We would like to express our sincere thanks to the two reviewers for their careful reading and constructive suggestions, which have helped us improve the quality of this manuscript. We have addressed all their comments carefully and revised the manuscript accordingly. The detailed responses to their concerns and comments are presented as follows.

Anonymous Referee #1

This paper summarizes the overall performance of several fully-coupled air quality models that participated in the MICS-Asia III intermodal comparison study. It is the first part of a multi-part study. While the paper is well organized and the discussion is straight-forward, there are numerous places where the grammar needs to be fixed. I have tried to make some suggestions in my specific comments; however, the authors should not assume I have found all the problems. There are some aspects of the manuscript that are not explained well, such as the rationale for this paper in relation to future parts and the rationale of the intercomparison framework. By the time I finished the paper, I feel like I did not learn anything substantially new; therefore, the authors have not adequately highlighted the new results of this study.

Major Comments: 1) In the introduction, the authors talk about Topic 3 of MICS-Asia phase 3 which is the subject of this paper. At the end of the paper I felt like I did not get any information regarding the feedbacks. Perhaps the paper title is implying that those details will be included in subsequent parts. It would be useful at the end of the introduction to have a clear understanding of what the objectives are for this paper, versus subsequent parts that will appear.

Response: Thanks for this great suggestion. In the last paragraph of introduction, we claim that "This paper presents and overview of the MICS-ASIA III Topic 3, serving as the main repository of the information linked to Topic 3 simulations and comparisons". To make it clear, we added in the manuscript: "Specifically, this paper aims to archive the information of participating models, how the experiments, and results of model evaluation. The results of the MICS-Asia Topic 3 experiments looking at the direct and indirect 135 effects during heavy haze events will be published in a companion paper, part II."

2) The authors do speculate why there are differences among the models; however, the paper would be improved significantly if they went into more detail into a few instances to find more concrete reasons for the differences. This might require more analysis of the results. But as the

paper stands, it does not shed any new light on why the air quality models could differ. In the conclusion, the authors state that the paper provides "some directions of future model developments", but I see no evidence of that in the paper. The authors could also do a better job at citing papers that examine processes that might be missing or poorly represented in the air quality models.

Response: Thank you for pointing out. We added more explanations on why model differs in the revised manuscript. For example, the radiation differences in M6 is due to the use of different meteorological boundary conditions, and the averaged values were taken from 3hourly outputs. More importantly, high liquid water path in M6 lead to lower radiation from M6 in north China.

We have cited more papers on the reasons for underestimation of sulfate and SOA, and clearly claim the future directions for future model developments: "X. Huang et al. (2014) found including natural and anthropogenic mineral aerosols can enhance sulfate production through aqueous-phase oxidation of dissolved SO2 by O3, NO2, H2O2 and transition metal. Gao et al. (2016), Wang et al. (2014), and Zhang et al. (2015) also emphasized the importance of multiphase oxidation in winter sulfate production. However, these processes are currently not incorporated in the participating models for this study, which might be responsible for the apparent under-predictions of sulfate concentration";

"An et al. (2013) incorporated photoexcited nitrogen dioxide molecules, heterogeneous reactions on aerosol surfaces, and direct nitrous acid (HONO) emissions into the WRF-Chem model and found these additional HONO sources can improve simulations of HONO and nitrate in north China. M7 also predict high nitrate concentrations (N2O5 and NO2 gases react with liquid water, Zheng et al., 2015), and the predicted lower nitrate concentrations from other models are probably due to missing aqueous phase and heterogeneous chemistry, or the implementations of different gas phase oxidation in these models. Many studies have been conducted regarding sulfate formation issues. Nitrate also account for a large mass fraction in PM2.5 during winter haze events in north China, yet less attention was attracted to fully understand its formation. It is worth furtherly digging into the details about how different processes contribute to high nitrate concentrations in future studies";

"R. Huang et al. (2014) also suggested that low temperature does not significantly reduce SOA

formation rates of biomass burning emissions. Most models over-simplified SOA formation"; "Zhao et al. (2015) comprehensively assessed the effect of organic aerosol aging and intermediate-volatile emissions on OA formation and confirmed their significant roles. All these results suggest more complicated SOA scheme are needed to improve organic aerosol simulations during haze events".

3) The purpose of the MICS-Asia phase 3 is to look at feedback effects. I gather that is not that subject of this paper, and this paper is showing the initial evaluation of the aerosol simulations that will be important when looking at feedback effects later. The authors go into some detail on evaluating aerosol composition, but do not say anything about size distribution. Size distribution will be just as important for optical and CCN properties. I suggest adding a section that compares the predicted size distribution in some manner. I assume there are some sort of size distribution measurements that could be utilized. If not, it would still be useful in the context of the subsequent papers.

