINN precipitation in 2006, respectively. Comparing to  
384 the observations, both FNL-FINN and ERA-FINN have produced more light rain  
385 events, and this appears to be the reason behind the model precipitation bias. Despite  
386 the model overestimation in averaged total precipitation, the temporal correlation of  
387 normalized rainfall anomaly between FNL-FINN (ERA-FINN) and TRMM is 0.69  
388 (0.90) and the spatial correlation is 0.86 (0.85) during 2002–2014. The comparisons  
389 show that simulated rainfall generally agrees with the observation in space and time,  
390 especial when ERA-Interim reanalysis is used (i.e., in ERA-FINN).

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### 3 Assessment of the impact of fire aerosols on the visibility in Southeast Asia visibility.

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#### 3.1 Impact of fire aerosols on the visibility in four selected cities

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394 We first to focus our analysis on four selected cities in the region, Bangkok (Thailand),  
395 Kuala Lumpur (Malaysia), Singapore (Singapore), and Kuching (Malaysia), all located  
396 close to the major Southeast fire sites ranging from the mainland to the islands of  
397 Southeast Asia. Specifically, Bangkok is a smoke receptor city of the fire events in the

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398 mainland of Southeast Asia (s1) while Kuala Lumpur and Singapore are two cities  
399 frequently under the influence of Sumatra (s2) as well as Borneo fires (s3). Kuching is in  
400 the ~~eastcoastal~~ area of Borneo ~~so that and~~ directly affected by Borneo fire events (s3).

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401 ~~The low visibility events in these four near fire site cities during the fire seasons~~  
402 ~~from 2002 to 2014, defined as days with daily averaged visibility lower or equal to 10~~  
403 ~~km, or Low Visibility Days (LVDs), have been identified using the daily GSOD visibility~~  
404 ~~database and then compared with modeled results (Fig. 4). We find that the model~~  
405 ~~has reasonably captured the LVDs despite certain biases. Specifically, for the Very~~  
406 ~~Lower Visibility Days (VLVDs), here defined as events with daily averaged visibility~~  
407 ~~lower or equal to 7 km, the modeled and observed results display a good correlation~~  
408 ~~despite a model overestimate in visibility value or underestimate in degrading~~  
409 ~~visibility in certain events. In Southeast Asia, severe haze events equivalent to the~~  
410 ~~VLVDs in visibility degradation are largely caused by fire aerosol pollutions.~~  
411 ~~Assuming this is true, the performance of our model in reproducing the major fire~~  
412 ~~events is very good since only 10% or fewer VLVDs observed in the past decade were~~  
413 ~~not captured by the model (Table 2; Fig. 4). Note that other than these VLVDs, for~~  
414 ~~many LVDs fire aerosol might not be the only reason responsible for the degradation~~  
415 ~~of visibility.~~

416 ~~In addition to the visibility data, we have also obtained the ground based~~  
417 ~~observations of PM<sub>2.5</sub> concentration in recent years from the National Environment~~  
418 ~~Agency (NEA) of Singapore. Figure 5a shows the comparison of time series of~~  
419 ~~observed and FNL\_FINN simulated daily PM<sub>2.5</sub> during 2013-2014. The surface~~  
420 ~~observational data of PM<sub>2.5</sub> concentration among these four cities are only available in~~

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421 Singapore since 2013 from the National Environment Agency (NEA) of Singapore. We  
422 thus firstly used these data along with visibility data to evaluate model's performance for  
423 fire-caused haze events reported in Singapore during 2013-2014 (Fig. 3). Note that the  
424 observed PM<sub>2.5</sub> level reflects the influences of both fire and non-fire aerosols, whereas the  
425 modeled PM<sub>2.5</sub> only includes the impact of fire aerosols. ~~However, We find that the~~ model  
426 still predicted clearly high PM<sub>2.5</sub> concentrations during most of the observed haze events,  
427 especially in June 2013, and in spring and fall seasons of 2014 (highlighted green areas),  
428 though with underestimates in particle concentration of up to 30-50%, likely due to the  
429 ~~model resolution, a model overestimation of rainfall, and the errors in emission~~  
430 ~~inventory. Once again, the model has shown a solid performance in capturing all the~~  
431 ~~major known haze events caused by fire PM in Singapore (Fig. 5b). Specifically to the~~  
432 ~~observed VLVDs, we evidence that fire aerosol is the main reason behind these~~  
433 ~~events~~ model's exclusion of non-fire aerosols, coarse model resolution, overestimated  
434 rainfall, or errors in the emission inventory. Figure 4 shows observed visibility versus  
435 modeled visibility in FNL FINN during the fire events shown in Fig. 3. Note that all these  
436 events have an observed visibility lower than or equal to 10 km, or can be identified as  
437 LVDs. In capturing these fire-caused haze events, the model only missed about 22% of  
438 them, or reporting a visibility larger than 10 km in 40 out of 185 observed LVDs as marked  
439 with purple color in Fig. 4. When observed visibility is between 7 and 10 km, model results  
440 appear to align with observations rather well. For cases with visibility lower than 7 km,  
441 the model captured all the events (by reporting a visibility lower than 10 km, or LVD)  
442 although often overestimated the visibility range. These results imply that the VLVDs only  
443 count a very small fraction in LVDs and thus are episodic events. It is very likely that the

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444 size of concentrated fire plumes in VLVDs might be constantly smaller than the 36 km  
445 model resolution; therefore, the model results could not reach the peak values of PM<sub>2.5</sub>  
446 concentrations of these plumes.

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447 Furthermore, the LVDs in the four selected near-fire-site cities during the fire seasons  
448 from 2003 to 2014 have been identified using the daily GSOD visibility database and then  
449 compared with modeled results (Fig. 5). It is difficult to identify all the fire caused haze  
450 events beyond Singapore even in recent years. However, in Southeast Asia, severe haze  
451 events equivalent to the VLVDs in visibility degradation are known to be largely caused  
452 by fire aerosol pollution. Therefore, we used the observed VLVDs in the four selected  
453 cities to evaluate the performance of the model. We find that the modeled result displays  
454 a good performance in capturing VLVDs despite an overestimate in visibility range during  
455 certain events compared with the observation. The model in general only missed about  
456 10% or fewer VLVDs observed in the past decade (Table 3; Fig. 5). In addition, the model  
457 has reasonably captured the observed LVDs despite certain biases (Fig. 5), likely due to  
458 the fact that fire aerosol might not be the only reason responsible for the degradation of  
459 visibility during many LVDs.

460 We find that the annual mean LVDs in Bangkok has increased from 46%47% (172  
461 days) in the first 5-year period of the simulation duration (2002-20092003-2007) to 74%  
462 (272 days) in the last 5-year period (2010-2014), so does the). The LVDs caused by fire  
463 aerosols has increased as well (Fig. 6a). Overall, fire aerosols are responsible for more  
464 than one third of these LVDs (i.e. 38. 39% in average; Table 23). The largest source of  
465 fire aerosols affecting Bangkok is burning of agriculture waste and other biomass burning  
466 in s1 during the dry season of spring (Fig. 7a; Table 34). During the fire season, abundant

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467 fire aerosols degrade visibility and even cause VLVDs to occur (~~Fig. 6e~~). ~~Ninety-~~  
468 ~~eight percent of VLVDs in Bangkok occurred~~, from December to April (~~Fig. 6c~~). Based  
469 on our model results, ~~99.87%~~ of VLVDs can be identified as fire caused.

470 In Kuala Lumpur, the percentage of LVDs also gradually increases since 2006 to reach  
471 a peak in 2011 and again in 2014 (Fig. 6b). During 2005-2010 the frequency of total LVDs  
472 have increased 10-15% each year, mainly attributing to the pollution sources other than  
473 fires. However, fire-caused LVDs ~~are become~~ more evident after 2009. Seasonal wise,  
474 there are two peaks of fire aerosol influence, one in February-March and another in August  
475 (Fig. 6f), corresponding to the trans-boundary transport of fire aerosols from  
476 ~~Mainland~~ ~~mainland~~ Southeast Asia (s1) in the winter monsoon season and from Sumatra  
477 (s2) in the summer monsoon season, respectively (Fig. 7b). Three quarter of VLVDs ~~are~~  
478 ~~occurred in the summer monsoon season due to Sumatra fires.~~ ~~Noted~~ ~~Note~~ that in  
479 November and December the percentage of LVDs is over 50% and dominated by ~~the~~  
480 pollutants other than fire aerosols. These non-fire aerosols come from either local sources  
481 or the areas further inland riding on the winter monsoon circulation. Overall, fire pollution  
482 is responsible for ~~35% or~~ ~~36%~~, a substantial fraction of total low visibility events in Kuala  
483 Lumpur during ~~2002~~ ~~2003~~-2014 (Table 23).

484 The percentage of LVDs in Singapore has been rapidly increasing since 2012 (Fig.  
485 6c). ~~Except for 2014~~ ~~During the simulation period~~, this increase ~~is appears to be~~ mostly  
486 from anthropogenic pollution other than fires, especially in 2012 and 2013. ~~High~~  
487 ~~percentage of LVDs in November and December could be induced by aerosols from~~  
488 ~~further inland of Mainland Southeast Asia through long range transport driven by the~~  
489 ~~monsoon circulation (Fig. 6g).~~ ~~Similar~~ ~~In monthly variation, similar~~ to Kuala Lumpur,

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490 ~~there are~~ two peaks of fire aerosol influence, ~~one appear~~ in February-March and ~~another~~  
491 ~~in~~ September-October, ~~respectively~~ (Fig. 6g). ~~The~~In February and March, the trans-  
492 boundary ~~transported fire aerosols can transport~~ of fire aerosols come from mainland  
493 ~~Southeast Asia (s1), while in the summer monsoon season fire aerosols~~ come from both  
494 Sumatra (s2) and Borneo (s3) ~~in the summer monsoon season~~ (Fig. 7c). Except for the  
495 severe haze events in June 2013, VLVDs basically occur in September and October (i.e.,  
496 92%) due to both Sumatra and Borneo fires. In general, ~~up to~~ 34% of LVDs in Singapore  
497 are caused by fire aerosols ~~in~~based on the FNL\_FINN simulation and the rest by local and  
498 long-range transported pollutants (Table 2). ~~Fire3~~. Nevertheless, ~~fire~~ aerosol is still the  
499 major reason for the episodic severe haze conditions.

500 Because of its geographic location, Kuching is affected heavily by local fire events  
501 during the fire season (Fig. 7d). Fire aerosols can often degrade the visibility ~~easily~~ to  
502 ~~lower than~~below 7 km and even ~~reach~~reaching 2 km (Fig. 4d5d). The LVDs mainly  
503 occur in August and September during the fire season (Fig. 6d and h). The frequency of  
504 LVDs in Kuching is similar to Singapore; however, 25% of those LVDs are considered to  
505 be VLVDs in Kuching while only 4% are in Singapore in comparison (Table 23).

### 506 3.2 Impact of fire aerosols on the visibility ~~in~~over the ~~greater~~whole Southeast

#### 507 Asia

508 Air quality degradation caused by fires apparently occurs in regions beyond the above-  
509 analyzed four cities. To examine such degradation ~~in~~over the ~~greater~~whole Southeast  
510 Asia, we have extended our analysis to cover 50 cities of the ~~Association of Southeast~~  
511 ~~Asian Nations (ASEAN)~~. The impact of particulate pollution on the ~~greater~~whole  
512 Southeast Asia is measured by ~~a metric of the~~“Haze Exposure Day” (HED). ~~HED can be~~

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513 as defined in a population-weighted format for the 50 analyzed cities, indicating the  
514 relative exposure of the populations in these cities to the low visibility events caused  
515 by particulate pollution, thus calculated as Section 4.1.

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$$516 \quad HED_{pw} = \sum_{i=1}^N C_{pw}(i) \quad (2)$$

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$$518 \quad C_{pw}(i) = \frac{pop(i) \cdot C(i)}{\sum_{i=1}^N pop(i)} \quad (3)$$

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519 where  $N$  equals to the total number of cities, or 50,  $i$  is the index for the 50 analyzed  
520 cities,  $C_{pw}(i)$  is the population-weighted fraction of the total Haze Exposure Days and  
521  $pop(i)$  is the population for a given city,  $C(i)$  represents the annual LVDs for that city  
522 calculated from the GSOD dataset. Note that we assume that the population of each  
523 city is constant throughout the analyzed period. Another assumption of  $HED_{pw}$  is that  
524 everyone in a given city would equally expose to the particulate pollution. The top  
525 four among the 50 cities that made the largest contributions to the  $HED_{pw}$  are Jakarta,  
526 Bangkok, Hanoi, and Yangon, (Fig. 8a), with population ranking of 1, 2, 4, and 5,  
527 respectively (Fig. 8a).

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528 In addition, HED can be also defined in an arithmetic mean format, assuming each city  
529 weights equally regardless of its population. Its value hence emphasizes on the relative  
530 exposure of each area within the analyzed region.

$$531 \quad HED_{ar} = \sum_{i=1}^N C(i)/N, \quad (4)$$



532 ~~Apparently, both  $HED_{pw}$  and  $HED_{ar}$  can be also calculated using~~  
533 ~~fire-caused LVDs (here using the results of FNL\_FINN) to define the~~  
534 ~~absolute and relative contributions of fire aerosols to the total low visibility events in the~~  
535 ~~region. We will label the fire-caused HED as  $fHED_{pw}$  and  $fHED_{ar}$  thereafter.~~

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536 Table S1).

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537 We find that both  $HED_{pw}$  and  $HED_{ar}$  increase rather steadily over the past decade (Fig.  
538 8b), demonstrating that the exposure to haze events either weighted by population or not  
539 has become worse in the region. Generally speaking, the fire aerosols are responsible for  
540 up to 40-60% of the total ~~exposures~~ exposure to low visibility across the region. In both  
541 measures, the increase of fire-caused HED (2.64 and 3.37 days per year for population-  
542 weighted and arithmetic mean, respectively) is similar to that of overall HED (2.61 and  
543 3.59 days per year for population-weighted and arithmetic mean, respectively) (Fig. 8b),  
544 suggesting that fire aerosol has taken the major role in causing the degradation of air quality  
545 in Southeast Asia ~~comparing~~ compared to the non-fire particulate pollution. The result that  
546  $HED_{pw}$  is higher than  $HED_{ar}$  in most of the years indicates that the particulate pollution is  
547 on average worse over more populous cities than the others. Interestingly, the discrepancy  
548 of these two variables, however, has become smaller in recent years and even reversed in  
549 2014, implying an equally worsening of haze event occurrence across from the smaller to  
550 ~~the~~ bigger cities in terms of population in the region. The reason behind this ~~result~~ could  
551 be a ~~widely~~ wider spread of fire events in the region, ~~particularly~~ causing acute haze events  
552 in ~~the~~ cities ~~even~~ with relatively low populations. Regarding the increase of fire-caused  
553 HED, because biomass burning, especially peatland burning, usually occurs in the rural  
554 areas, higher fire emissions would extend low visibility ~~condition~~ conditions to a larger

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555 area regardless of its population. On the other hand, ~~air pollution caused by due to~~  
556 industrialization, urbanization, and other factors such as population growth ~~increases~~  
557 ~~rapidly, air pollution has become worse~~, across the region so ~~that~~, even cities with lower  
558 ~~population~~ ~~populations~~ now increasingly suffer from low visibility from fossil fuel burning  
559 and other sources of particulate pollution. Therefore, the mitigation of air quality  
560 degradation needs to consider both fire and non-fire sources.

