Author Response

Anonymous Referee #3

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The authors are taking advantages of an open biomass burning episode observed at five stations in the Yangtze River Delta area in China to study the processes leading to such an episode and its impact on PM2.5, organic carbon, and elementary carbon loads. Based on observations first, the study is then completed by HYSPLIT back-trajectory analyses and an air quality modeling study which both help in understanding the regional origins of the pollution observed at each station.
The manuscript contains interesting findings and the scientific contents match the ACP standard. However, some points in the manuscript, listed below, remain unclear and need to be addressed before this article deserved to be publish in ACP. Technical or spelling issues are also listed below.

- We appreciate the reviewer for his/her careful review of the paper. The paper has been modified to address all the comments and a detail point-by-point response is also given below.

Comments:
Abstract:
In the abstract, the sentence L 11 starting with "Daily minimum mixing..." is not corresponding to any point discuss in the paper. It should be remove or alternatively, the authors should discuss the low mixing depth values within the text.

- We have followed your suggestion and revised the sentence to "Mixing depth during the stagnant period was 240-399 m". Such statement can be found in section 3.2 of the manuscript.

Introduction:
p. 30690, L16-27: Please be more precise with the time scale of the numbers and percentages you're giving in this section. Are the values corresponding to decadal mean, annual mean, monthly mean? This information will be important when discussing your data.

- The numbers are monthly mean or seasonal mean. We agree with the reviewer the time scale information is important and have added such information in the revised manuscript. The revised text is given below,
"For the prescribed burning, the concentration contribution is estimated to vary at 0.3–5.1 μgm^{-3}, 2.8–43 % of the monthly ambient $PM_{2.5}$ (particles with aerodynamic diameters no more than 2.5 μm) load in Australia and the United States (Reisen et al., 2013; Tian et al., 2009). The contribution of residential wood heaters is at the range of 3.2–9.8 μgm^{-3}, 27–77 % of the seasonal $PM_{2.5}$ load in winter of southeastern United States and Australia (Reisen et al., 2013; Zhang et

al., 2010). In the winter of Portugal, the contributions of residential wood heaters to seasonal organic carbon (OC) and elemental carbon (EC) reaches 12.3 and 1.8 μgm^{-3}, accounting for 64 % and 11 % respectively (Gelencsér, 2007). The biomass burning contribution to seasonal ambient $PM_{2.5}$ mass is much higher in China, i.e., 12–27 μgm^{-3} (15–24 %) in Beijing (Cheng et al., 2013; Song et al., 2007; Wang et al., 2009), 5.4–25.4 μgm^{-3} (4–19 %) in Guangzhou (Wang et al., 2007) and 8–64 μgm^{-3} (<70 %) in Southeast Asia and south China (Fu et al., 2012)."

p. 30691, L1-3: A reference is needed here to support this statement.
Other studies on biomass burning episodes in China using modeling approach to identify source regions need to be mention in the introduction (e.g., Fu et al. 2012; Wang et al. 2007). A short paragraph must be added here in which similar studies could be used to put this study in context.

-The paragraph for p. 30691, L1-3 has been rewritten. The revised part is as follows:

"The biomass burning contribution to seasonal ambient $PM_{2.5}$ mass is much higher in China, i.e., 12–27 μgm^{-3} (15–24 %) in Beijing (Cheng et al., 2013; Song et al., 2007; Wang et al., 2009), 5.4–25.4 μgm^{-3} (4–19 %) in Guangzhou (Wang et al., 2007) and 8–64 μgm^{-3} (<70 %) in Southeast Asia and southern China (Fu et al., 2012). For the YRD region, contribution of biomass burning to the ambient $PM_{2.5}$ concentrations are seldom quantified and reported, especially for heavy haze episode. Such information is vital for development of further pollution control strategies."

Materials and methods:
p. 30692, L. 13: "Soil was... ". I'm not sure to understand what the term "Soil" is used for. Can you clarify this terminology? p. 30692, L. 15-16: The sentence "The trace elements..." is unclear and need to be reword.

- The term of "soil" means the crustal material (elements and their oxides) from the Earth, while trace elements represents the metal elements (usually heavy metal) that are mainly emitted by anthropogenic sources. In order to avoid confusion, we change these two terms to "crustal material" and "trace species" throughout the paper according to the definition of the following reference.