Response: Thanks for this good suggestion. We are aware that size distribution is crutial to optical properties and CCN formation, and thus direct and indirect effects. The participating models use mode and sectional approaches with different degree of complexity, we present the basic information on size distribution in each model, such as geometric mean radius and standard deviation and the number and range of size bin, and briefly mention about its potential influence on AOD simulation. We know this is not enough and we will investigate this issue in more detail in the companion paper (part II) by conducting additional sensitivity simulations with different size distribution while keeping other conditions the same. For this paper, we add more information on aerosol size distributions in the models in Table 1. Except M2 using 8-bin MOSAIC sectional approach, the other models use mode and bulk approach, with similar mean radius and standard deviation for anthropogenic aerosols or no size information. Unfortunately, we don't measurement of size distribution for comparison.

Specific Comments:

Line 43: Change "resolutions" to "resolution".

Response: We have changed.

Line 56: I would change “are consistent” to “are similar”. “consistent” can imply that the model results are good, but they could all consistently disagree with data. Change “haze event” to “haze events”.

Response: We have changed.

Line 58: “some brief senses” is an awkward phrase and should be replaced. The abstract could be shortened so that it contains only the most important findings. For example, the sentences in lines 44 – 48 could be removed. The whole abstract is rather weak.

Response: We have changed “provide some brief sense of” to “present”; we remove lines 44-48. This abstract is mainly used to introduce Topic 3 and how simulations were done and analyzed. Major findings out of Topic 3 will be published in the following companion papers.

Line 66: Change “but primarily in Asia” to “but most deaths occur primarily in Asia.”

Response: We have made this change.

Line 74: I would not use a semi-colon here and just have two sentences, although the second would need to be rephrased slightly.

Response: We have made this change.

Lines 134-139. There appears to be no underlying motivation for how the air quality models are compared. The only constraint on the models was the use of the same emissions inventories and they had to provide a set of variables. To better isolate the differences among the models, it would have been useful to have similar domains, grid spacings, and boundary conditions. I understand it would make the setting up the models a bit more difficult, but it would significantly reduce the differences arising from boundary conditions and spatial resolution. There are already many differences associated with the internal treatments of meteorology, chemistry, and aerosols. What I am looking for here is some further explanation as to why MICS-Asia organizers found the current configuration sufficient.

Response: We tried our best to constrain the differences, by requesting the modeling groups to use the same emission inventories, domains, grid size and boundary conditions, but some models has been constructed and tested on their own configurations, such as M5 and M6, and all the modeling groups use a similar resolution ~50km (Table 1) except M4 (15km). M4 actually covers a nested domain of M3. We include this simulation to further look at the impacts of grid spacing on aerosol feedbacks, which will be documented in the companion paper.

Models are quite different from each other, and it is difficult to keep all the inputs the same. Even using the same boundary conditions, mapping species from global chemistry simulations to different chemical mechanism each model uses can also lead to differences. We describe what we provided in the methodology section was intended to reduce the differences in inputs. Results from these model configurations have been published to report aerosol feedbacks and aerosol direct and indirect effects in Asia. The underlying motivation is to look at how current reported values differ, and provide possible range of these values. Given this motivation and our efforts on constraints, we think our current configuration could be sufficient although not ideal.

Line 153: The Grell reference is correct, but it only describes the initial model which did not discuss any of the feedback processes – which seems to be the focus of MICS-Asia Phase III. Those feedbacks were first implemented in Fast et al. (2006) and revised in subsequent manuscripts.

Response: We have added Fast et al. (2006).

Line 182: For VBS, need to cite Adhamov et al. (2012) in which it was developed and described.

Response: We confirmed that M1 did not use VBS, so we deleted description here.

Lines 190-191: The sentence regarding SOA is not correct and misleading. A VBS SOA treatment has been available in the public version of WRF-Chem for several years. What the authors mean to say is that the version of MOSAIC used in this study includes no SOA. The correct language here should imply that the users have chosen not to include SOA.

Response: We changed it to "The MOSAIC version used in M2".

Section 2.1, in general. The description of the models is uneven. Some sections to into some details about the aerosol model, such as noting the geometric means of the modes (e.g. M5) but not going into the same detail as another model (e.g. M1). For one model the details of how optical properties and hygroscopicity are discussed, but then another model the same level of detail is not discussed. The authors need to revise this section to have the appropriate level of detail for all models.

Response: Thanks for these good suggestion. We reorganize the whole section to make it easier to read and keep details for all models even.

Lines 232 – 239: The text discusses differences in the physics configurations as it should, but I assume the other models have physics differences too. Why not state that?