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### 561 3.3 The influence of wind and precipitation on fire aerosol life cycle

562 Seasonal migrations of the ITCZ and associated summer and winter monsoons  
563 dominate seasonal wind flows that drive fire aerosol transport. Additionally, as discussed  
564 previously, certain small-scale or short-term phenomena such as sea ~~breeze,~~  
565 ~~typhoon breezes, typhoons,~~ and topography-forced circulations also play important roles  
566 in distributing fire aerosols. Nevertheless, we focus our ~~discussions~~ ~~discussion~~ here on the  
567 former.

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568 ~~From~~ February to April is the main fire season in ~~Mainland~~ ~~mainland~~ Southeast Asia  
569 (s1). In the FNL\_FINN simulation, ~~the~~ seasonal mean concentration of PM<sub>2.5</sub> within the  
570 ~~planetary boundary layer (PBL)~~ can exceed 20  $\mu\text{g m}^{-3}$  in this region. ~~(note that the air~~  
571 ~~quality standard suggested by World Health Origination is 10  $\mu\text{g m}^{-3}$  for annual mean and~~  
572 ~~25  $\mu\text{g m}^{-3}$  for 24-h mean).~~ During this fire season, the most common wind direction is  
573 from northeast to southwest across the region (Fig. 9a). Fire aerosol plumes with  
574 ~~concentration~~ ~~concentrations~~ higher than 0.1  $\mu\text{g m}^{-3}$  can ~~transport with the main~~  
575 ~~wind~~ ~~be transported~~ westward as far as 7000 km from the burning sites. In contrast,  
576 February to April is not the typical burning season in the islands. Low fire emissions ~~added~~

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577 ~~by in combination with~~ a lack of long-range transport of fire aerosols from the mainland  
578 due to the seasonal circulation result in a low PM<sub>2.5</sub> level over these regions (Fig. 9b - d).

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579 Wet scavenging is a major factor ~~to determinedetermining~~ the lifetime and thus  
580 abundance of suspended fire aerosols in the air. The effect of wet scavenging of fire  
581 aerosols is reflected from the wet scavenging time calculated using the modeled results:

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582 ~~The wet scavenging time, which~~ is a ratio of ~~the~~ aerosol mass concentration ~~and to the~~  
583 scavenging rate, ~~the latter is~~ (a function of precipitation rate). Thus, short scavenging  
584 ~~timetimes~~ often ~~indicatesindicate~~ high scavenging ~~raterates~~ except for the sites with

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585 extremely low aerosol concentration. During February-April, at the ITCZ's furthest  
586 southern extent, the short scavenging time < 1 day around 10°S shows a quick removal of  
587 fire aerosols by heavy precipitation ~~that has prevented~~ preventing the southward

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588 transport of aerosols (Fig. 9f). ~~Whereas~~ ~~On the other hand~~, the long scavenging time (> 5  
589 days) in the Western Pacific warm pool, South China Sea, the Indochina peninsula, Bay of  
590 Bengal, and Arabian Sea leads to a long suspending time of aerosols transported to these

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591 regions. During the same season, over the islands of Sumatra and Borneo, the abundance  
592 ~~along with the likelihood of being transported to other places~~ of fire aerosols, either  
593 emitted locally or trans-boundary transported, are greatly limited by the high scavenging

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594 rate (short scavenging time) over ~~this~~ ~~these~~ regions (Fig. 9g and h). ~~The~~ South China Sea  
595 ~~is in a dry condition~~ ~~has little precipitation~~ during this time period; therefore, fire aerosols  
596 from the northern part of ~~Philippine~~ ~~the Philippines~~ can be transported to this region and

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597 stay longer than 5 days (Fig. 9i).

598 The months of August to October, when the ITCZ reaches its furthest northern extent,  
599 mark the major fire season of Sumatra, Borneo, and some other islands in the ~~Maritime~~

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600 ~~Continent~~MC (Fig. ~~10bS5b~~ - d). Australia fires also mainly occur in this season (Fig.  
601 ~~10eS5c~~). Mean wind flows are from southeast to northwest in the Southern Hemisphere,  
602 and turn to the northeast direction once ~~passpast~~ the Equator. Within the MC the seasonal  
603 variation of rainfall is small during this time, ~~with~~ heavy precipitation and thus short  
604 scavenging ~~times~~ (< 3 days) ~~mostly exist~~existing along the MJO path (Fig. ~~10fS5f~~ -  
605 i) (Wu and Hsu, 2009). The high scavenging rate in the regions close to the fire sites in  
606 the islands shortens the transport distance of fire aerosol plumes with PM<sub>2.5</sub> concentration  
607 > 0.1  $\mu\text{g m}^{-3}$  to less than 3000 km (Fig. ~~10bS5b~~ - d). Long scavenging ~~times~~ (> 5  
608 days) ~~primarily exists~~exist in the Banda Sea and northern Australia due to the ITCZ  
609 location. Fire aerosols from Java ~~Island~~(s2) (Fig. ~~10gS5g~~), Papua New Guinea (s4) (Fig.  
610 ~~10iS5i~~), and northern Australia (s5) (Fig. ~~10jS5j~~) can thus ~~suspend~~be suspended in the air  
611 for a relatively long time over these regions.

612 The above-discussed seasonal features of precipitation and aerosol scavenging  
613 ~~strength~~rate help us to better understand the variability of haze occurrence and also to  
614 identify the major source regions of fire aerosols influencing selected Southeast Asian  
615 cities (Fig. 7). For example, the geographic location of Bangkok, which is inside the s1  
616 emission region, determines that ~~about 99%~~ ~~nearly all the~~ fire aerosols ~~is~~(99%) are from  
617 sources within the region from December to April (Fig. 7a and Table 34). Fire aerosols  
618 from all the other burning sites stay at very low ~~level~~levels even during the burning seasons  
619 there due to circulation and precipitation scavenging. For Kuala Lumpur and Singapore,  
620 over 90% of ~~total~~the fire aerosols ~~reached~~reaching both cities come from  
621 ~~Mainland~~mainland Southeast Asia (s1) in January–April due to the dominant winter  
622 monsoon circulation. During May-October, however, the major sources of fire aerosols

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623 shift to Sumatra (s2) and Borneo (s3) ~~aiding~~aided by northward wind (Fig. ~~10bS5b~~10bS5b and c).  
624 The monthly variations of PM<sub>2.5</sub> concentration in Kuala Lumpur and Singapore also have  
625 a largely similar pattern (Fig. ~~8b7b~~8b7b and d). The annual mean contribution of different  
626 emission regions in Kuala Lumpur are 43% from ~~Mainland~~mainland Southeast Asia (s1),  
627 ~~49.50%~~49.50% from Sumatra (s2), 4% from Borneo (s3), 3% from the rest of Maritime Continent  
628 (s4), and 0.43% from northern Australia (s5) in FINL\_FINN (Table 34). Similar to Kuala  
629 Lumpur, there are two peak seasons of the monthly low visibility days contributed by fire  
630 aerosols in Singapore (Fig. 6g), well correlated with modeled high fire PM<sub>2.5</sub> concentration  
631 (Fig. 7c). The low visibility days in March and April mainly are caused by fire aerosols  
632 from ~~Mainland~~mainland Southeast Asia (s1) under southward wind pattern (Fig. 9a), and  
633 those in May to October are affected by Sumatra (s2) first in May to June, and then by both  
634 s2 and s3 (Borneo) during August to October due to north- or northwest-ward monsoonal  
635 circulation (Fig. ~~10bS5b~~10bS5b and c; also Table 34). Kuching, similar to Bangkok, is strongly  
636 affected by local fire aerosols (s3) during the fire season (July – October). The annual  
637 mean contribution from Borneo (s3) is 85% ~~while%, with~~ only 78% from  
638 ~~Mainland~~mainland Southeast Asia (s1) and 5% from Sumatra (s2) (Table 34).  
639 Reddington et al. (2014) applied two different models, a 3D global chemical transport  
640 model and a Lagrangian ~~atmospheric transport~~tracer model to examine the long-term  
641 mean contributions of fire emissions ~~to PM<sub>2.5</sub>~~ from different regions ~~to PM<sub>2.5</sub> in several~~  
642 ~~cities~~ in Southeast Asia. ~~The~~Their estimated contribution from ~~Mainland~~mainland  
643 Southeast Asia to the above-discussed four selected cities was lower than our result during  
644 January-May, likely due to their use of a different emission inventory and the coarse  
645 resolution of their global model. The FINNv1.5 dataset used in our study specifically

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646 provides higher PM<sub>2.5</sub> emissions from agriculture fires (the major fire type in  
647 ~~Mainland~~mainland Southeast Asia) than GFED4.1s does, ~~the~~ the latter is an updated version  
648 ~~of~~ the dataset (GFEDv3) used in Reddington et al. (2014) (Fig. 2). The ~~detail~~detailed  
649 comparison of FNL\_FINN and FNL\_GFED will be discussed in the following section.

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#### 650 4 Influence of different ~~reanalysis~~meteorological datasets and emission 651 inventories on modeled fire aerosol abundance

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652 As discussed in the previous section, meteorological conditions, particularly wind field  
653 and precipitation, could substantially influence the life cycle and transport path of fire  
654 aerosols during the fire ~~reasons; therefore~~seasons. ~~Therefore~~, it is necessary to examine  
655 ~~any~~ potential ~~discrepancies~~discrepancy in modeled particulate matter abundance  
656 ~~attributing to~~arising from the use of different meteorological datasets.

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657 ~~When~~ comparing ~~the~~ two of our simulations, one ~~was~~ driven by the ~~NCAR\_NCEP-~~  
658 ~~FNL~~ (i.e., FNL\_FINN), ~~another~~ and the ~~other~~ by the ERA-Interim (i.e., ERA\_FINN)  
659 meteorological input, we find that the ERA\_FINN run consistently produces less  
660 precipitation than ~~the~~ FNL\_FINN run during the ~~raining~~rainy seasons over ~~the~~ past decade

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661 (Fig. 2) ~~(c)~~ also see the comparison results of both runs with observations in Section 2.4.2.

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662 Regarding fire aerosol life cycle, less rainfall in ERA\_FINN results in ~~a~~ weaker wet  
663 scavenging ~~condition~~ and thus higher abundance of fire ~~aerosol concentration~~aerosols

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664 than in FNL\_FINN. We find that ~~the~~ annual mean concentration of fire PM<sub>2.5</sub> produced in  
665 the ERA\_FINN run in Bangkok, Kuala Lumpur, Singapore, and Kuching is ~~8.89~~2, 5.48,

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666 3.4, and 7.97  $\mu\text{g m}^{-3}$ , respectively, clearly higher than the corresponding results of the

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667 FNL\_FINN run of ~~8.0~~4.95, 5.3, 3.0, and ~~7.16~~9  $\mu\text{g m}^{-3}$  (Table 34). In ~~Mainland~~

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668 Southeast Asia, a twenty-one percent lower rainfall in ERA\_FINN causes the  
669 significantly different general fire PM<sub>2.5</sub> concentration comparing to in ERA\_FINN is  
670 about 10% higher than in FNL\_FINN result in. However, the occurrence of low visibility  
671 events is less sensitive to the fire season (February–April) (Fig. 2a and 11a). In Kuala  
672 Lumpur differences in rainfall in places near the burning areas such as Bangkok and  
673 Kuching, as indicated by a nearly negligible enhancement of VLVDs in the ERA\_FINN  
674 run in Bangkok and Kuching (~1%) (Table 3). In comparison, the difference in fire PM<sub>2.5</sub>  
675 concentration wind field between these two runs mainly comes from Sumatra (s2)  
676 during June to September; however, in Singapore and Kuching the concentration  
677 difference comes from both Sumatra (s2) and Borneo (s3) in August to October (Fig.  
678 11b–d and Table 3), all corresponding to the discrepancy of rainfall between  
679 FNL\_FINN and ERA\_FINN in these regions (Fig. 2b and c). has a much smaller impact  
680 than that of precipitation on modeled particulate matter abundance.

681 The difference in aerosol scavenging between ERA\_FINN and FNL\_FINN extends  
682 to a difference as high as 7% and 12% in the resulted LVDs of Bangkok and Kuching,  
683 respectively, (Table 2), though its influence on the results of Kuala Lumpur and  
684 Singapore is much smaller (3~4%). In general, fire PM<sub>2.5</sub> concentration in ERA\_FINN  
685 is about 10% higher than in FNL\_FINN; however, the substantial impact of fire  
686 aerosols on LVDs is more sensitive in places near the burning areas, i.e., Bangkok and  
687 Kuching. Interestingly, a mild increase of VLVDs in the ERA\_FINN run in Bangkok and  
688 Kuching (~1%) (Table 2) implies that the occurrence of severe haze events is less  
689 affected by the rainfall difference in the burning areas.