Yang, F.; Tan, J.; Zhao, Q.; Du, Z.; He, K.; Ma, Y.; Duan, F.; Chen, G., Characteristics of PM2.5 speciation in representative megacities and across China. Atmospheric Chemistry and Physics 2011, 11, (11), 5207-5219.

P 30693, L. 14: Please add words to clarify that you are now writing about your study. (e.g., "In this study, non-soil...")

- We have revised the sentence as you suggested.

Section 2.4: WRF/CMAQ model:
(i) References are needed for the WRF and CMAQ models.

- Three references used in the US, south Asia and China are added as ", which are

widely used over the world (Knipping et al., 2006; Wang et al., 2010; Fu et al., 2012),"

Knipping, E. M., Kumar, N., Pun, B. K., Seigneur, C., Wu, S.-Y., and Schichtel, B. A.: Modeling regional haze during the BRAVO study using CMAQ-MADRID: 2. Source region attribution of particulate sulfate compounds, Journal of Geophysical Research: Atmospheres, 111, D06303, doi:10.1029/2004JD005609, 2006.

Wang, S., Zhao, M., Xing, J., Wu, Y., Zhou, Y., Lei, Y., He, K., Fu, L., and Hao, J.: Quantifying the Air Pollutants Emission Reduction during the 2008 Olympic Games in Beijing, Environmental Science & Technology, 44, 2490-2496, doi:10.1021/es9028167, 2010.

Fu, J. S., Hsu, N. C., Gao, Y., Huang, K., Li, C., Lin, N. H., and Tsay, S. C.: Evaluating the influences of biomass burning during 2006 BASE-ASIA: a regional chemical transport modeling, Atmospheric Chemistry and Physics, 12, 3837-3855, doi:10.5194/acp-12-3837-2012, 2012.

(ii) p. 30963, L. 26. "...while the WRF domain was a 12 km extension in four directions." This sentence is unclear and need to be clarify. Are the WRF and CMAQ domains different?

- Yes, WRF has a larger domain to avoid numerical effects associated in the vicinity of the WRF domain boundaries. Two rows of grid cells at each boundary were trimmed off when we run CMAQ. This sentence has been deleted to avoid unclear expression and revised to "The CMAQ modeling domains were shown in Fig. 1a, with the outer domain of 36 km×36 km for China, the medium domain of 12 km×12 km for eastern China and the inner domain of 4 km×4 km for the YRD area."

(iii) How are distributed the fourteen vertical levels? How many of them are representing the boundary layer?

-There are twenty-four vertical layers. The information on vertical layers has been added in the revised manuscript,

"Twenty-four vertical layers were included from the height of the surface to 100 mbar (about 16 km), of which thirteen layer are included under the boundary layer height of 2 km."

(iv) The authors referred to a paper by Fu et al. (2013) in which "the detail model configuration and parameters" are given. However, the Fu et al. (2013) paper is describing an emission inventory and no mention of a modeling study (at least involving WRF/CMAQ) is done in it. Please replace this reference by the correct one or alternatively, insert a description of the model configuration and set-up in the text.
(v) Whatever the way the point (iv) is addressed, a brief description of the aerosol scheme used in the simulations is necessary (e.g., modal or sectional approach, organic carbon specific treatment?)

-Yes, we have replaced the reference with Fu et al.(2014). The related text has also been revised to explain the model configuration,

"The first guess fields of WRF model were from the analysis data of the National Center for Environmental Prediction (NCEP), as well as the Automated Data Processing (ADP) data used for four-dimensional data assimilation. The updated 2005 carbon bond gas-phase mechanism (CB05) (Whitten et al., 2010) and the AERO6 aerosol module with updates of primary organic aerosol (POA) aging (Simon and Bhave, 2012) and secondary organic aerosol (SOA) yield parameterization were used in CMAQ model. The detailed information about WRF and CMAQ model configuration and parameters were given in Fu et al. (2014)."