Response: Thanks for these good suggestion. We have added other physical configurations in Table 1 and in the texts.

Line 286: What does regridding mean? To handle the emissions inventories properly, the emissions need to be reapportioned so that mass is neither gained or lost. Regridding implies interpolation, that to me indicates a poor method of handling the emissions from one domain to another.

Response: Yes, the emissions were handled using mass conservative method to make sure that mass is neither gained nor lost, not simple interpolation.

Line 409: Change “are frequently happening” to “frequently happen”.

Response: We have changed.

Section 4, in general: this study relies on comparing model output to relatively few (at least for the PM data) point measurements. However, some discussion is needed to put the proper context of this type of comparison since the grid size differs among the models so there are issues of representativeness that must be considered.

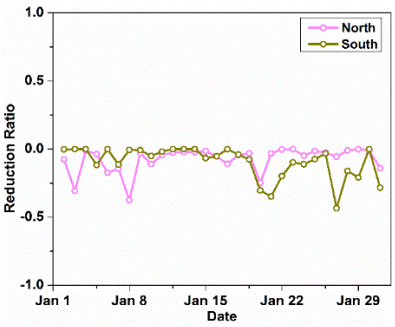
Response: This is a good point. We required the modeling groups use similar model resolutions. For most models, ~50km resolution is used. Except model evaluation, the model resolution would also affect simulations of meteorological fields and then chemical fields. We are preparing a manuscript discussing the influences of model resolution be keeping all other factors the same, which will be published in this topic series. Based on this, we think the comparison representativeness would be close for participating models. For model observation comparison, this is a common problem, given limited available observations in this study. In our previous experiences, the comparisons are done mostly in urban regions, and measurements during haze events show high homogeneity when we compared concentrations from different city sites in previous study. Besides, the in situ measurements at city sites exhibit a similar variation trend during haze events, reflecting haze pollution at regional scale rather than local scale, thus the concentration difference resulting from grid size would reduce.

Line 442: Turbulent mixing is missing from this description, which is not the same as transport.

Response: We have added turbulent mixing.

Lines 469-483: What is missing from this discussion is how clouds affect the prediction of downward shortwave radiation. I assume that the clouds are the main factors controlling clouds, but there is no mention of this. Would be useful to include what the clear-sky values are in Figure 5.

Response: Thanks for this great point. Since clear sky radiation is not submitted by modeling groups and it is not a default model outputs, it might not be appropriate to show it in Figure 5. However, we do think it is important to include the discussion here because we agree with you that it is one of the main factors controlling radiation. We add a figure to show the reduction ratio of downwards shortwave radiation due to clouds (SW-SWclearsky)/ SWclearsky in the SI, and discussion on clouds: "Clouds are also important to alter radiation. To exclude its impacts on the radiation shown here, we calculated the reduction ratio of radiation due to clouds. During the severe haze period (16-19 January 2010), the averaged reduction fraction is 5.9% in north China and 4.2% in south China, suggesting the relatively lower radiation during this period shown in Figure 5(d) is mainly caused by aerosols, while the lowest radiation on 20 January was caused by clouds (Figure 5(d))."



Line 493: Change "larger near surface" to "larger near the surface".

Response: We have changed.

Line 498: Awkward sentence – need to revise.

Response: We removed "only".

Line 513-514: Change to "All models produce similar CO predictions" based on how I understand this sentence.

Response: We have changed.

Line 522: It is rather surprising that the models produce better PM than ozone. Usually it is the other way around.

Response: This is probably because previous good ozone simulations mostly occur in summer. The study period is frequently affected by heavy haze in winter, when photochemistry is weaker and other chemical processes maybe more complicated. Another reason is that primary PM (including POA, BC etc) is of similar magnitude to secondary aerosol in winter haze period, which means meteorological and chemical processes are equally important.

Line 548: I doubt that sea-salt emissions are responsible for differences in PM10.

Response: We recheck the model and possible reason for the differences in PM10. Fig 7d shows that M5 largely overpredict PM10 concentration during 1-8 January at Rishiri, where is less affected by continental aerosols, overestimation of sea salt emission under certain meteorological conditions (such as large wind) is responsible for such positive biases, whereas in the rest of January, M5 perform relatively better and is generally consistent with most of the models.

Line 569-580: Chen et al., ACP (2016) is just but one paper that describes possible missing reactions associated with sulfate. It would be useful for the authors to delve a bit deeper into the literature to find such issues associated with models. Most community models are inherently dated and do not necessarily have the most up-to-date chemistry treatments since it takes time for new research findings to make there was in to those community models.