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690 In addition to meteorological inputs, ~~differences various using different~~ fire emission  
691 ~~estimations estimates~~ could also affect the modeled results. To examine such an influence,  
692 we have compared two simulations with the same meteorological input but different fire  
693 emission inventories, the FNL\_FINN using FINNv1.5 and FNL\_GFED using GFEDv4.1s.  
694 The main differences between the two emission inventories appear mostly in  
695 ~~Mainland mainland~~ Southeast Asia (s1) and northern Australia (s5) (Fig. 2a and e, Fig. 12a  
696 and e). ~~For instance, the peak month of fire PM<sub>2.5</sub> concentration in Bangkok shifts~~  
697 ~~from March in FNL\_FINN to January in FNL\_GFED (Fig. 11a), owing to the difference~~  
698 ~~in temporal pattern between the two fire emission inventories (Fig. 2a). Comparing,~~  
699 ~~Compared~~ to FINNv1.5, fire emissions in GFEDv4.1s over ~~Mainland mainland~~ Southeast  
700 Asia are more than 66% lower (Fig. 2a), and this results in a ~~4043~~% lower fire PM<sub>2.5</sub>  
701 ~~concentration~~ in Bangkok (Fig. 11a and Table 34). The lower fire PM<sub>2.5</sub> concentration in  
702 FNL\_GFED actually ~~produced produces~~ a visibility that matches better with  
703 ~~observation observations~~ in Bangkok comparing to the result of FNL\_FINN (Fig. S1a).  
704 ~~The difference in monthly S5a). This implies that the fire emissions over the islands~~  
705 ~~between the two emission inventories is small, with the fire emission in FINNv1.5~~  
706 ~~generally higher than that in GFEDv4.1s (Fig. 2b – d). However, fire emissions in~~  
707 ~~GFEDv4.1s are much higher during the fire season in the dry years (i.e. 2004, 2006~~  
708 ~~and 2009) over s2 and s3 (Fig. 12b and c), leading to a modeled mean PM<sub>2.5</sub>~~  
709 ~~concentration by FNL\_GFED in Kuala Lumpur and Singapore that is higher than that~~  
710 ~~by FNL\_FINN during the fire season (Fig. 11b and c). On the other hand, the higher~~  
711 ~~PM<sub>2.5</sub> concentration simulated in FNL\_GFED during the June 2013 severe haze event~~  
712 ~~in Kuala Lumpur and Singapore is due to the spatiotemporal distribution of fire spots~~

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713 ~~rather than absolute perhaps overestimated in mainland Southeast Asia. In northern~~  
714 ~~Australia, fire aerosol emissions. Based on our simulations, fire aerosols from Sumatra~~  
715 ~~(s2) are mainly responsible for the severe haze event in June 2013 (Fig. 7b – c and~~  
716 ~~Fig. S2b – c). During this event, the total amount of fire emissions in Sumatra (s2) is~~  
717 ~~lower in GFEDv4.1s than FINNv1.5, however, distributed rather more densely over a~~  
718 ~~smaller area (Fig. 13c and d). As a result, under the same meteorological condition,~~  
719 ~~the simulated PM<sub>2.5</sub> in the FNL\_GFED simulation reaches Singapore in a higher~~  
720 ~~concentration that also matches better with observation than the result of FNL\_FINN~~  
721 ~~(Fig. 13b). A similar result also appears in Kuching, where the difference in modeled~~  
722 ~~PM<sub>2.5</sub> concentration between the two model runs is likely related to the difference in~~  
723 ~~spatial or temporal distributions rather than the mean quantities of PM<sub>2.5</sub> emissions~~  
724 ~~since the latter suggested by FINNv1.5 are almost the same in both fire emissions~~  
725 ~~inventories.~~

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726 ~~The most evident difference between the two emission inventories occurs in~~  
727 ~~northern Australia, where FINNv1.5 suggests an almost negligible fire aerosol~~  
728 ~~emission comparing compared to GFEDv4.1s (Fig. 2e). Therefore, in the FNL\_GFED~~  
729 ~~simulation, Australia fire aerosols play an important role in Singapore air quality,~~  
730 ~~contributing to about 22% of the modeled PM<sub>2.5</sub> concentration in Singapore. In contrast,~~  
731 ~~Australia fires have nearly no effect on Singapore air quality in the FNL\_FINN run (Table~~  
732 ~~3). Our results raise the important issue of the sensitivity of modeled aerosol~~  
733 ~~concentration in downwind areas to the spatiotemporal distribution, besides the~~  
734 ~~absolute emission amount from the fire spots. A further study regarding this topic~~  
735 ~~would be much needed.4).~~

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736 We would also like to point out the importance of spatiotemporal distribution of fire  
737 emission to the modeled results. For example, during the June 2013 severe haze event in  
738 Kuala Lumpur and Singapore, the total amount of fire emissions from Sumatra (s2) in  
739 GFEDv4.1s are lower than those of FINNv1.5 (Fig. S6a) but distributed rather more  
740 densely over a smaller area (Fig. S6c and d). As a result, under the same meteorological  
741 conditions, the simulated PM<sub>2.5</sub> in the FNL GFED simulation reaches Singapore in a  
742 higher concentration that also matches better with observations than the result of  
743 FNL FINN (Fig. S7b).

## 744 **5 Summary and Conclusions**

745 We have examined the extent of the biomass burning aerosol's impact on the air  
746 quality of Southeast Asia in the past decade using surface visibility and ~~surface~~ PM<sub>2.5</sub>  
747 measurements along with the WRF model with a modified fire tracer module. The model  
748 has shown a good performance in capturing 90% of the observed severe haze events  
749 (visibility < 7 km) caused by fire aerosols occurred ~~in over~~ past decade in several cities that  
750 are close to the major burning sites. ~~Such events are known to be induced mainly by~~  
751 ~~biomass burning. On the more general cases of particulate pollution, our~~ Our study  
752 also suggests that fire aerosols are responsible for a substantial fraction of the low visibility  
753 days (visibility < 10 km) in ~~several these~~ cities: ~~38~~ up to 39% in Bangkok, ~~35~~ 36% in Kuala  
754 Lumpur, 34% in Singapore, and ~~32~~ 33% in Kuching.

755 ~~The life cycle and transport path, and thus spatial and temporal distributions of~~  
756 ~~fire aerosols are all influenced by meteorological conditions, especially the seasonal~~  
757 ~~precipitation distribution and atmospheric circulations. These impacts are well~~

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758 ~~reflected from the variations of abundance of fire aerosols in the selected cities in~~  
759 ~~analysis. In general, Mainland~~In attributing the low visibility events to fire emissions  
760 ~~from different sites, we find that mainland~~ Southeast Asia is the major contributor during  
761 the Northeast or winter monsoon season in Southeast Asia. In the Southwest or summer  
762 monsoon season, ~~however,~~ most fire aerosols come from Sumatra and Borneo.  
763 Specifically, fires in ~~Mainland~~mainland Southeast Asia are accounted for the largest  
764 percentage of the total fire PM<sub>2.5</sub> in Bangkok (99.2%), and fires from Sumatra are the major  
765 contributor in Kuala Lumpur (5150%) and Singapore (4241%). Kuching receives 8885%  
766 of fire aerosols from local Borneo fires.

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767 By comparing the results from two modeled runs with the same fire emissions but  
768 driven by different meteorological inputs, we have examined the ~~potential,~~ sensitivity of  
769 modeled results to meteorological datasets. The discrepancy in ~~modeling the modeled~~ low  
770 visibility events ~~due to arising from the use of~~ different meteorological datasets is clearly  
771 evident, especially in the results of Bangkok and Kuching. However, using different  
772 meteorological input datasets does not appear to have influenced the modeled very low  
773 visibility events, or the severe haze events in the cities close to burning sites.

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774 We have also examined the sensitivity of modeled results to the use of different  
775 emission inventories. We find that significant discrepancies of fire emissions in  
776 ~~Mainland~~mainland Southeast Asia and northern Australia between ~~the~~ two emission  
777 inventories used in ~~the our~~ study have caused ~~significant a~~ substantial difference in  
778 modeled fire aerosol concentration and visibility, ~~particularly~~especially in Bangkok and  
779 Singapore. For instance, the contribution to fire aerosol in Singapore from northern  
780 Australia changes from nearly zero in the simulation driven by FINNv1.5 to about 22% in

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781 another simulation driven by GFEDv4.1s. We have also identified the influence of the  
782 ~~discrepancy~~difference in spatiotemporal distribution rather than total emitted quantities  
783 from the fire hotspots on modeled PM<sub>2.5</sub> concentration. ~~Further analysis on this direction~~  
784 ~~is much needed.~~

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785 To further assess the impacts of ~~fire events~~particulate pollution on the ~~air~~  
786 ~~quality~~surface visibility of the ~~great whole~~Southeast Asia ~~and to estimate the fire aerosol's~~  
787 ~~contribution.~~ we have defined and derived a metric of “Haze Exposure Days” (HEDs), by  
788 integrating annual low visibility days of 50 cities of the Association of Southeast Asian  
789 Nations and weighted by population or averaged arithmetically. We find that a very large  
790 population of Southeast Asia has been exposed to relatively persistent hazy  
791 ~~condition~~conditions. The top four cities in the HED ranking, Jakarta, Bangkok, Hanoi,

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792 and Yangon, with a total population exceeding two millions, ~~all~~ have experienced more  
793 than 200 days per year of low visibility due to particulate pollution over the past decade.

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794 Even worse is that the number of annual low visibility days have been increasing steadily  
795 not only in high population cities but also those with relatively low populations, suggesting  
796 a ~~widely wide~~ spread of particulate pollutions ~~into the great across~~ Southeast Asian  
797 ~~region.~~ Generally speaking, the fire aerosols are found to be responsible for ~~up to~~ about

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798 half of the total exposes to low visibility ~~aerossin~~ the region. Our result suggests that in  
799 order to improve the air quality in Southeast Asia, besides reducing or even prohibiting  
800 planned or unplanned fires, mitigation policies targeting at pollution sources other than  
801 fires need to be ~~put in effect~~implemented as well.

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802  
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816

## 817 Reference

- 818 Carlson, K. M., Curran, L. M., Ratnasari, D., Pittman, A. M., Soares-Filho, B. S., Asner,  
819 G. P., Trigg, S. N., Gaveau, D. A., Lawrence, D., and Rodrigues, H. O.:  
820 Committed carbon emissions, deforestation, and community land conversion from  
821 oil palm plantation expansion in West Kalimantan, Indonesia, Proceedings of the  
822 National Academy of Sciences, 109, 7559-7564, 10.1073/pnas.1200452109,  
823 2012.
- 824 Chang, C. P., Wang, Z., McBride, J., and Liu, C.-H.: Annual Cycle of Southeast Asia—  
825 Maritime Continent Rainfall and the Asymmetric Monsoon Transition, *Journal of*  
826 *Climate*, 18, 287-301, 10.1175/JCLI-3257.1, 2005.
- 827 Dennis, R., Mayer, J., Applegate, G., Chokkalingam, U., Colfer, C. P., Kurniawan, I.,  
828 Lachowski, H., Maus, P., Permana, R., Ruchiat, Y., Stolle, F., Suyanto, and  
829 Tomich, T.: Fire, People and Pixels: Linking Social Science and Remote Sensing  
830 to Understand Underlying Causes and Impacts of Fires in Indonesia, *Hum Ecol*,  
831 33, 465-504, 10.1007/s10745-005-5156-z, 2005.
- 832 Emmanuel, S. C.: Impact to lung health of haze from forest fires: The Singapore  
833 experience, *Respirology*, 5, 175-182, 10.1046/j.1440-1843.2000.00247.x, 2000.

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Line spacing: single

- 834 Field, R. D., van der Werf, G. R., and Shen, S. S. P.: Human amplification of drought-  
 835 induced biomass burning in Indonesia since 1960, *Nature Geosci*, 2, 185-188,  
 836 [http://www.nature.com/ngeo/journal/v2/n3/supinfo/ngeo443\\_S1.html](http://www.nature.com/ngeo/journal/v2/n3/supinfo/ngeo443_S1.html), 2009.
- 837 Fujii, Y., Iriana, W., Oda, M., Puriwigati, A., Tohno, S., Lestari, P., Mizohata, A., and  
 838 Huboyo, H. S.: Characteristics of carbonaceous aerosols emitted from peatland  
 839 fire in Riau, Sumatra, Indonesia, *Atmospheric Environment*, 87, 164-169,  
 840 <http://dx.doi.org/10.1016/j.atmosenv.2014.01.037>, 2014.
- 841 Giglio, L., Randerson, J. T., and van der Werf, G. R.: Analysis of daily, monthly, and  
 842 annual burned area using the fourth-generation global fire emissions database  
 843 (GFED4), *Journal of Geophysical Research: Biogeosciences*, 118, 317-328,  
 844 10.1002/jgrg.20042, 2013.
- 845 Grell, G., Freitas, S. R., Stuefer, M., and Fast, J.: Inclusion of biomass burning in WRF-  
 846 Chem: impact of wildfires on weather forecasts, *Atmos. Chem. Phys.*, 11, 5289-  
 847 5303, 10.5194/acp-11-5289-2011, 2011.
- 848 Grell, G. A., ~~A. Mangold, M. K., O. Mohler, H. Saathoff, H. Teichert, V. Ebert,~~ Peckham,  
 849 S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.:  
 850 Fully coupled “online” chemistry within the WRF model, *Atmospheric*  
 851 *Environment*, 39, ~~6957-6975~~ [10.1016/j.atmosenv.2005.04.027](https://doi.org/10.1016/j.atmosenv.2005.04.027), 2005.
- 852 Heil, A., Langmann, B., and Aldrian, E.: Indonesian peat and vegetation fire emissions:  
 853 Study on factors influencing large-scale smoke haze pollution using a regional  
 854 atmospheric chemistry model, *Mitig Adapt Strat Glob Change*, 12, 113-133,  
 855 10.1007/s11027-006-9045-6, 2007.
- 856 Huffman, G. J., Bolvin, D. T., Nelkin, E. J., Wolff, D. B., Adler, R. F., Gu, G., Hong, Y.,  
 857 Bowman, K. P., and Stocker, E. F.: The TRMM Multisatellite Precipitation  
 858 Analysis (TMPA): Quasi-Global, Multiyear, Combined-Sensor Precipitation  
 859 Estimates at Fine Scales, *Journal of Hydrometeorology*, 8, 38-55,  
 860 10.1175/JHM560.1, 2007.
- 861 Johnston, F. H., Henderson, S. B., Chen, Y., Randerson, J. T., Marlier, M., Defries, R. S.,  
 862 Kinney, P., Bowman, D. M., and Brauer, M.: Estimated global mortality  
 863 attributable to smoke from landscape fires *Environ. Health Perspect.* , 120 695–  
 864 701, 2012.
- 865 ~~Kiehl, J. T., Schneider, T. L., Rasch, P. J., Barth, M. C., and Wong, J.: Radiative forcing~~  
 866 ~~due to sulfate aerosols from simulations with the National Center for Atmospheric~~  
 867 ~~Research Community Climate Model, Version 3, *Journal of Geophysical*~~  
 868 ~~*Research: Atmospheres*, 105, 1441-1457, 10.1029/1999JD900495, 2000.~~
- 869 Kim, D., Wang, C., Ekman, A. M. L., Barth, M. C., and Rasch, P. J.: Distribution and  
 870 direct radiative forcing of carbonaceous and sulfate aerosols in an interactive size-  
 871 resolving aerosol–climate model, *Journal of Geophysical Research: Atmospheres*,  
 872 113, D16309, 10.1029/2007jd009756, 2008.
- 873 Koe, L. C. C., Arellano Jr, A. F., and McGregor, J. L.: Investigating the haze transport  
 874 from 1997 biomass burning in Southeast Asia: its impact upon Singapore,  
 875 *Atmospheric Environment*, 35, 2723-2734,  
 876 ~~2001~~ [http://dx.doi.org/10.1016/S1352-2310\(00\)00395-2](http://dx.doi.org/10.1016/S1352-2310(00)00395-2), 2001.
- 877 Kunii, O., Kanagawa, S., Yajima, I., Hisamatsu, Y., Yamamura, S., Amagai, T., and  
 878 Ismail, I. T. S.: The 1997 Haze Disaster in Indonesia: Its Air Quality and Health

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879 Effects, Archives of Environmental Health: An International Journal, 57, 16-22,  
880 10.1080/00039890209602912, 2002.

881 Langner, A., Miettinen, J., and Siegert, F.: Land cover change 2002–2005 in Borneo and  
882 the role of fire derived from MODIS imagery, Global Change Biology, 13, 2329-  
883 2340, 10.1111/j.1365-2486.2007.01442.x, 2007.