Fu, X., Wang, S. X., Cheng, Z., Xing, J., Zhao, B., Wang, J. D., and Hao, J. M.: Source, transport and impacts of a heavy dust event in the Yangtze River Delta, China, in 2011, Atmos. Chem. Phys., 14, 1239-1254, doi:10.5194/acp-14-1239-2014, 2014.

Simon, H. and Bhave, P. V.: Simulating the Degree of Oxidation in Atmospheric Organic Particles, Environ. Sci. Technol., 46, 331–339, doi:10.1021/es202361w, 2012.

Whitten, G. Z., Heo, G., Kimura, Y., McDonald-Buller, E., Allen, D. T., Carter, W. P. L., and Yarwood, G.: A new condensed toluene mechanism for Carbon Bond CB05-TU, Atmos. Environ., 44, 5346–5355, doi:10.1016/j.atmosenv.2009.12.029, 2010.

Results and Discussion:

p. 30694, Were the five stations set-up for a specific field campaign or are they measuring continuously all over the years? If long-term measurements are available, it would be great to compare the haze episode daily average with the daily average over the entire year.

-Yes, the five stations are measuring continuously all over the years. The daily average PM concentration over the entire year has been added and compared with that of the episode in the revised manuscript.

"During the entire year (from 1 May 2011 to 30 April 2012), the daily average concentration of the five sites is 86 μgm^{-3} for PM_{10} and 50 μgm^{-3} for $PM_{2.5}$. The average PM_{10} and $PM_{2.5}$ concentrations of the episode are 44% and 76% higher than the average of the entire year. In addition, the $PM_{2.5}/PM_{10}$ mass ratio was 66% during the episode, 58% higher than the annual average."

p. 30695, L. 23: Please mention that you now move to observations from another instrument/technique. Are PM 2.5 observations from the filters in agreement with these from TEOM?

-Thanks for the suggestion. We have added some text to explain the different dataset and to make comparison between different datasets in the revised manuscript as:

"The daily average concentrations of $PM_{2.5}$ species during the episode, which are from the laboratorial analytical result from the sampling filters, are reconstructed

and shown in Fig. 3. The gap between the sum of reconstructed $PM_{2.5}$ species and gravimetric mass, which is marked as "Others" in Fig. 3, is 11% for the average of the three sites."

p. 30696, L. 2-4: The sentence starting with "The increase in OM, ..." Why were the meteorological conditions responsible for the secondary aerosols formation enhancement? More explanations are needed here such as a reference to support this statement.

-The sentence has been revised and one reference has been added. The revised text is given as below,

"Increase of OM, sulfate and nitrate indicates that meteorological conditions might have enhanced the formation of secondary aerosols through accumulating and increasing the concentrations of gaseous precursors like SO_2, NOx and VOCs, and their oxidation rates (Fu et al., 2008)."

Fu, Q., Zhuang, G., Wang, J., Xu, C., Huang, K., Li, J., Hou, B., Lu, T., and Streets, D. G.: Mechanism of formation of the heaviest pollution episode ever recorded in the Yangtze River Delta, China, Atmospheric Environment, 42, 2023-2036, doi:10.1016/j.atmosenv.2007.12.002, 2008.

p. 30696, L. 13-24: Are all the information given regarding the overall stations? If yes, please mention it in the text.

- Yes, all the information are about the average of five sites. The explanation for all the numbers has been added in the revised manuscript.

p. 30696, L. 19-20: "...increased more rapidly..." This expression doesn't sound scientifically rigorous, please reword.

- Yes, the sentence was revised to "Maximum daily Non-soil K^+ concentration among the five sites reaches 5.4–18.3 μgm^{-3} in Phase II, 3.5–15 times of that in Phase I."

p. 30696, L. 22-24: Sulfate and Nitrate aerosols are anthropogenic pollution markers. Can their concentrations increased be linked to an anthropogenic influence, in addition to the biomass burning described in the paper?