Response: We have added several papers describing the model problems and how missing mechanism can explain the underestimation of sulfate and other model problems. All the participating models have not implemented such mechanism, which is an important source for the underprediction of sulfate in this study.

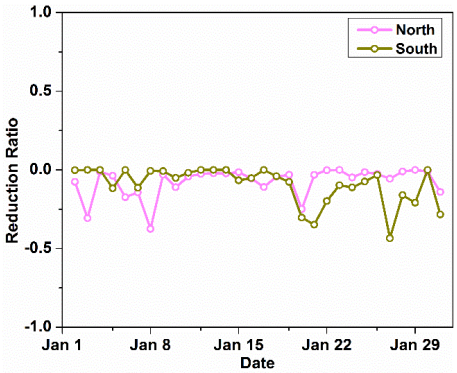
Line 593-594: There are probably other reasons as well for errors in nitrate predictions.

Response: Thanks for mentioning this. We added the spatial plots of NOx in the SI. It is shown that NOx from M5 is the highest, yet the nitrate produced from M5 is lower than other models except M1, which suggest there could be some missing nitrate formation pathways or stronger deposition of nitrate and its precursors in M5. Other than the mentioned aqueous phase and heterogeneous chemistry, the implementation of gas phase chemistry in different models might also played a role here. We added this to the revised sentence: “, or the implementations of different gas phase oxidation in these models. Many studies were conducted for sulfate formation issues. Nitrate also account for a large mass fraction in PM2.5 during winter haze

events in north China, yet less attention was attracted to fully understand its formation. It is worth furtherly digging into the details about how different processes contribute to high nitrate concentrations in future studies."

Line 607: The authors list deposition, but this usually means dry deposition. What about wet scavenging? Same comment applies to line 610.

Response: Thanks for this point. Here we meant both dry and wet deposition. To make it clearer, we added "(dry deposition and wet scavenging)". We added a figure in the SI to check how often were clouds present during this month. There were clouds only in a few days, but they can be very important for wet scavenging for hydrophilic aerosols. Thus, we added wet scavenging in Line 607 and 610.



Line 607: Change predicted BC to "predicted BC at the surface"

Response: We have changed.

Line 609: I think the authors mean horizontal grid resolution and not "horizontal grid interpolation." I have no idea what the latter means in this context. Please be more specific.

Response: We have changed.

Line 612: Since POC is about the same from the models, then BC should be as well. So it is a bit of a mystery why BC from M2 and M7 are higher than the other models.

Response: It depends on the models treat deposition and aging processes. For example, in the GOCART aerosol model (M3 and M4), 80% of BC are assumed to be hydrophobic and then undergo aging to become hydrophilic in an e-folding time of 1.2 days. Hydrophilic aerosols will go through wet deposition. But in other models like M2 and M7, BC is assumed to be hydrophobic, thus the wet removal is less actived. We added these explanations in the revised manuscript.

Line 621: Find a reference for this comment – there are lots of papers to cite here.

Response: Thanks. We added the following citation.

Heald, C. L., Henze, D. K., Horowitz, L. W., Feddema, J., Lamarque, J.-F., Guenther, A., Hess, P. G., Vitt, F., Seinfeld, J. H., Goldstein, A. H., and Fung, I., 2008. Predicted change in global secondary organic aerosol concentrations in response to future climate, emissions, and land use change, J. Geophys. Res., 113, D05211, doi:10.1029/2007JD009092.

Line 638: This implies the model is missing a feedback, and I thought this study was about in the inclusion of feedbacks (see line 106 on topic 3).

Response: Yes, we are trying to say less VOCs should be able to convert to SOA under hazy conditions, but the model still used 10% yield, which could overestimates SOA.

Line 642: "dust deflation" is an odd phrase. What is that?

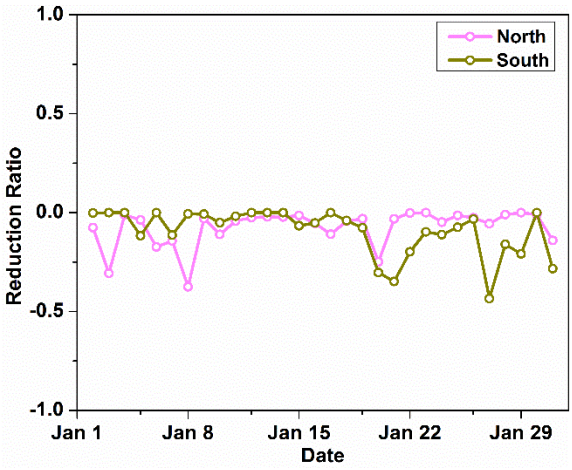
Response: We change to "wind-blown dust"

Lines 650-652: These sentences are poorly written. Suggest changing to "Only the sulfate predictions from M5 are close to the observed values. Sulfate is much lower than observed for all other models, except M6 which is too high. M2 and M7 predict reasonable nitrate concentrations. M3 and M4 overpredict OC during the haze period, but other models underpredict OC concentrations."