884 Lin, N.-H., Tsay, S.-C., Maring, H. B., Yen, M.-C., Sheu, G.-R., Wang, S.-H., Chi, K. H.,  
885 Chuang, M.-T., Ou-Yang, C.-F., Fu, J. S., Reid, J. S., Lee, C.-T., Wang, L.-C.,  
886 Wang, J.-L., Hsu, C. N., Sayer, A. M., Holben, B. N., Chu, Y.-C., Nguyen, X. A.,  
887 Sopajaree, K., Chen, S.-J., Cheng, M.-T., Tsuang, B.-J., Tsai, C.-J., Peng, C.-M.,  
888 Schnell, R. C., Conway, T., Chang, C.-T., Lin, K.-S., Tsai, Y. I., Lee, W.-J.,  
889 Chang, S.-C., Liu, J.-J., Chiang, W.-L., Huang, S.-J., Lin, T.-H., and Liu, G.-R.:  
890 An overview of regional experiments on biomass burning aerosols and related  
891 pollutants in Southeast Asia: From BASE-ASIA and the Dongsha Experiment to  
892 7-SEAS, Atmospheric Environment, 78, 1-19,  
893 ~~2013~~<http://dx.doi.org/10.1016/j.atmosenv.2013.04.066>, 2013.

894 Madden, R. A., and Julian, P. R.: Detection of a 40–50 Day Oscillation in the Zonal  
895 Wind in the Tropical Pacific, Journal of the Atmospheric Sciences, 28, 702-708,  
896 10.1175/1520-0469(1971)028<0702:DOADOI>2.0.CO;2, 1971.

897 ~~Marlier, M., Defries, R. S., Kim, P. S., Koplitz, S. N., Jacob, D. J., Mickley, L. J., and~~  
898 ~~Myers, S. S., Mauderly, J. L., and Chow, J. C.: Health effects of organic aerosols,~~  
899 ~~Inhalation Toxicology, 20, 257-288, 2008.~~

900 ~~Miriam, E. M., Ruth, S. D., Patrick, S. K., David, L. A. G., Shannon, N. K., Daniel, J. J.,~~  
901 ~~Loretta, J. M., Belinda, A. M., and Samuel, S. M.: Regional air quality impacts of~~  
902 ~~future fire emissions in Sumatra and Kalimantan, Environmental Research~~  
903 ~~Letters, 10, 054010, 2015a.~~

904 ~~Miriam, E. M., Ruth, S. D., Patrick, S. K., Shannon, N. K., Daniel, J. J., Loretta, J. M., and~~  
905 ~~Samuel, S. M.: Fire emissions and regional air quality impacts from fires in oil~~  
906 ~~palm, timber, and logging concessions in Indonesia, Environmental Research~~  
907 ~~Letters, 10, 085005, 2015b~~2015a.

908 ~~Marlier, M. E., DeFries, R. S., Kim, P. S., Gaveau, D. L. A., Koplitz, S. N., Jacob, D. J.,~~  
909 ~~Mickley, L. J., Margono, B. A., and Myers, S. S.: Regional air quality impacts of~~  
910 ~~future fire emissions in Sumatra and Kalimantan, Environmental Research~~  
911 ~~Letters, 5, 054010 pp., 2015b.~~

912 ~~Mauderly, J. L., and Chow, J. C.: Health effects of organic aerosols, Inhalation~~  
913 ~~Toxicology, 20, 257-288, 2008.~~

914 ~~McBride, J. L., Haylock, M. R., and Nicholls, N.: Relationships between the Maritime~~  
915 ~~Continent Heat Source and the El Niño–Southern Oscillation Phenomenon,~~  
916 ~~Journal of Climate, 16, 2905-2914, 10.1175/1520-~~  
917 ~~0442(2003)016<2905:RBTMCH>2.0.CO;2, 2003.~~

918 Page, S. E., Siegert, F., Rieley, J. O., Boehm, H.-D. V., Jaya, A., and Limin, S.: The  
919 amount of carbon released from peat and forest fires in Indonesia during 1997,  
920 Nature, 420, 61-65, 2002.

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921 Randerson, J. T., Chen, Y., van der Werf, G. R., Rogers, B. M., and Morton, D. C.:  
922 Global burned area and biomass burning emissions from small fires, *Journal of*  
923 *Geophysical Research: Biogeosciences*, 117, G04012, 10.1029/2012JG002128,  
924 2012.

925 Rasmusson, E. M., and Wallace, J. M.: Meteorological Aspects of the El Niño/Southern  
926 Oscillation, *Science*, 222, 1195-1202, 10.1126/science.222.4629.1195, 1983.

927 Reddington, C. L., Yoshioka, M., Balasubramanian, R., Ridley, D., Toh, Y. Y., Arnold,  
928 S. R., and Spracklen, D. V.: Contribution of vegetation and peat fires to  
929 particulate air pollution in Southeast Asia, *Environmental Research Letters*, 9,  
930 094006, 2014.

931 Reid, J. S., Xian, P., Hyer, E. J., Flatau, M. K., Ramirez, E. M., Turk, F. J., Sampson, C.  
932 R., Zhang, C., Fukada, E. M., and Maloney, E. D.: Multi-scale meteorological  
933 conceptual analysis of observed active fire hotspot activity and smoke optical  
934 depth in the Maritime Continent, *Atmos. Chem. Phys.*, 12, 2117-2147,  
935 10.5194/acp-12-2117-2012, 2012.

936 Reid, J. S., Lagrosas, N. D., Jonsson, H. H., Reid, E. A., Sessions, W. R., Simpas, J. B.,  
937 Uy, S. N., Boyd, T. J., Atwood, S. A., Blake, D. R., Campbell, J. R., Cliff, S. S.,  
938 Holben, B. N., Holz, R. E., Hyer, E. J., Lynch, P., Meinardi, S., Posselt, D. J.,  
939 Richardson, K. A., Salinas, S. V., Smirnov, A., Wang, Q., Yu, L., and Zhang, J.:  
940 Observations of the temporal variability in aerosol properties and their  
941 relationships to meteorology in the summer monsoonal South China Sea/East Sea:  
942 the scale-dependent role of monsoonal flows, the Madden-Julian Oscillation,  
943 tropical cyclones, squall lines and cold pools, *Atmos. Chem. Phys.*, 15, 1745-  
944 1768, 10.5194/acp-15-1745-2015, 2015.

945 Saji, N. H., Goswami, B. N., Vinayachandran, P. N., and Yamagata, T.: A dipole mode in  
946 the tropical Indian Ocean, *Nature*, 401, 360-363, 1999.

947 See, S. W., Balasubramanian, R., and Wang, W.: A study of the physical, chemical, and  
948 optical properties of ambient aerosol particles in Southeast Asia during hazy and  
949 nonhazy days, *Journal of Geophysical Research: Atmospheres*, 111, D10S08,  
950 10.1029/2005JD006180, 2006.

951 Seinfeld, J., and Pandis, S.: *Atmospheric Physics and Chemistry. From Air Pollution to*  
952 *Climate Change*, Second Edition ed., New York (NY): JohnWiley & Sons, 2006.

953 Sekiguchi, M., Nakajima, T., Suzuki, K., Kawamoto, K., Higurashi, A., Rosenfeld, D.,  
954 Sano, I., and Mukai, S.: A study of the direct and indirect effects of aerosols using  
955 global satellite data sets of aerosol and cloud parameters, *Journal of Geophysical*  
956 *Research: Atmospheres*, 108, 4699, 10.1029/2002JD003359, 2003.

957 ~~Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang,~~  
958 ~~X.-Y., Wang, W., and Powers, J. G.: A Description of the Advanced Research WRF~~  
959 ~~Version 3, NCAR Technical Note, NCAR/TN-475+STR, 2008.~~

960 Smith, A., Lott, N., and Vose, R.: The Integrated Surface Database: Recent  
961 Developments and Partnerships, *Bulletin of the American Meteorological Society*,  
962 92, 704-708, doi:10.1175/2011BAMS3015.1, 2011.

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Line spacing: single



963 Stauffer, D. R., and Seaman, N. L.: Multiscale Four-Dimensional Data Assimilation,  
964 Journal of Applied Meteorology, 33, 416-434, 10.1175/1520-  
965 0450(1994)033<0416:mfdda>2.0.co;2, 1994.

966 Tosca, M. G., Randerson, J. T., Zender, C. S., Flanner, M. G., and Rasch, P. J.: Do  
967 biomass burning aerosols intensify drought in equatorial Asia during El Niño?,  
968 Atmos. Chem. Phys., 10, 3515-3528, 10.5194/acp-10-3515-2010, 2010.

969 Tosca, M. G., Randerson, J. T., Zender, C. S., Nelson, D. L., Diner, D. J., and Logan, J. A.:  
970 Dynamics of fire plumes and smoke clouds associated with peat and deforestation  
971 fires in Indonesia, Journal of Geophysical Research: Atmospheres, 116, n/a-n/a,  
972 10.1029/2010JD015148, 2011.

973 van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P.  
974 S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire  
975 emissions and the contribution of deforestation, savanna, forest, agricultural, and  
976 peat fires (1997–2009), Atmos. Chem. Phys., 10, 11707-11735, 10.5194/acp-10-  
977 11707-2010, 2010.

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

980 Wang, J., Ge, C., Yang, Z., Hyer, E. J., Reid, J. S., Chew, B.-N., Mahmud, M., Zhang,  
981 Y., and Zhang, M.: Mesoscale modeling of smoke transport over the Southeast  
982 Asian Maritime Continent: Interplay of sea breeze, trade wind, typhoon, and  
983 topography, Atmospheric Research, 122, 486-503,  
984 2013-<http://dx.doi.org/10.1016/j.atmosres.2012.05.009>, 2013.

985 Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando,  
986 J. J., and Soja, A. J.: The Fire INventory from NCAR (FINN): a high resolution  
987 global model to estimate the emissions from open burning, Geosci. Model Dev.,  
988 4, 625-641, 10.5194/gmd-4-625-2011, 2011.

989 Wu, C.-H., and Hsu, H.-H.: Topographic Influence on the MJO in the Maritime  
990 Continent, Journal of Climate, 22, 5433-5448, 10.1175/2009JCLI2825.1, 2009.

991 Wu, R., Wen, Z., and He, Z.: ENSO Contribution to Aerosol Variations over the  
992 Maritime Continent and the Western North Pacific during 2000–10, Journal of  
993 Climate, 26, 6541-6560, 10.1175/JCLI-D-12-00253.1, 2013.

994 Zhang, C.: Madden-Julian Oscillation, Reviews of Geophysics, 43, RG2003,  
995 10.1029/2004RG000158, 2005.

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Table 1. WRF physics scheme configuration

Physics Processes	Scheme
microphysics	Morrison (2 moments) scheme
longwave radiation	rrtmg scheme
shortwave radiation	rrtmg scheme
surface-layer	MYNN surface layer
land surface	Unified Noah land-surface model
planetary boundary layer	MYNN 2.5 level TKE scheme
cumulus parameterization	Grell-Freitas ensemble scheme

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Table 2. The spatial and temporal correlation of monthly rainfall between model and observation during 2003-2014. FMA, MJJ, ASO, NDJ and All indicate February-April, May-July, August-October, November-January and whole year, respectively.

	<u>FNL FINN vs. TRMM</u>		<u>ERA FINN vs. TRMM</u>	
	<u>Spatial cor.</u>	<u>Temporal cor.</u>	<u>Spatial cor.</u>	<u>Temporal cor.</u>
<u>FMA</u>	<u>0.89</u>	<u>0.61</u>	<u>0.89</u>	<u>0.89</u>
<u>MJJ</u>	<u>0.83</u>	<u>0.69</u>	<u>0.81</u>	<u>0.90</u>
<u>ASO</u>	<u>0.86</u>	<u>0.59</u>	<u>0.84</u>	<u>0.89</u>
<u>NDJ</u>	<u>0.88</u>	<u>0.60</u>	<u>0.88</u>	<u>0.85</u>
<u>All</u>	<u>0.86</u>	<u>0.68</u>	<u>0.85</u>	<u>0.90</u>

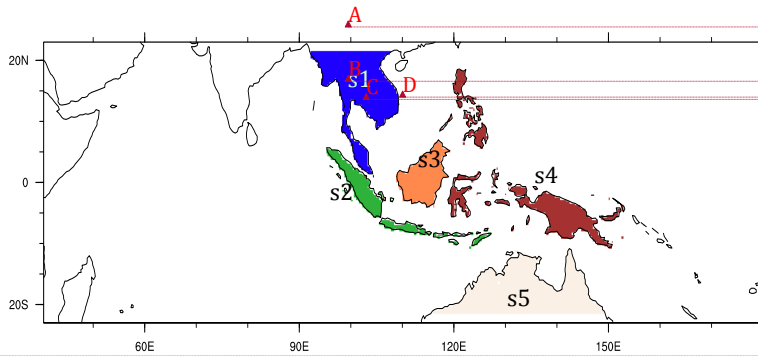
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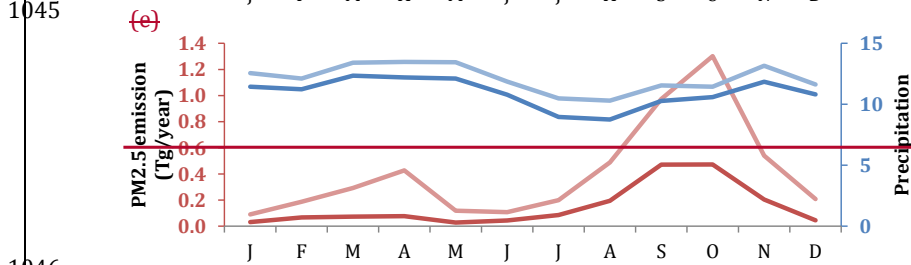
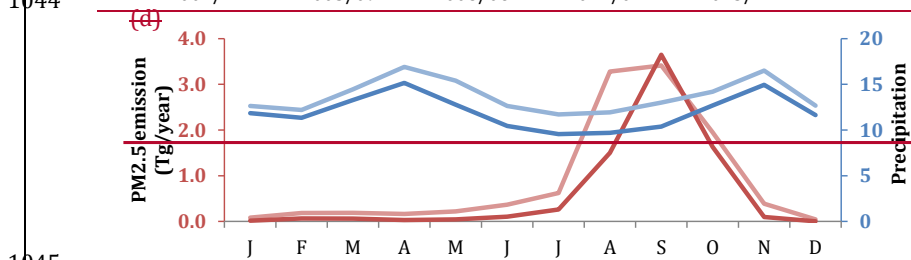
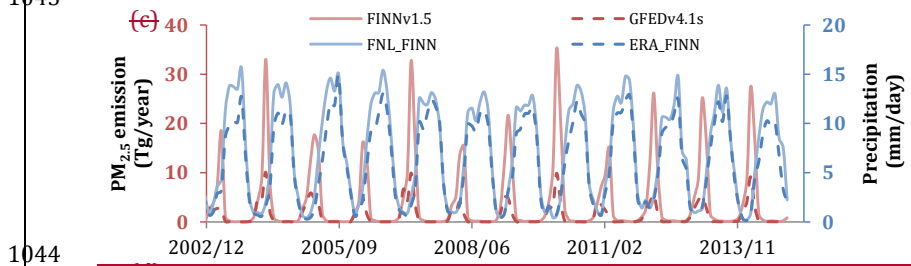
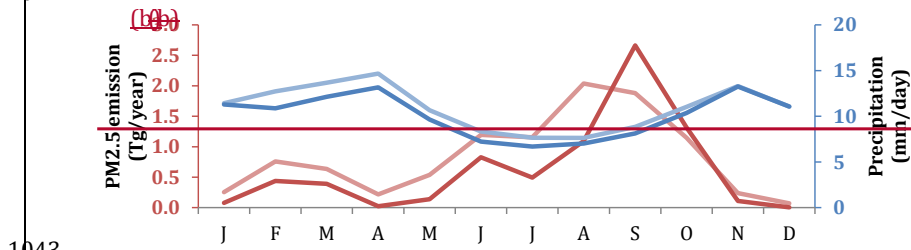
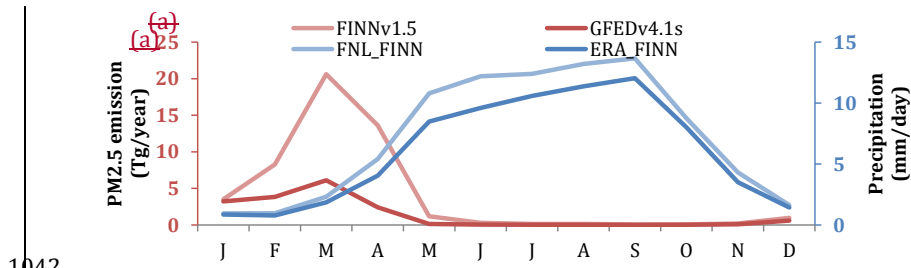