-Yes, we agree with the reviewer's comments. The modeling results between scenarios with and without biomass burning are compared and summarized. The related text is added in the revised manuscript:

"In order to investigate the sources of $PM_{2.5}$ and its chemical components, we compare the modeling concentration distance between the base scenario with biomass burning and the scenario without any biomass burning for Phase II. It is found that the biomass burning emissions increase the concentration of OM and EC 2.2–6.6-fold and 1.0–3.7-fold, respectively, while that of sulfate and nitrate only increase 2.0–4.2% and 19-38%, respectively. The modeling results illustrates that the high concentration of OM and EC in Phase II are mainly from biomass

burning. Nitrate is partly from biomass burning. The increase of sulfate shall be due to the accumulation of anthropogenic emissions under stagnant meteorological conditions rather than biomass burning emission."

p. 30696-30697: The sentence starting by "The map show..." should be rewrite since 28 and 29 May maps are not shown in Fig. 4.

- We apologize for the error. The related sentences about the situation of 28 and 29 May have been deleted in the revised manuscript.

p. 30698, L. 2: "The mixing depth of Nanjing site during..." Is is the mean value along the period? If it is, please mention it.

- Yes, the word of "average" has been added before "mixing depth".

p. 30698, L6-10: The statement given here that the (very) low increase in wind speed would enhance the horizontal dispersion and the result in the temporary reduction of PM concentrations should be support by a reference to a relevant study or remove.

- We investigate the reasons for the short-term PM peak during June 1 for Hangzhou and Suzhou again. We agree with the reviewer and delete the above statement in the revised manuscript.

Also, the visibility in Nanjing is anti-correlated with the relative humidity during phase I but also during phase II. Does this mean that the pollution doesn't impact the visibility in Nanjing as much as at the other sites? Why would that be? This should be discuss in the text.

- The reasons that the visibility in Nanjing is different from the other sites are summarized below. Firstly the starting time of Phase II in Nanjing is June 2, one day later than that in other sites due to the meteorological conditions and different biomass burning regions. As a result, the PM pollution of Phase II shows similar variation with the RH value for Nanjing site, which are also different from other sites and resulting in the visibility anti-correlated with the relative humidity. We have added explanation on this in the revised manuscript as follows:
"Ambient RH exhibits typical diurnal variation (shown in **Fig. 5**), usually with the peak value in the midnight and valley value in the noon due to sunshine. However, the visibility is affected both by the PM pollution level and RH value. For all the sites except for Nanjing, the PM pollution in Phase II was accumulated without notable diurnal variation, resulting in the continuous low visibility. For the Nanjing site, the PM pollution in Phase II also shows diurnal change, the same as that of RH variation. Hence the visibility in Nanjing site also varied diurnally with the valley value in the midnight and peak value in the noon during Phase II."

- The difference of Nanjing site pollution is mainly due to the difference of biomass burning area and transport. This is explained in detail in Section 3.2 as:
"On 31 May, fires are mainly located near Hangzhou Bay in northern Zhejiang Province, the southern border of the Shanghai municipality, and southern Jiangsu

Province around Tai Lake; only limited fires are found in the area close to Nanjing. For Shanghai, Hangzhou, Ningbo and Suzhou, the main air flow is from the south, mixing with the pollutants from fires along the path. As a result, the main hot spots of PM_{10} pollution concentrated in the area of Shanghai and northern Zhejiang Province with daily average concentration over 200 μgm^{-3}. The situation changes on 4 June. Compared with that on 31 May, most fire spots are located in the north, i.e., central Anhui Province and southern Jiangsu Province. The air flow is from the south for the five sites. As a result, high PM_{10} concentrations occur in Jiangsu Province. The daily $PM_{2.5}$ concentrations in Nanjing are between 150 and 290 μgm^{-3}, followed by Suzhou (104 μgm^{-3}). In contrast, the concentrations at the other three sites are all less than 70 μgm^{-3}, and not affected by the biomass burning."

p. 30698, L. 16-17: Regarding back-trajectories on Fig. 7. Information on the back-trajectories time scale and the altitude would be useful. Over how days are plotted the back-trajectories? Are the back-trajectories staying in the boundary layer? Several back-trajectories, especially on May 31 are coming from over the ocean. How this is influencing the observations? Also, Fig. 7 would be clearer if the regions presented in Fig 1a were highlighted in it. This would be especially useful when one will confront the results presented in Fig. 9 with the back-trajectories.

- The configuration and parameters has been explained in the section 2.3: "The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph, 2013; Rolph, 2013) was run in the back-trajectory mode at 100 m AGL starting at 12:00 LST of 31 May and 4 June, and every three hours repeated thereafter, for the running time of previous 24 h."