Response: We have changed following your suggestions.

Line 677: What about clouds? How often were AOD retrievals not possible due to cloudy conditions?

Response: Yes, it is possible. But in winter, there is relatively less cloud amount, so cloud is not a serious problem to AOD retrieval. For example, in Figure 14 and SI, about three days within a month (Jan. 2 Jan. 8 Jan 20) are due to clouds. We have added the influences of clouds in the text: "under serious pollution and cloudy conditions". Thanks for pointing out.



Line 680: The figure captions should also state that the AOD is a daily (daytime) value.

Response: We have changed following your suggestions: "daily (daytime) mean"

Line 686: Change "it's" to "it is".

Response: We have changed following your suggestions.

Line 718: Change "shows overprediction" to "shows an overprediction".

Line 724: Change "lower RH simulation" to "lower simulated RH".

Response: We have changed following your suggestions.

Line 726: Change "OC concentration" to "OC concentrations".

Response: We have changed following your suggestions.

Line 736-737: The authors have not shown this. It is very likely the size distribution and mixing state is treated differently. In this sense, the explanation provided previously in the paragraph is incomplete. I doubt one can really attribute the difference in AOD without a more rigorous analysis than the simple explanations presented here. At best, they are showing the range of AOD associated with all the differences among the models.

Response: Thank you for this question and suggestion. We added the mixing state of each model in Table 1. Only M6 used external mixing. Curci et al. (2015) discussed the impacts of mixing state on simulated AOD and found that external mixing state assumption significantly increase simulated AOD. M6 used external mixing but shows a relative lower AOD because of ignorance of other aerosol species like dust, sea-salt, etc.. Other models used internal mixing for major aerosol compositions. The size distribution treated in the models except M2 (sectional approach) and GOCART bulk approach is described by a lognormal distribution with similar geometric mean radius and standard deviation for different modes or species, such as

0.07 μm for inorganic aerosols, 0.01μm and 0.02 μm for BC and OC, but different bins for dust and sea salt (their concentrations are low in the north China Plain in winter). Thus we believe the differences in AOD shown here is mostly due to differences in simulated compositions. The size distribution in this sentence means the used modal size treatments in M1, M5, M6 and M7. To avoid misunderstanding, we change it to "lognormal treatments" and add the conclusion from Curci et al., (2015) about the impacts of mixing state.

Curci, G., et al. "Uncertainties of simulated aerosol optical properties induced by assumptions on aerosol physical and chemical properties: An AQMEII-2 perspective." Atmospheric Environment 115 (2015): 541-552.

Lines 773-775: I don't see how interpolation of emissions to the grid should lead to model uncertainties. Of course, there could be errors introduced to reapportion emissions from one grid to another. But these would only be large if the mathematical method of reapportionment is poorly treated. There are ways to ensure that such uncertainties are small.

Response: Thanks. We changed this sentence to "which might be caused by the treatment of aging and deposition (dry deposition and wet scavenging) processes."

Line 776: "Manifold" is a strange word to use in this context.

Response: We have changed to "various".

Line 783: And what are those improvements? I would that that such an intercomparison study such as this would provide more concrete recommendations.

Response: The above results provide some directions for future model development, such as new heterogeneous or aqueous pathways for sulfate and nitrate formation under hazy condition, SOA chemical mechanism with new VOC precursors, yield data and approaches, and the dependence of aerosol optical properties on size distribution and mixing state. We have added this in the revised manuscript.

Line 788- 792: This sort of general conclusion about model inter-comparisons studies is rather tired. It could have been written based on results already in the literature and by speculation, without even conducting this inter-comparison study. I wish he authors could be more specific here regarding the findings specific to this study. The authors have not investigated all these possibilities and only barely scratched the surface at isolating and ascribing the uncertainties to specific processes.

Response: This paper presents model evaluation and intercomparison for meteorological fields, aerosol concentrations and optical properties with a series of observations over East Asia. While these models exhibit a generally good performance for PM2.5 concentration, the simulations of chemical compositions differ largely, which lead to the large differences in optical properties, such as AOD, and this would further affect direct and indirect aerosol effects, and consequently radiation and cloud in this region. We will compare and examine how the different AOD levels and aerosol properties affect radiation and cloud and explore the strength and affecting factors of aerosol-radiation-weather interactions in the companion paper part II.