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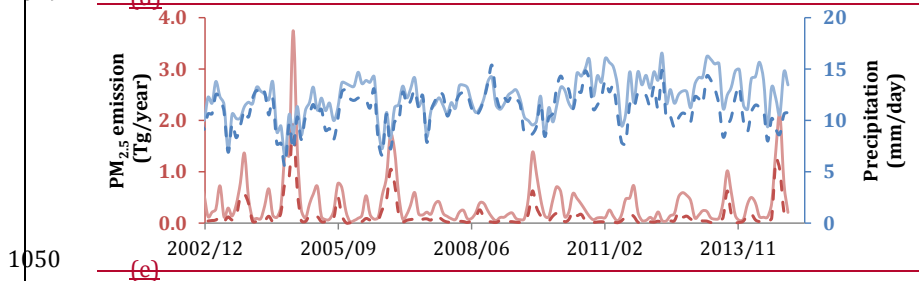
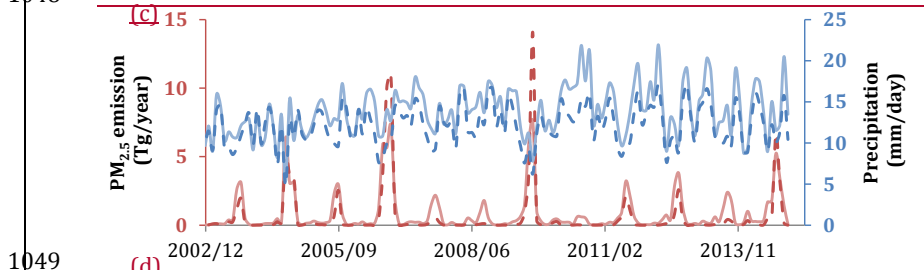
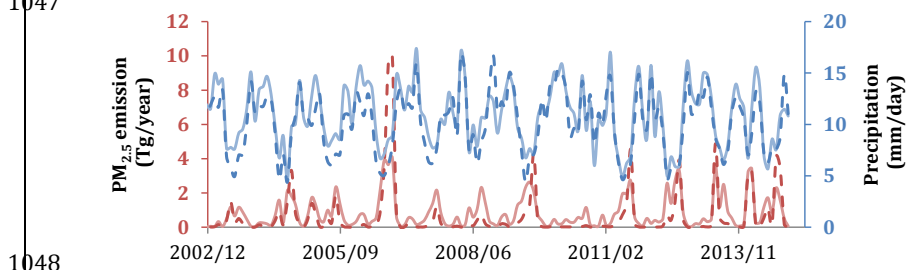
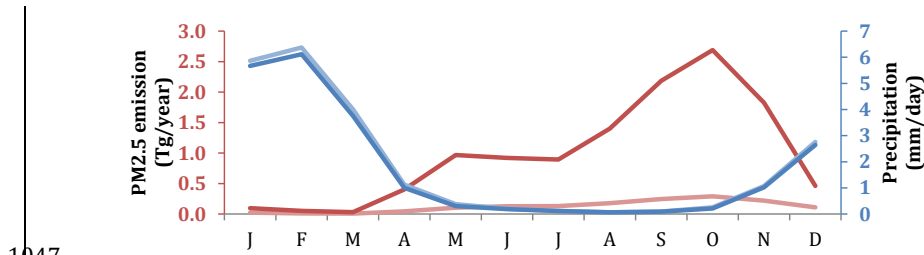
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Figure 1. Model domain used for simulations. Domain consists of 31 vertical levels, each with The domain has  $432 \times 148$  grid points with a horizontal resolution of 36 km. Five colored fire source regions, marked in different colors and labeled as s1, s2, s3, s4 and s5, represent Mainland, mainland Southeast Asia, (s1), Sumatra and Java islands, (s2), Borneo, (s3), the rest of Maritime Continent, (s4), and northern Australia, respectively. (s5). A, B, C and D indicate the location of four selected cities: Bangkok, (A), Kuala Lumpur, (B), Singapore (C) and Kuching, respectively. (D).

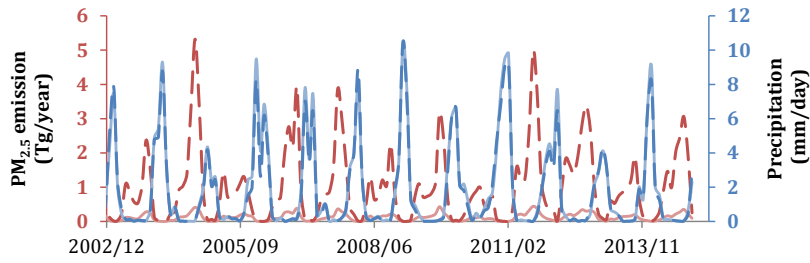
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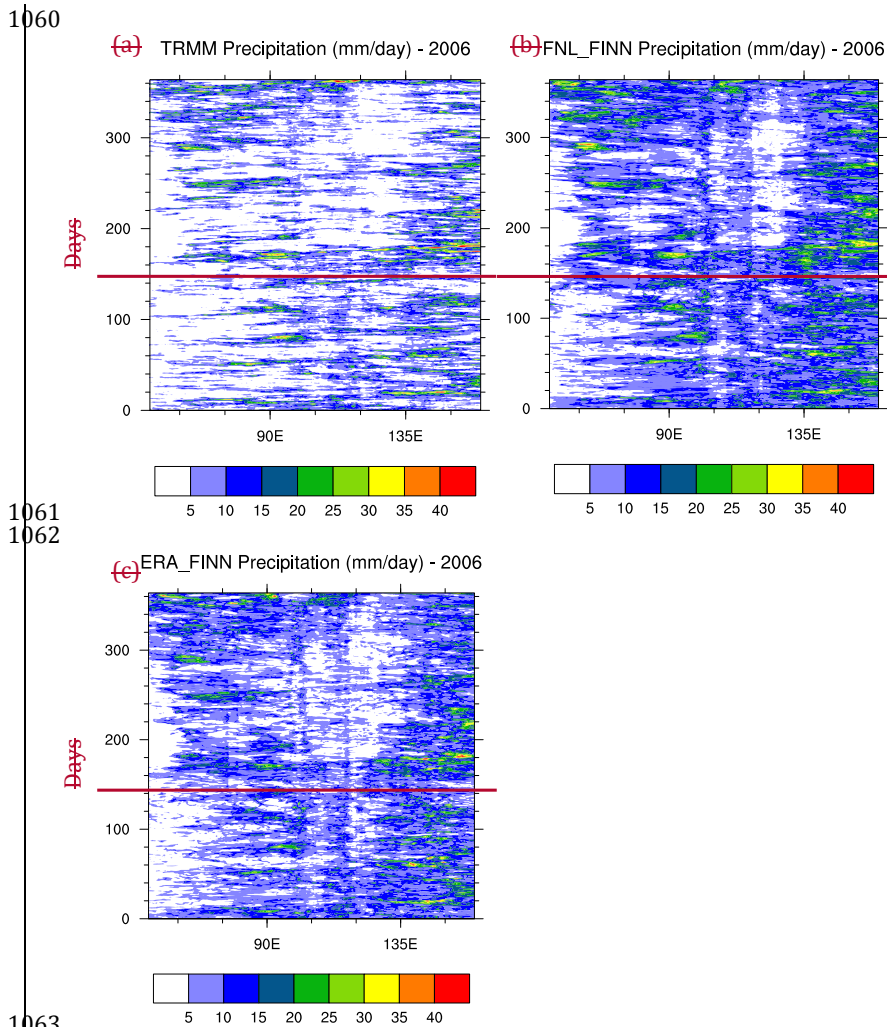




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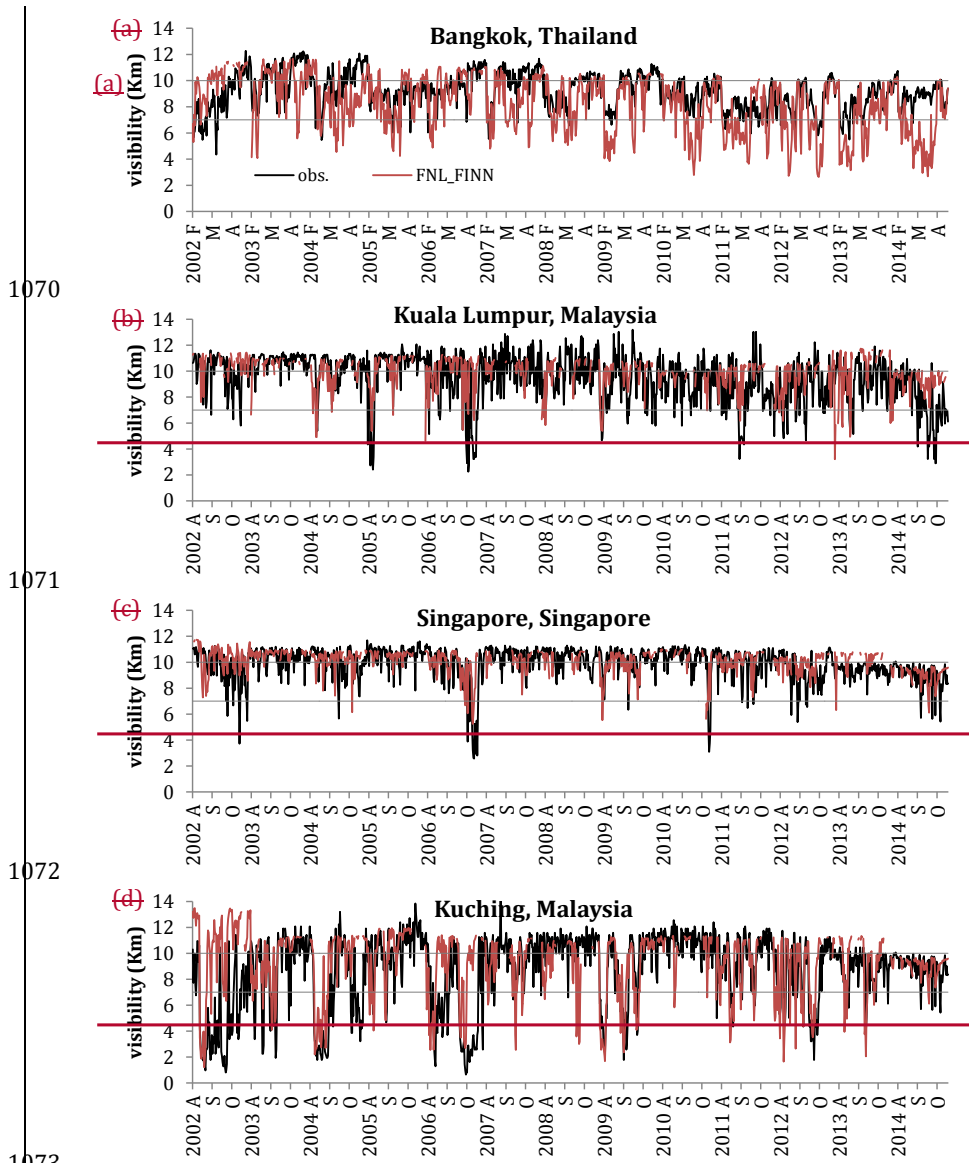
Figure 2. Monthly Time series of monthly  $PM_{2.5}$  emission (Tg year<sup>-1</sup>) in FINNv1.5 (red solid lines) and GFEDv4.1s (red dashed lines). Also shown are precipitation rates (mm day<sup>-1</sup>) simulated in FNL\_FINN (light blue solid lines) and ERA\_FINN (blue dashed lines). All data are averaged during 2002-2014 for: (a) Mainland Southeast Asia (s1), (b) Sumatra and Java islands (s2), (c) Borneo (s3), (d) the rest of the Maritime Continent (s4), and (e) northern Australia (s5). Note that GFEDv4.1s  $PM_{2.5}$  emission is averaged from 2003 to 2014.

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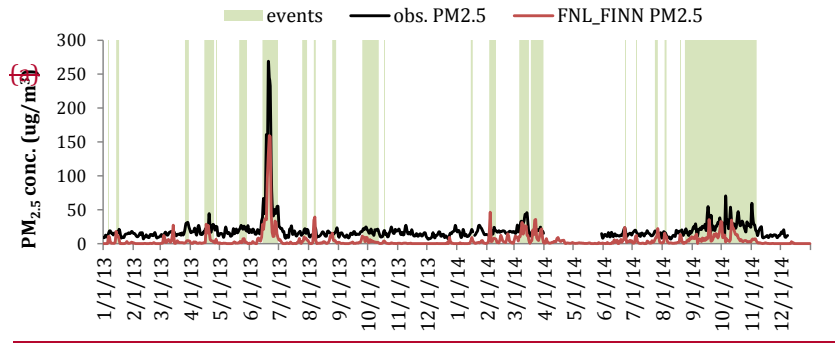
1065 Figure 3. Hovmöller (time vs. longitude) plot of daily precipitation in 2006 derived  
 1066 from: (a) TRMM, (b) FNL\_FINN, and (c) ERA\_FINN. Latitude average is from 10°S to  
 1067 10°N. Unit is mm day<sup>-1</sup>.