- We also investigated the height of back-trajectories on 31 May and 4 June, and found some of them are transported from over 1000 m, higher than the boundary layer 24 hr before. However, taking Shanghai and Suzhou of 31 May and Nanjing of 4 June for example(shown in the following images), the air mass go down quickly to below 500m at least 24 h, and below 200m at least 6 h before getting to the observation sites. The long-time horizontal transport under the boundary layer will provide sufficient chances of biomass burning emissions reaching the observation sites.

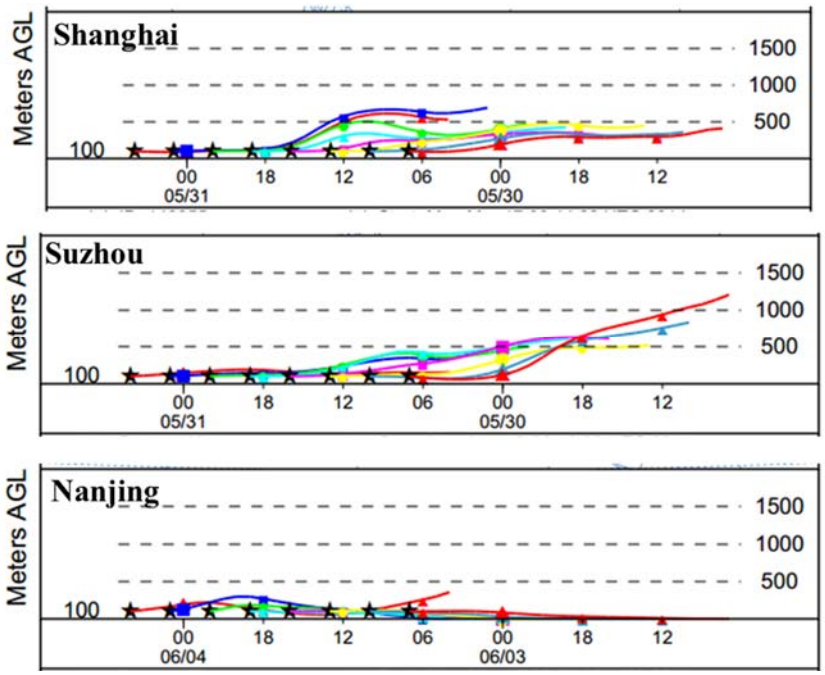

Fig. 7 has been updated by highlighting the four regions.