Conclusion: It would be useful to have some text that looks forward to the next part of the paper. Will the authors be looking at evaluating the feedbacks, which were not examined in this study? Another reason I am looking for some more concrete explanations in this paper for why the models differ is that the situation will only become more complicated when examining feedback effects.

Responses: Thank you for the good suggestion. We briefly describe the objective of the next part of the paper in the conclusion: "This paper focused on the evaluation of the predictions of meteorological parameters and the predictions of aerosol mass, composition and optical depth. These factors play important roles in feedbacks impacting weather and climate through radiative and microphysical processes."

Referee #2

The paper describes the setup of the MICS Asia Phase III model experiment. The results of seven simulations with online coupled meteorology-atmospheric chemistry models are shown and compared against observational data. The evaluation of online coupled air quality models and the outcome of the MICS-ASIA III model inter-comparison exercise are certainly worth to be published. However, the quality of the paper must be improved significantly before it can be published in ACP. In the first instance, the 'Results and discussions' section must be enhanced considerably. Attempts should be made to explain the reasons for the observed differences among the models. In particular, a more in depth discussion is necessary for those model results which look like outliers (for example solar radiation and ozone in Figure 5 and 6). Although the paper is overall well organized, it was nevertheless a though read for me.

It is sometimes difficult to keep track of the different models and their respective setups. Repetition of the model name along with the label once a while could improve this situation with only little effort. Enhancing the figure captions would also help. Finally, several sentences are quite convoluted and hard to understand. Splitting long sentences may improve the readability.

For these reasons and further reasons mentioned below, major revisions are required.

The specific comments include some suggestions how to improve the paper.

Response: We appreciate the detailed and constructive comments from referee #2 to help improve our manuscript. We have added more explanations for the shown model differences in the revised version. We also add model names in figure captions, rewrite some sentences, and split long sentences to improve the readability. The detailed point-by-point responses and changes are listed below.

Specific comments (also including minor points):

Abstract: In its current form the abstract raises the expectation that aerosol meteorology interactions will also be a topic of this paper. The reader may also expect that both episodes will be evaluated in the paper. Generally, the first part of the abstract is raising expectations, which are not fulfilled by the paper. Nevertheless, it is also OK to restrict the paper to the evaluation of first episode. However, the abstract must be reworded in this case in order to

avoid raising expectations which are not met later. It is not clear why the model evaluation is restricted to the first episode of the model inter-comparison and why the second episode is not considered.

Response: Thanks for pointing out. This manuscript is the overview paper for aerosol meteorology interactions topic, so we include it in the abstract to raise interests from audience in the companion papers. We deleted "Two winter months (January 2010 and January 2013) were selected as study periods, when severe haze occurred in North China." in the abstract to avoid the expectation. We restricted to the first case because all models submitted results for January 2010 and less models for January 2013. For other topics within MICS-ASIA III, only year 2010 was simulated. Besides, more measurements data are available for year 2010. We also found that we had enough materials (mostly project overview) to present for current manuscript, so decided to move the evaluations of the second case into part II manuscript. We have removed all descriptions for 2013 in the manuscript.

Introduction: The authors should also consult publications describing related work, e.g. the model evaluation papers related to HTAP as well as AQMEII Phase 2 and Phase 3.

Response: It is a great idea to include information from HTAP and AQMEII projects. In the introduction, we add "Other ongoing related modeling frameworks include the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) and the Air Quality Model Evaluation International Initiative (AQMEII). The TF HTAP was initiated to improve knowledge of the intercontinental or hemispheric transport and formation of air pollution, and its impacts on climate, ecosystems and human health (Galmarini et al., 2017; Huang et al., 2017). The AQMEII project specifically focuses on regional modeling domains over Europe and North America (Galmarini et al., 2017), within which aerosol meteorology interactions was studied (Forkel et al., 2015; Makar et al., 2015a, 2015b; San Jose et al., 2015) over Europe and North America."

Line 106: 'Various multi-scale models: Are there more than the five model and seven simulations described here?

Response: Yes. This sentence briefly describes a bigger picture of MICS-ASIA Phase III, not

just Topic 3. Other topics include other models, such as GEOS-Chem. To avoid misunderstanding, I add one more sentence in the context: "A detailed overview of MICS-Asia Phase III, including descriptions of different research topics and participating models, will be published in a companion paper".

Lines 114 –119: The references should not be restricted to Chinese authors.

Response: We include more references on this topic: "Forkel et al., 2015; Makar et al., 2015a, 2015b; San Jose et al., 2015" from AQMEII Phase 2.

Line 125: The paper claims to be 'serving as the main repository of the information linked to Topic 3 simulations and comparisons.' To achieve this aim, more details must be added to the descriptions of those models where die description is quite. Please add also some paragraphs in section 2.1 (similar to section 2.5) in order to make this section better readable.