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 1074 **Figure 4. Comparison of daily visibility between GSOD observation (black lines) and**  
 1075 **FNL\_FINN modeled result (red lines) in: (a) Bangkok, (b) Kuala Lumpur, (c)**  
 1076 **Singapore, (d) Kuching during the fire seasons from 2002 to 2014.**

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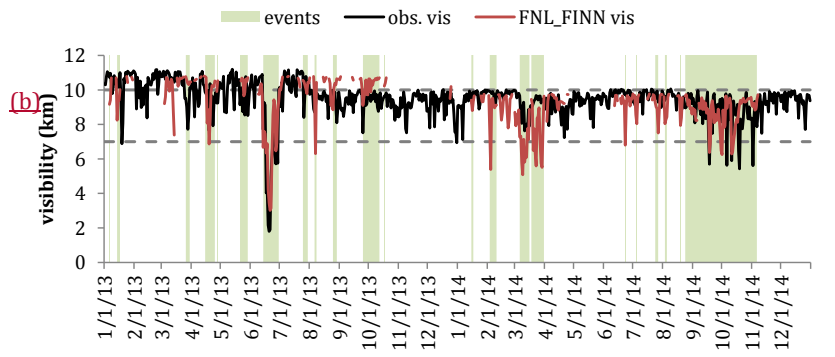
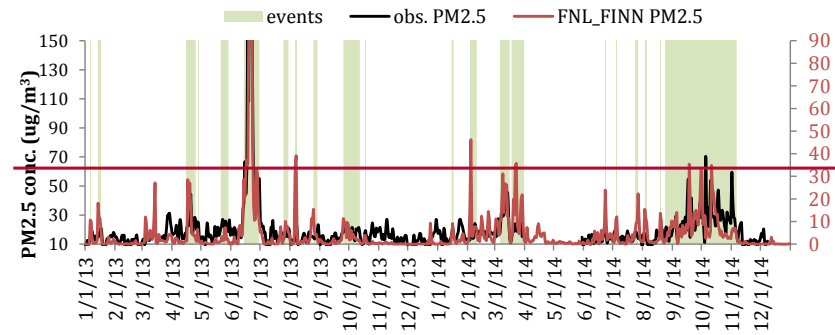
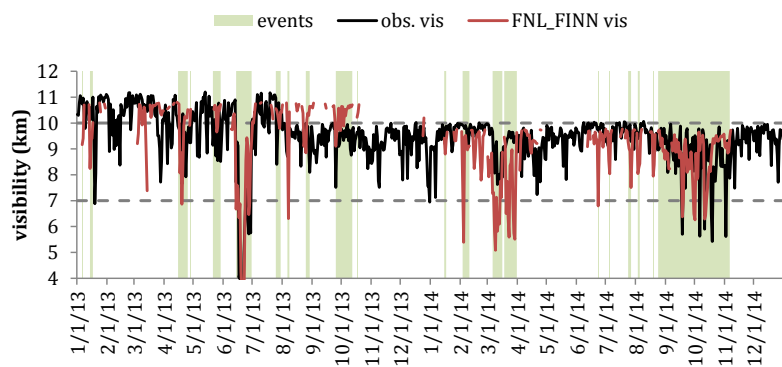


Figure 3. Two grey lines mark the visibility of 7 and 10 km, respectively.

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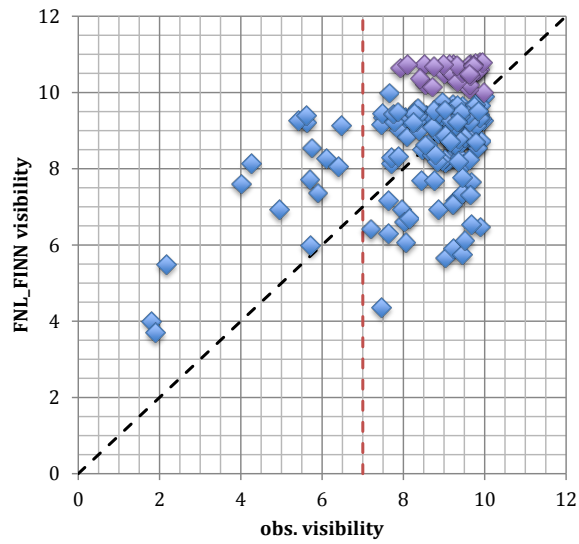




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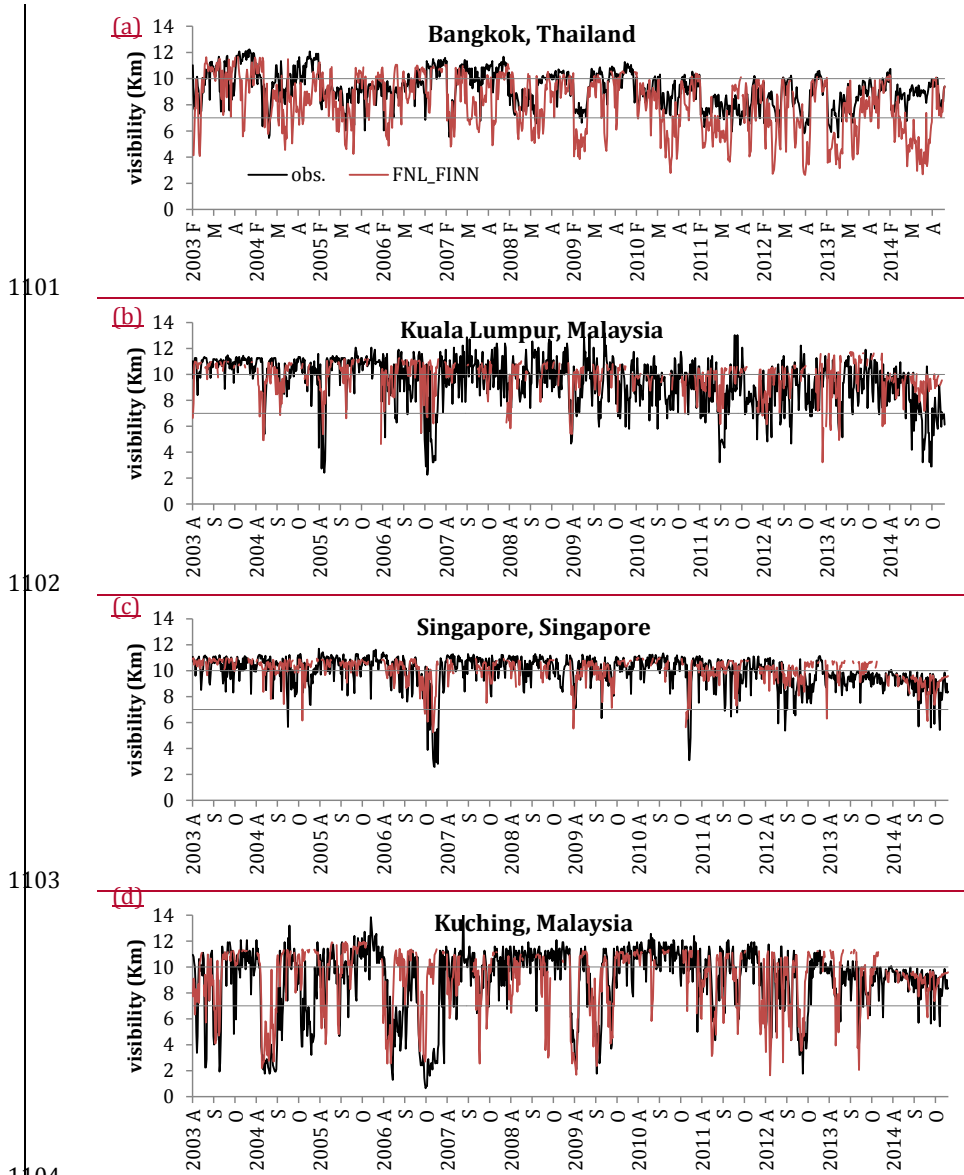
Figure 5. (a) Time series of daily surface PM<sub>2.5</sub> from the ground-based observations (black line) and FNL\_FINN simulated results (red line) in Singapore during 2013-2014. (b) Time series of daily visibility of GSOD observation (black line) and calculated result from FNL\_FINN (red line) in Singapore during 2013-2014. Highlighted green areas are known haze events caused by fire aerosols. Two gray lines mark the visibility of 7 and 10 km, respectively.

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Figure 4. A scatter plot of observed visibility and FNL FINN visibility during known fire events as labeled in Fig. 4b. Black dash line refers 1:1 line and red line is the threshold of VLVD (7 km). Data points marked with purple color are the events that model failed to produce a visibility qualified for LVD.



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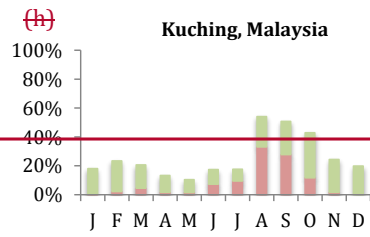
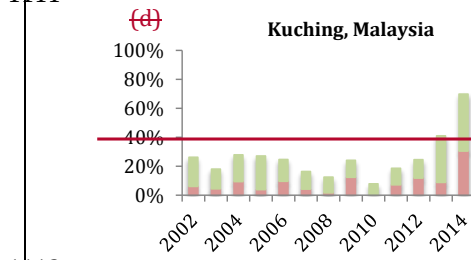
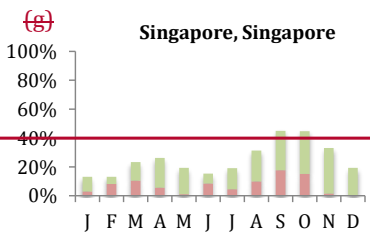
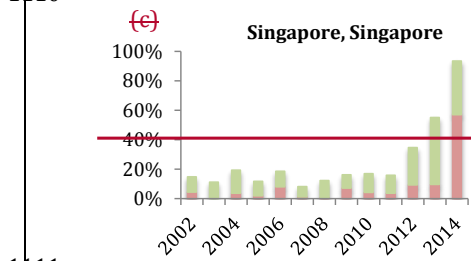
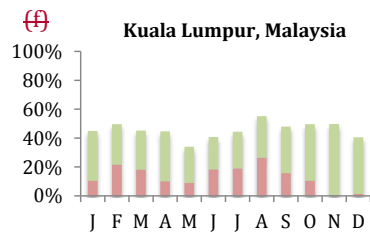
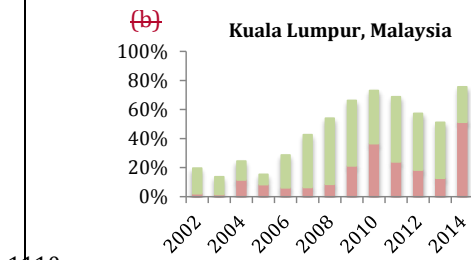
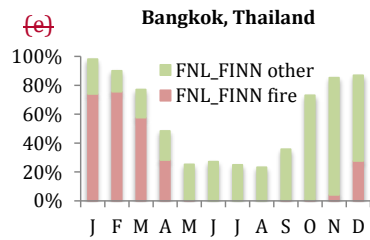
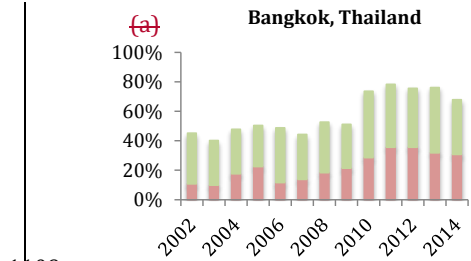
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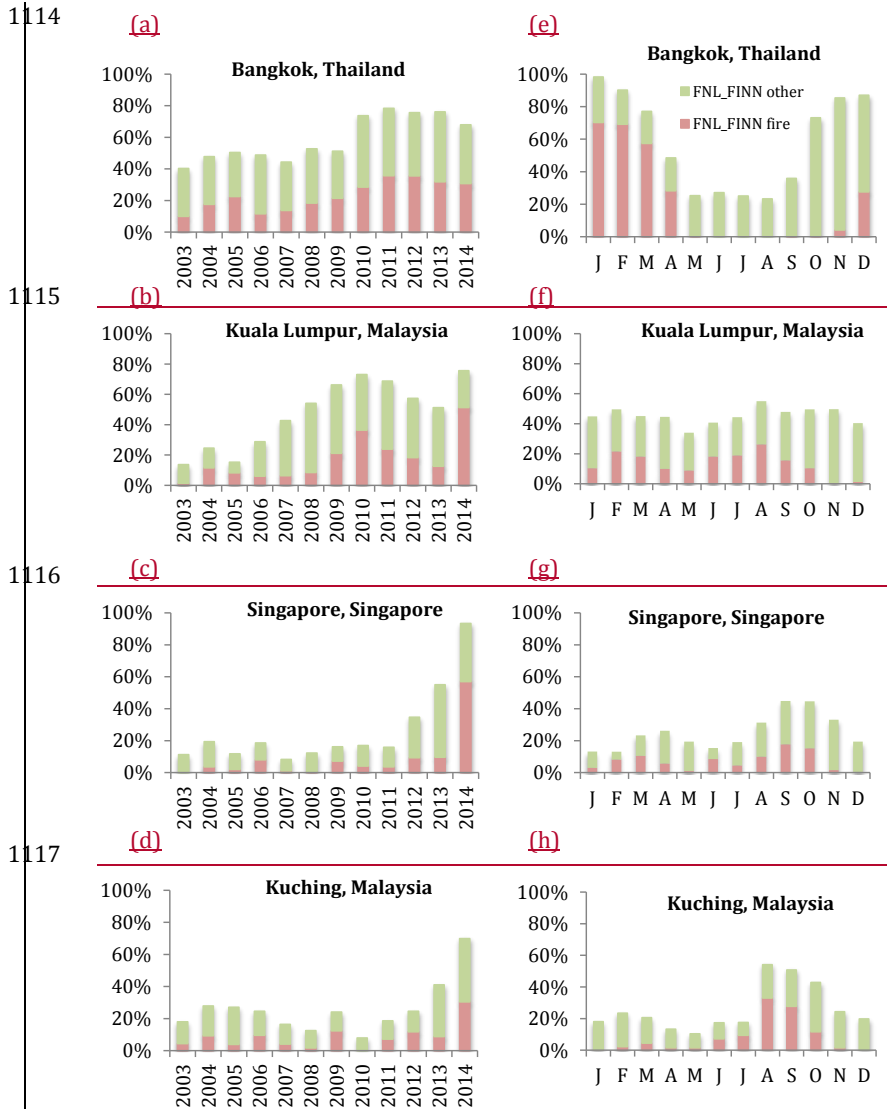
1105 Figure 5. Comparison of daily visibility between GSOD observation (black lines) and  
 1106 FNL FINN modeled result (red lines) in: (a) Bangkok, (b) Kuala Lumpur, (c) Singapore,  
 1107 (d) Kuching during the fire seasons from 2003 to 2014. Two grey lines mark the visibility  
 1108 of 7 and 10 km, respectively.