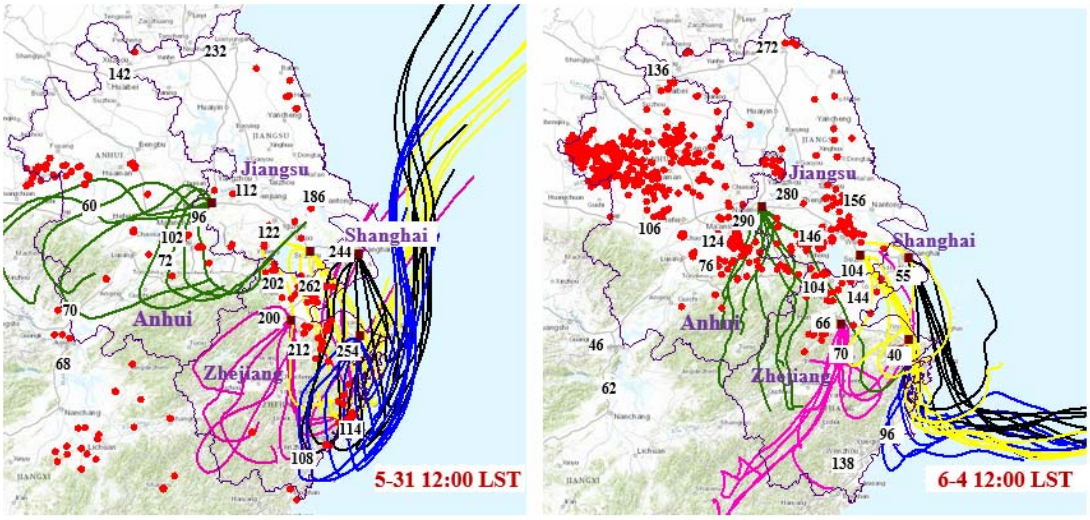

Fig. 7. HYSPLIT 24 h back–trajectories at 100m a.g.l. originating at each monitoring site (black squares) calculated every 3 h beginning at 12:00 LST and ending at 09:00 LST the previous day. Red dots represent the satellite-detected fires (FIRMS,Davies et al., 2009). Numbers are the daily average PM10 mass concentrations from air quality monitoring (http://datacenter.mep.gov.cn/report/air_daily/air_dairy.jsp). Back–trajectory colors are: Black-Shanghai, Blue-Ningbo, Pink-Hangzhou, Yellow-Suzhou, Green-Nanjing.

p. 30700, L. 4-8: The correlation coefficient between the observed and simulated PM2.5 concentrations would be very useful, in addition to normalized mean bias, to support the statement “Figure 8 compared the modeled and measured … as

measurements". The authors should add it in Fig. 8 or, alternatively, in a separate table.

- The correlation coefficient has been added to Fig. 8 and the related explanation has been added to the text in the revised manuscript.

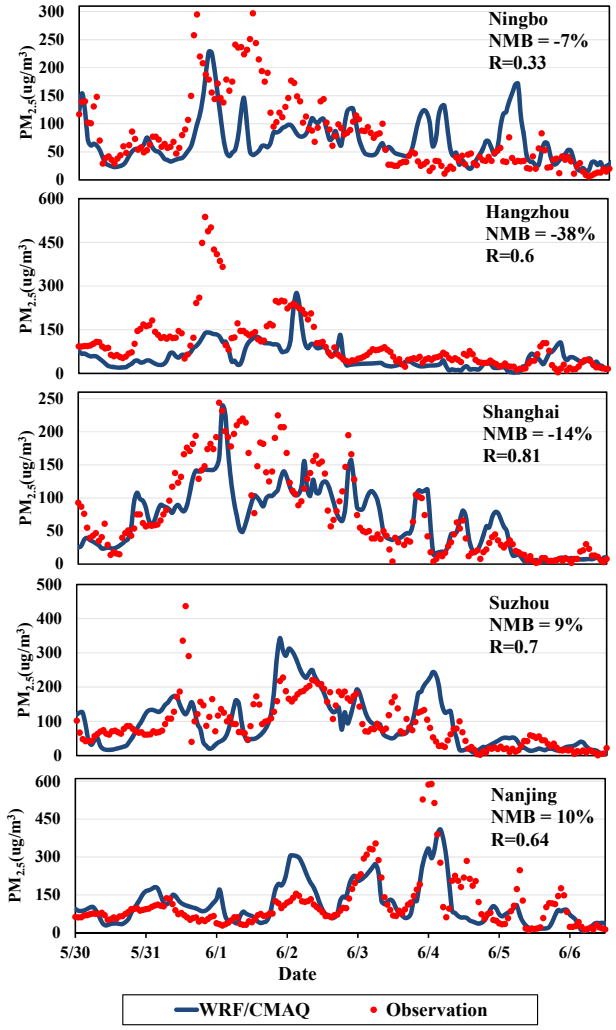

Fig. 8. Comparison of CMAQ simulations (blue lines) and TEOM–measured (red dots) hourly PM2.5 mass concentrations. NMB means normalized mean bias. R means the correlation coefficient.

p. 30700, L. 13-14: Conversely, the model is simulating PM 2.5 peaks while observations are flat. This is particularly true for Ningbo (on 4, 5 and 7 June) and Suzhou (2 and 4 June). Is this also due to the uncertainties in the biomass burning emissions? Using the modeling approach presented later in this paper, is it possible to identify the origins of these "fake" contributions?

Fu, J. S., Hsu, N. C., Gao, Y., Huang, K., Li, C., Lin, N.-H., and Tsay, S.-C.: Evaluating the influences of biomass burning during 2006 BASE-ASIA: a regional chemical transport modeling, Atmos. Chem. Phys., 12, 3837-3855, doi:10.5194/acp-12-3837-2012, 2012.