Response: Sorry for the relatively poor presentation in section 2.1. We have reorganized section 2.1 following the format in section 2.5 to make it more readable.

Table 1: The numbers attached to 'WRF-Chem' and 'NU-WRF' are unnecessary and confusing. Therefore, they must be removed. On the other hand, the model version is an important information which must be added wherever applicable. The contents of the table are not precise: For example, for M1 'RACM' must be replaced by 'RACM-ESRL' and 'MADE' by 'MADE/VBS'. What climatological data are applied as boundary conditions for M6? Please consider to add also information about the details of the radiation calculation. As shown by Curci et al. (2015, Atmospheric Environment, Vol. 115) the inherent assumption have a strong influence on the calculated AODs.

Response: Thanks for these great suggestions. We delete the numbers attached to WRF-Chem and NU-WRF and add the model version. We replaced 'RACM' and 'MADE', and add climatological boundary data for each model. We add both longwave and shortwave parameterization information and mixing state information for each model to Table 1 and cite Curci et al. (2015) to emphasize the importance of radiation calculation on AOD.

Table S1: Please check also whether this table needs to be more specific (similar to Table 1). Please add also information about the two models which are missing in the table. Please consider also moving this table into the main part of the paper.

Response: To make it more specific, we add information for other missing models, including

climatological boundary data. We also merge S1 and Table 1.

Line 338: Please mention the source of the climatological data here or in Table 1.

Response: We add this in Table 1 in the revised manuscript.

Line 281: Please add the information which model uses the prescribed BVOC emissions and which one does the internal calculation (could also be added to Table 1).

Response: We add this in Table 1 in the revised manuscript.

Section 2.1: Please mention clearly in the text which aerosol meteorology interactions are switched on for each model.

Response: We include this information in Table 1 and add this in the revised manuscript.

Section 2.2: Please add some information on soil dust emissions (also to be included in Table 1).

Response: We add this information in Table 1 in the revised manuscript.

Section 2.3: Please include some information about the meteorological boundary conditions.

Response: We add this information in Table 1 in the revised manuscript.

Section 2.3: Please consider to add some information about the differences between the boundary values from the different data sources for selected variables (eventually to be included in the supplementary material). Looking into the boundary values may also help to understand differences between results of the different model runs.

Response: We add this information in the supplementary material as shown below. For aerosol species and gases except ozone, boundary conditions from the two models are pretty similar to each other. MOZART simulated ozone is higher than GEOS-Chem. Previous tests have showed that the influences of different boundary conditions have negligible impact on PM simulations, but larger impact on ozone (Abdalah et al., 2016). Topic 3 focuses on aerosol-weather-climate interactions in North China, so the impacts of different chemical boundary conditions are not quite important on our results. But it might be part of reason for poor ozone performance for some models. Thanks for this great suggestion. We have added this discussion in the revised paper.

Abdallah, C., Sartelet, K. and Afif, C., 2016. Influence of boundary conditions and anthropogenic emission inventories on simulated O 3 and PM 2.5 concentrations over Lebanon. Atmospheric Pollution Research, 7(6), pp.971-979.