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 1119 Figure 6. (a) – (d) The annual variation of the percentage of LVDs per year from GSOD  
 1120 observational visibility in Bangkok, Kuala Lumpur, Singapore, and Kuching,  
 1121 respectively. (e) – (h) The monthly variation of the percentage of LVDs derived using  
 1122 from GSOD observational visibility observations in Bangkok, Kuala Lumpur, Singapore,  
 1123 and Kuching, respectively. (e) – (h) The percentage of LVDs averaged over 2002.

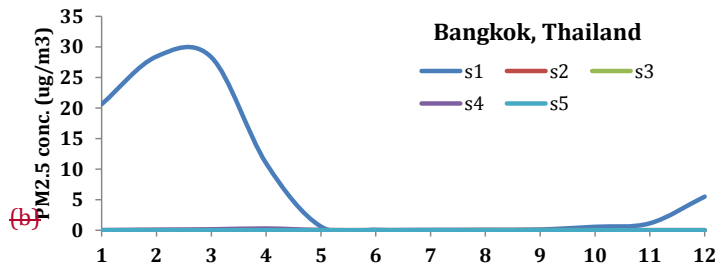
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1|24 2014,2003-2014, derived using GSOD visibility observations in Bangkok, Kuala Lumpur,  
1|25 Singapore, and Kuching, respectively. Each bar presents the observed LVDs in each year  
1|26 or month. Red color shows the partition of fire-caused LVDs (captured by model) while  
1|27 green color presents non-fire LVDs (observed – modeled).

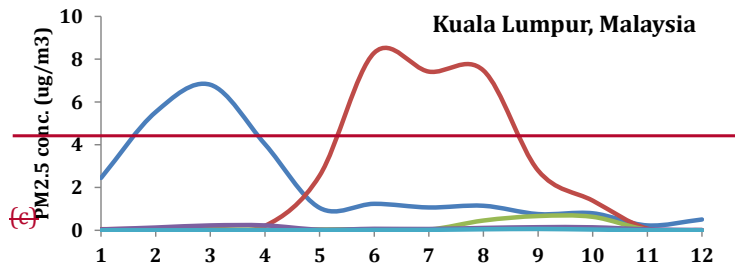
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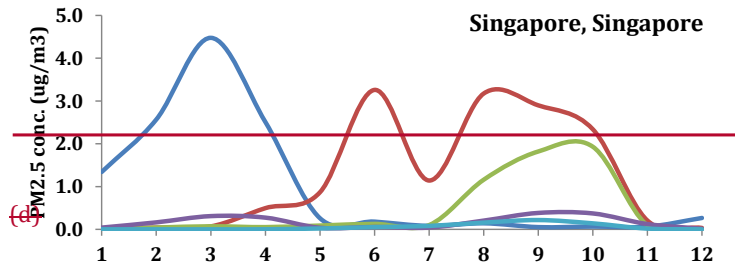
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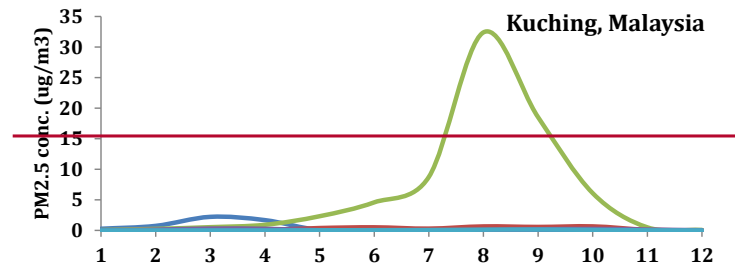
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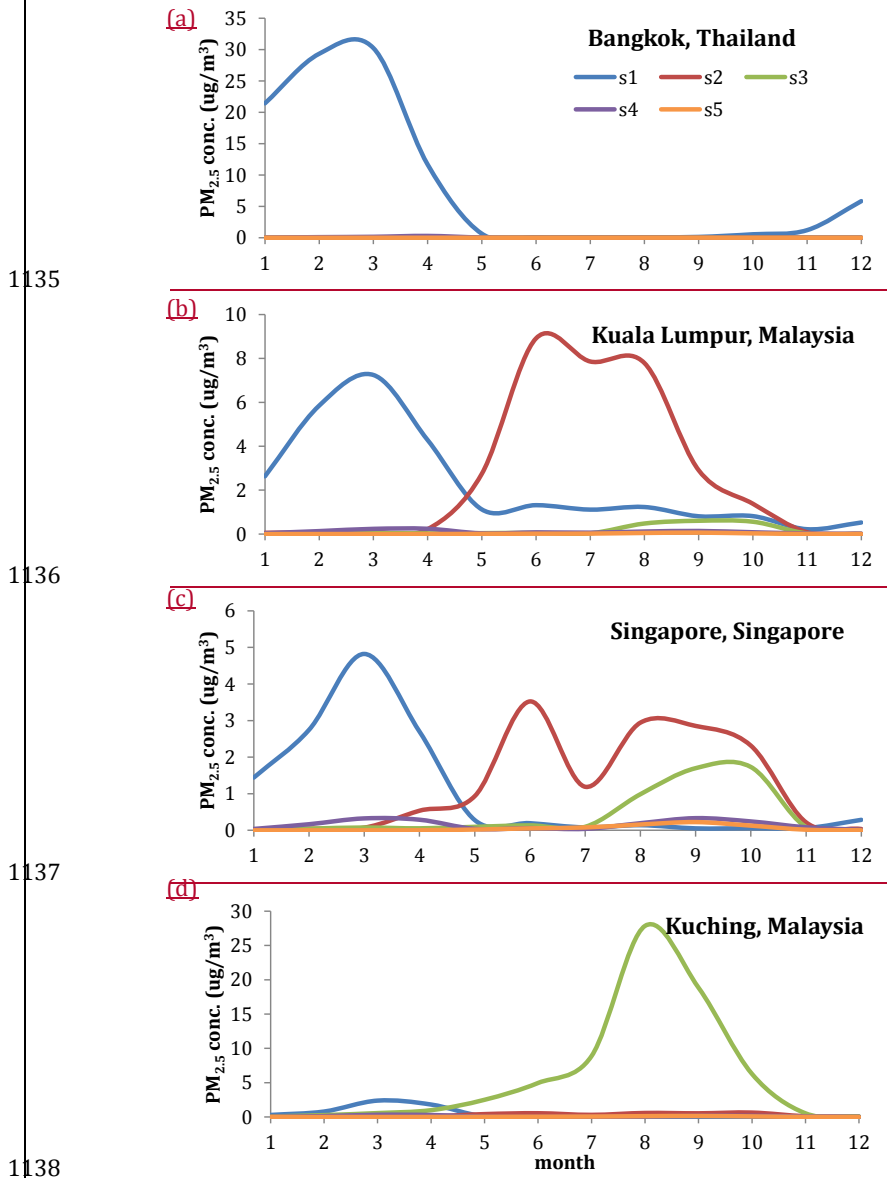
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Figure 7. The monthly variation of mean fire PM<sub>2.5</sub> concentration from each concentrations within the PBL attributed to different emission regions (s1 - s5) in (a)

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1142 Bangkok, (b) Kuala Lumpur, (c) Singapore and (d) Kuching, all derived from FNL\_FINN  
1143 simulation and averaged over the period ~~2002 of 2003~~-2014.

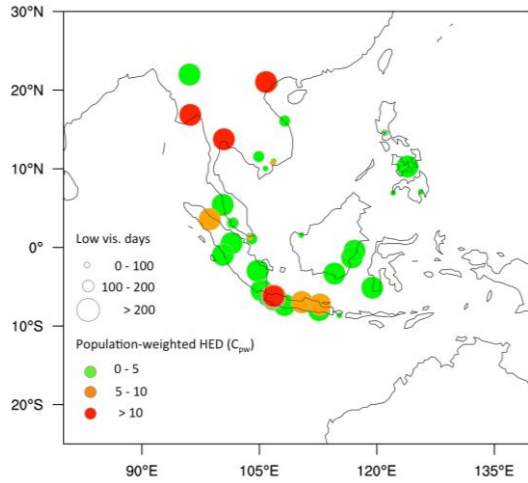
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(a)



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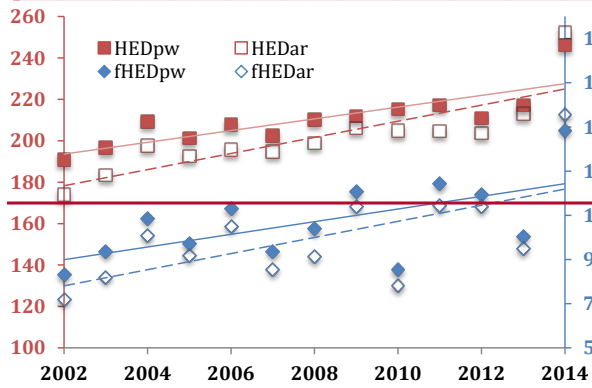
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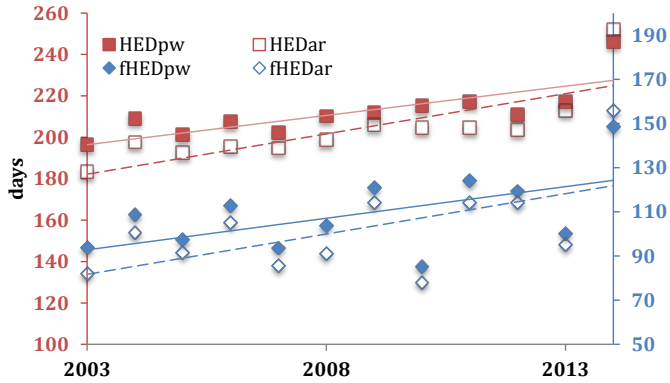
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(b)



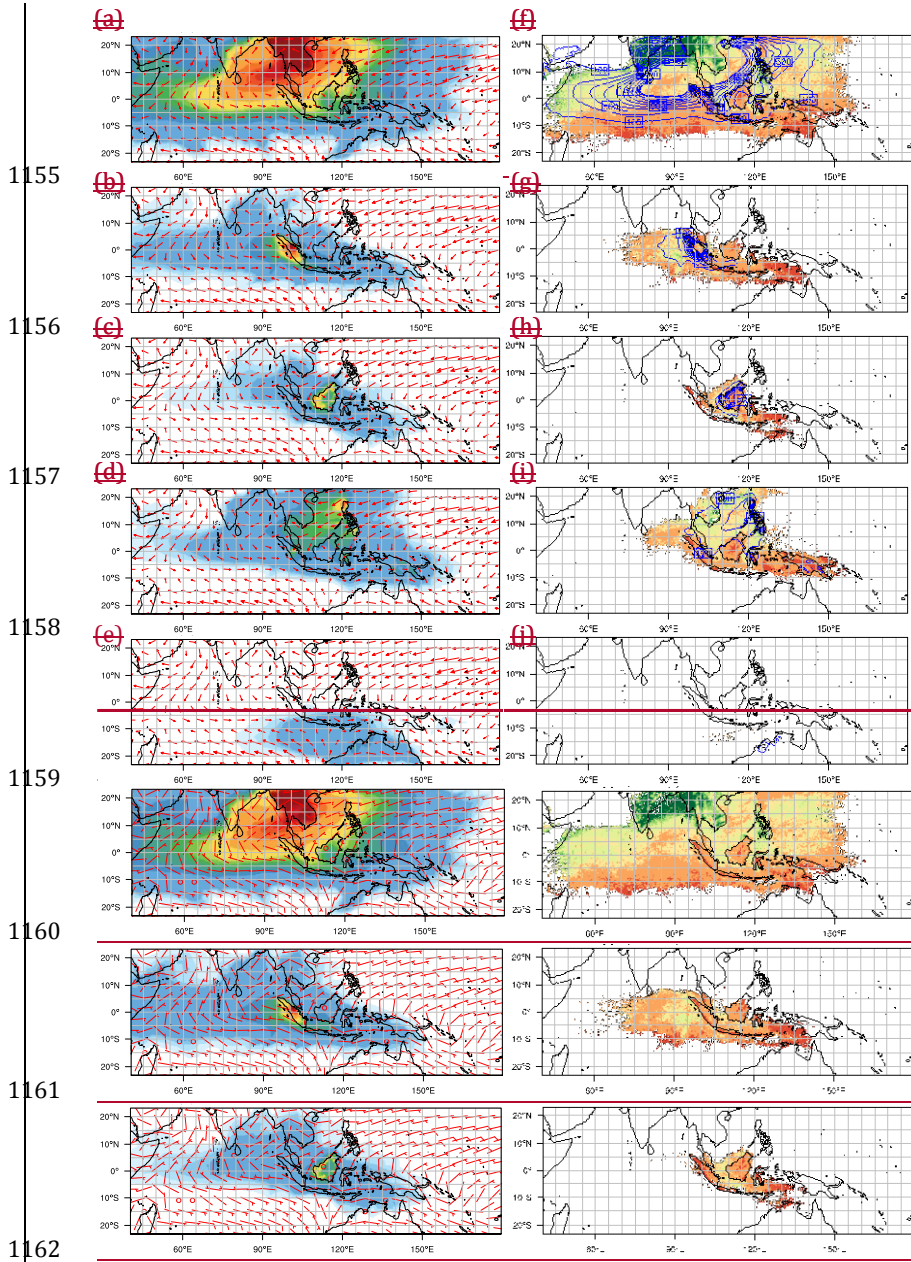
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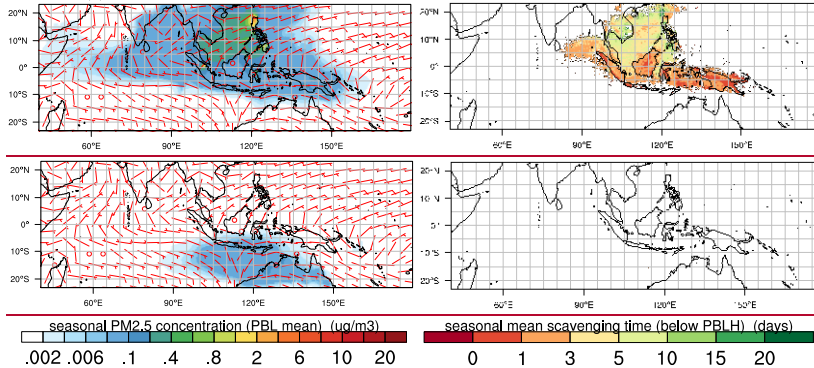
Figure 8. (a) The mean low visibility days (circles) per year from 2002 to 2014 in 50 ASEAN cities and their. The size of the circles indicates the number of days. The colors refer to population-weighted fraction in the total Haze Exposure Days (HED; colors). (b) Annual variation of population-weighted HED (HED<sub>pw</sub>) and arithmetic mean HED (HED<sub>ar</sub>). Fire-caused HED are labeled as fhED<sub>pw</sub> and fhED<sub>ar</sub>. Units are in days. Note that the y-axes are in different scales.