Qiaoqiao Wang, Min Shao, Ying Liu, Kuster William, Goldan Paul, Xiaohua Li, Yuan Liu, Sihua Lu, Impact of biomass burning on urban air quality estimated by organic tracers: Guangzhou and Beijing as cases, Atmospheric Environment,

Volume 41, Issue 37, December 2007, Pages 8380-8390, ISSN 1352-2310, http://dx.doi.org/10.1016/j.atmosenv.2007.06.048.

- Thank you for the comments. The model overestimates $PM_{2.5}$ peaks in Ningbo (on 4, 5 and 7 June) and in Suzhou (2 and 4 June) mainly because of the uncertainty in the temporal and spatial distribution of biomass burning emissions. We distribute the emission from biomass open burning based on the number and brightness of active fire data. But considering the influence of clouds, satellite data from May 20 to June ten were used. This may lead to extra emissions along the transport path and overestimating the $PM_{2.5}$ concentration in monitoring sites. Further studies shall be conducted to improve the temporal and spatial distribution of biomass burning emissions.

p. 30700, L. 15-17: It is claimed here that "the simulated meteorological fields and other anthropogenic emissions have been verified at other sites". This should be move to the beginning of the paragraph and completed with more information, i.e., a table with numbers, a figure with time series, or a reference to a paper which is providing such comparisons.

- More information has been added:
"Firstly the meteorological parameters of WRF model are compared with the observation dataset of National climate data center (NCDC) of the US. The average biases between the two datasets are acceptable with 0.44 ms^{-1} for wind speed, 1.03 degree for wind direction, -0.55 K for temperature and 0.26 g kg^{-1} for relative humidity."

- In addition, we revised this sentence and explained in more details:
"The better simulation performance during Phase I and III, which are less affected by biomass burning, illustrates that the non-biomass burning anthropogenic emission inventory and its distribution is reasonable and acceptable. Conversely, the outliers during the Phase II indicates that some uncertainties of the biomass burning emission amount and its spatial distribution still exists, especially for the time with thick cloud cover which affects the quality of satellite information."

p. 30700, L. 24-25: The sentence "Another reason...of supporting data." is senseless and need to be reword.

- The sentence has been changed to "Another reason is that the air quality transport model and receptor model use different source apportionment methods, as well as different inputs."

Technical review:

Abstract:

L. 14, "air model simulation", do you mean "air quality model simulations?"

- Yes, revised as suggested.

Section 2:

p. 30692, L. 3 replace "TOEM" by "TEOM"

- Revised as suggested.

p. 30692, L. 13 “measurements”

- Changed to “measurements”.

p. 30694, L. 11 replace “... and an additional five runs dropped biomass emissions...“ by “... and five additional runs in which biomass burning emissions were dropped...” Are they dropped to zero? If yes, mention it.

- Yes, the biomass burning emissions were dropped to zero. The sentence has been revised to “and followed by additional five runs in which biomass burning emissions for each sub-region were dropped to zero in sequence”.

p. 30694, L. 15 Instead of “grids”, you probably mean “grid cells” here?

- Yes, it should be “grid cells”. We have revised it as you suggested.

Section 3:

p. 30695, L. 12 replace “...measured the average concentrations of PM2.5 and PM10 of...” by “...measured PM2.5 and PM10 average concentrations of...”

- We have revised it as you suggested.

p. 30697, L. 20: replace Table 2 by Table 1.

- We have corrected it in the revised manuscript.

p. 30698, L. 17: Replace “... fires under high cloud cover...” by “...fires due to high cloud cover...”

- We have revised the sentence as you suggested.

p. 30698, L. 19: Switch “Nanjing” and “the four other sites”.

- In order to make it clear, we split the expression of these two days, and the sentence was revised to “hence 4 June was selected to represent Phase II for Nanjing while 31 May for the other four sites.”

p. 30699, L. 1: replace “for five sites” by “for the five sites”.

- We have revised the sentence as you suggested.

p. 30699, L. 12: The expression “...which is well understood...” should be replace.

- The sentence has been revised to:
“Our findings for the biomass burning regions that affected Nanjing agree with the above studies, indicating that the crop locations might not have changed in recent years.”