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Figure 9. (a)-(e) Seasonal mean fire  $PM_{2.5}$  concentration ( $\mu g m^{-3}$ ) and wind within the PBL modeled in FNL\_FINN during February to April, 2002-2003-2014 in: Mainland for fire  $PM_{2.5}$  source region from (a) mainland Southeast Asia (s1), (b) Sumatra and Java island (s2) islands, (c) Borneo (s3), (d) the rest of the Maritime Continent (s4), and (e) northern Australia (s5), respectively. (f)-(g) Same as (a)-(e) but for seasonal mean wet scavenging time (days; shaded) and column intergraded  $PM_{2.5}$  concentration ( $\mu g m^{-2}$ ; contours) within the PBL height. (e)

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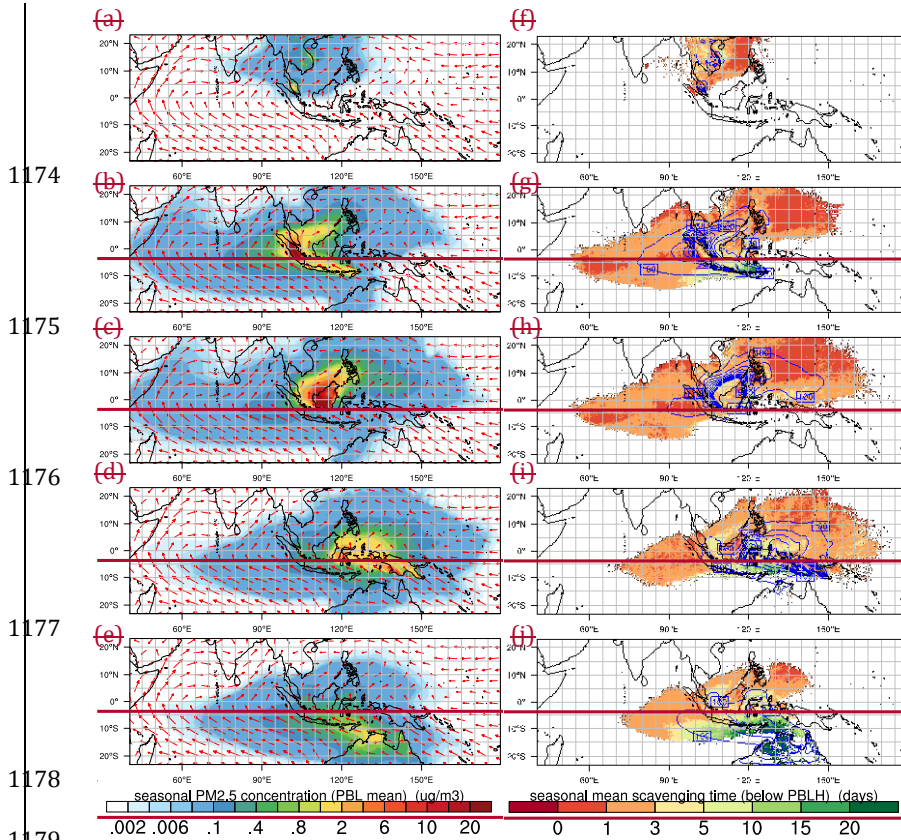


Figure 10. Same as Fig. 9 but during August to October, 2002–2014.

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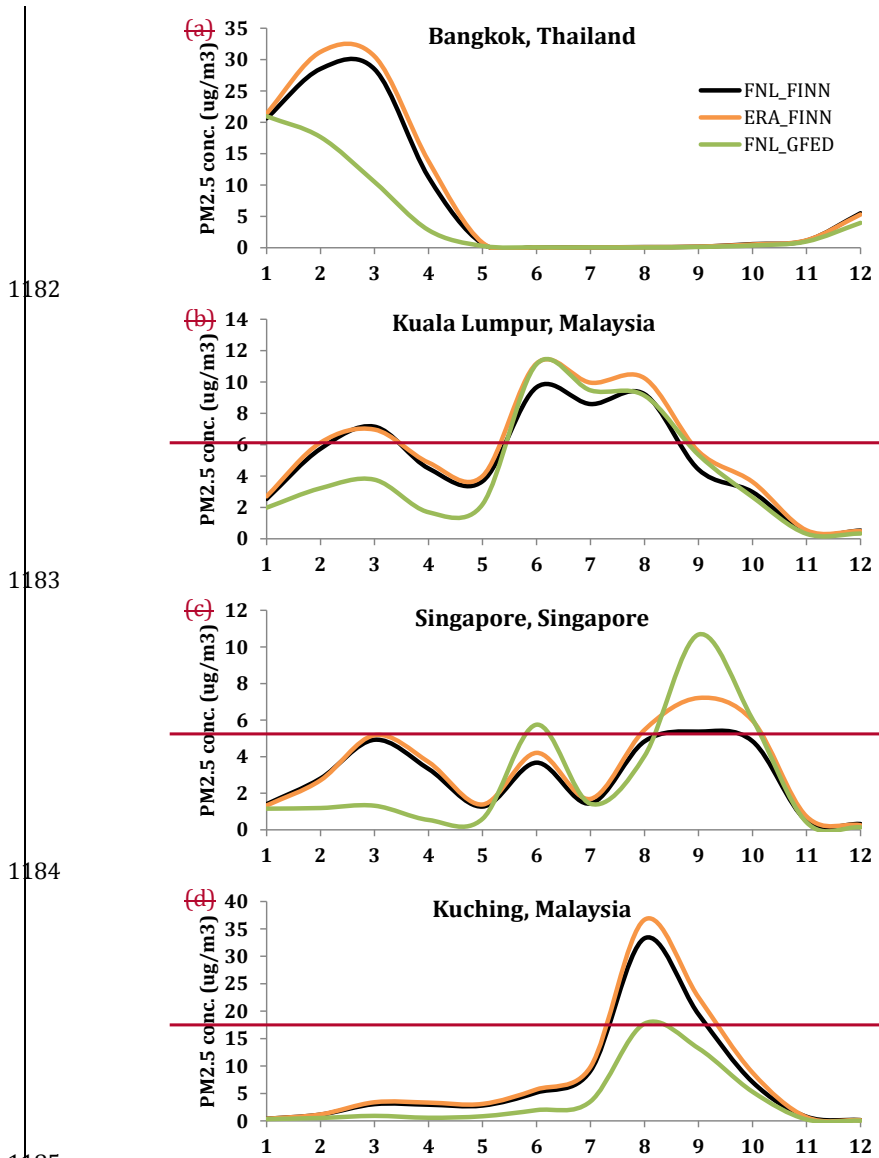
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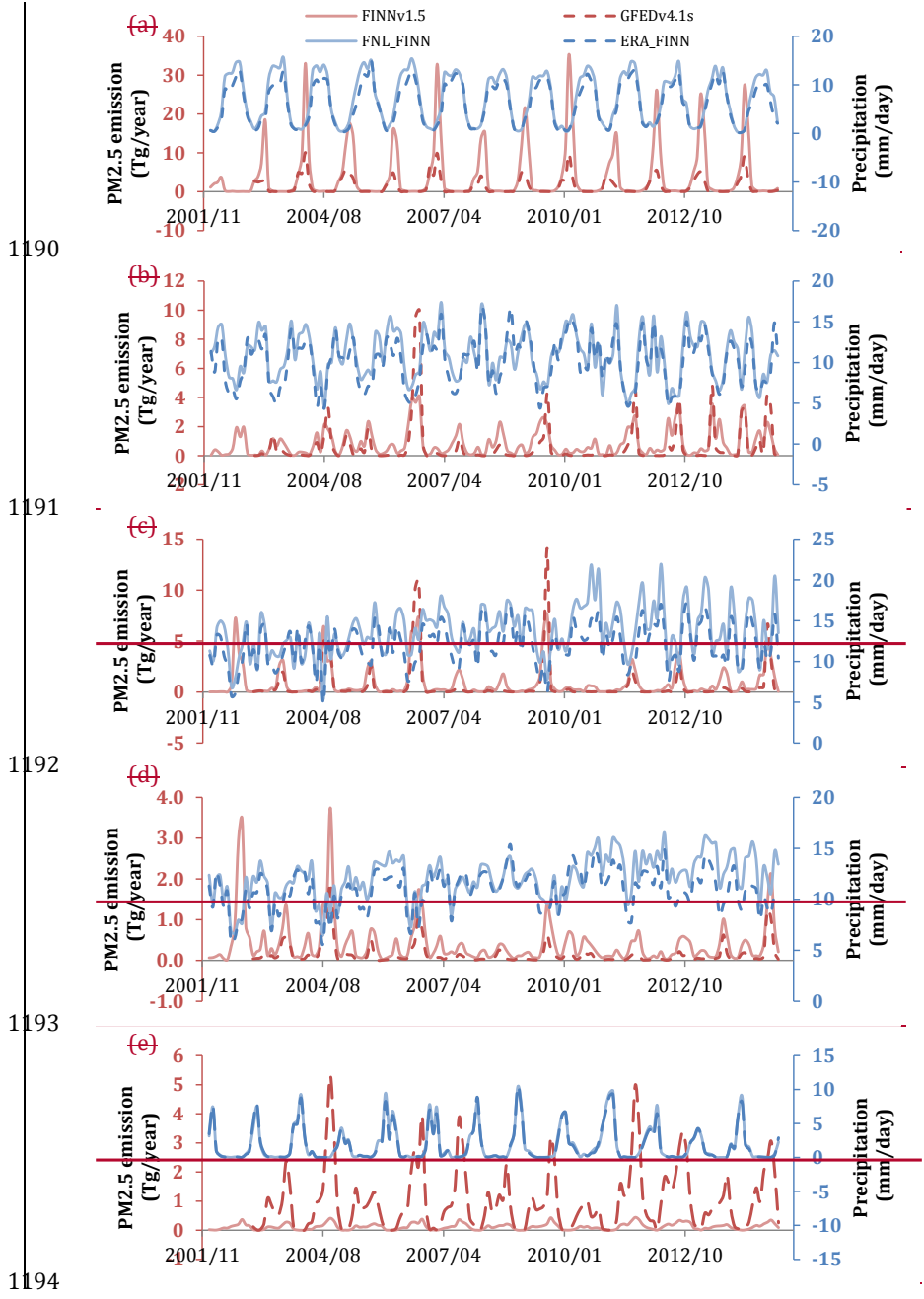
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Figure 11. The monthly variation of mean PM<sub>2.5</sub> concentration in FNL\_FINN, ERA\_FINN, and FNL\_GFED in: (a) Bangkok, (b) Kuala Lumpur, (c) Singapore, and (d) Kuching over the period 2002-2014 (FNL\_GFED is from 2003 to 2014).



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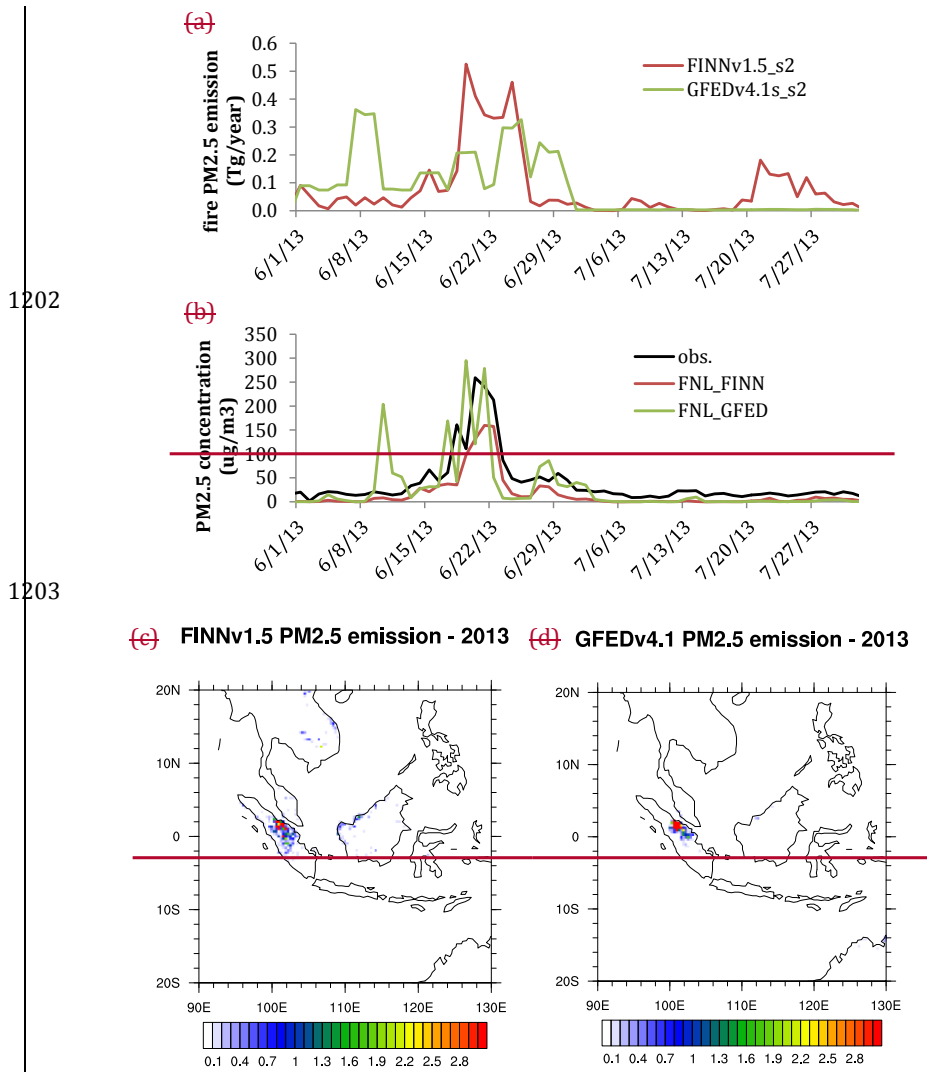
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1195 Figure 12. Temporal variation of monthly  $PM_{2.5}$  emission ( $Tg\ year^{-1}$ ) in FINNv1.5  
1196 (pink solid lines) and GFEDv4.1s (red dashed lines). Also shown are precipitation  
1197 rates ( $mm\ day^{-1}$ ) simulated in FNL\_FINN (light blue solid lines) and ERA\_FINN (blue  
1198 dashed lines) during 2002-2014 in: (a) Mainland Southeast Asia (s1), (b) Sumatra  
1199 (s2), (c) Borneo (s3), (d) the rest of the Maritime Continent (s4), and (e) northern  
1200 Australia (s5).  
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 1205 **Figure 13.** (a) Time series of daily mean PM<sub>2.5</sub> emissions (Tg year<sup>-1</sup>) in Sumatra (62)  
 1206 from FINNv1.5 (red line) and GFEDv4.1s (green line). (b) Time series of daily mean  
 1207 PM<sub>2.5</sub> concentration (µg m<sup>-3</sup>) in Singapore from observation (black line), and modeled  
 1208 results from FNL\_FINN (red line) and FNL\_GFED (green line). (c) Monthly mean PM<sub>2.5</sub>  
 1209 emissions (Tg year<sup>-1</sup>) from FINNv1.5 in June 2013. (d) same as (c) but from  
 1210 GFEDv4.1s.  
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