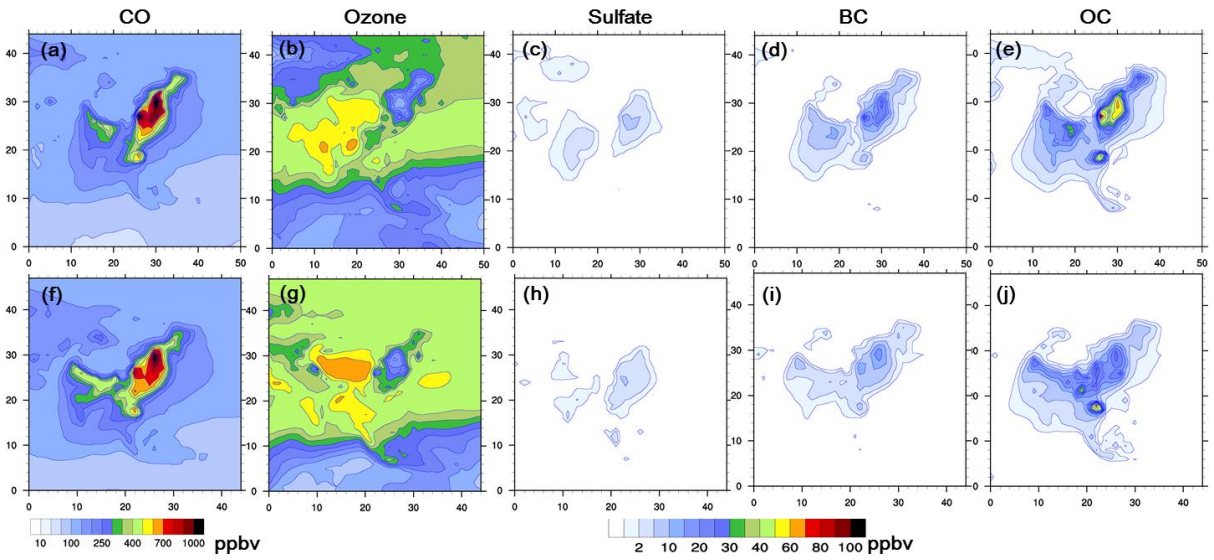


Figure Monthly mean near surface CO, ozone, sulfate, BC and OC from GEOS-Chem (a-e) and MOZART (f-i)

Line 389: This sentence is confusing. According to Table S1 all simulations considered here are performed with aerosol meteorology interactions switched on. Were the simulations additionally performed also without aerosol meteorology interactions for the investigation of feedback effects? Anyway, this could be mentioned in the introduction as well as in the outlook, but not at this place.

Response: Yes, Table S1 only specifies what kind of interactions were turned on. We also performed without interactions to check the differences. The differences between with and without interactions will be present in paper part II on aerosol-meteorology interactions. Following your suggestion, we move this sentence to the introduction section.

Line 392 –393: Why are abbreviations given for temperature, humidity, and wind, and units for the shortwave radiation?

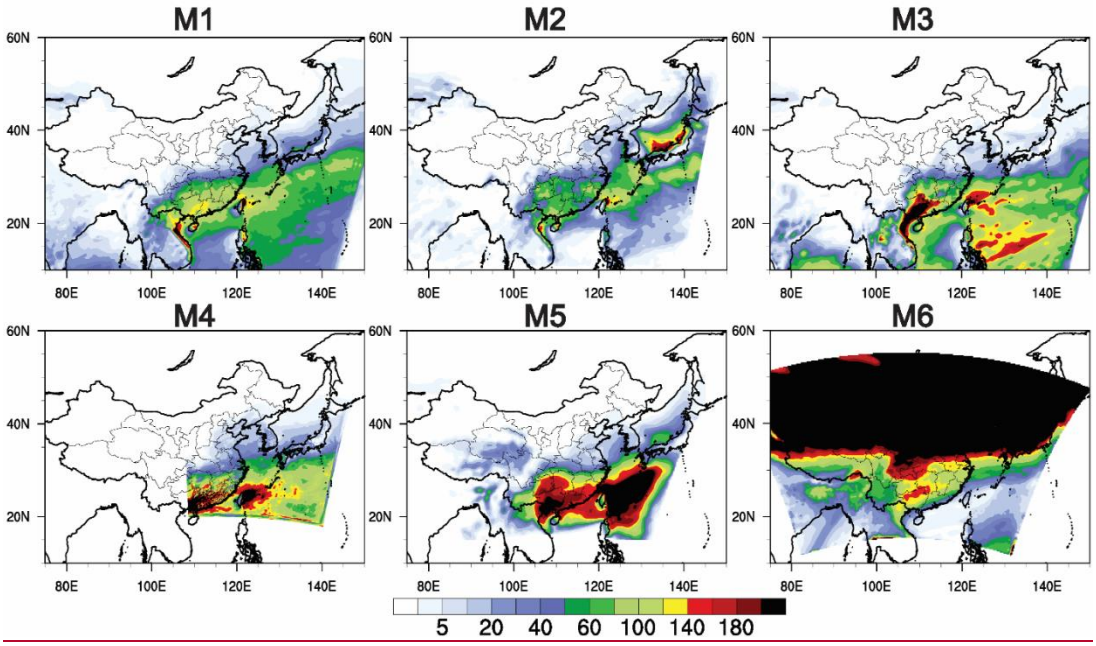
Response: To make it easier to show in figures.

Section 3: Why is the year 2013 described here, if it not discussed in the rest of the paper? Figure 3 could eventually be moved to the supplementary material.

Response: Following your suggestion, we move it to the supplementary material and remove corresponding sentences in the text.

Section 4.1: Cloud optical depth and integrated liquid water are important and should also be discussed (even, if no observational data are available).

Response: Following your suggestion, we add plots of integrated liquid water. We found this plot is good to explain why SWDOWN in north China is extremely low for M6. We added this point in the revised manuscript.



Line 427: This topic is not addressed in this paper.

Response: Thanks for pointing out. We delete it here and will provide discuss it the forthcoming companion paper.

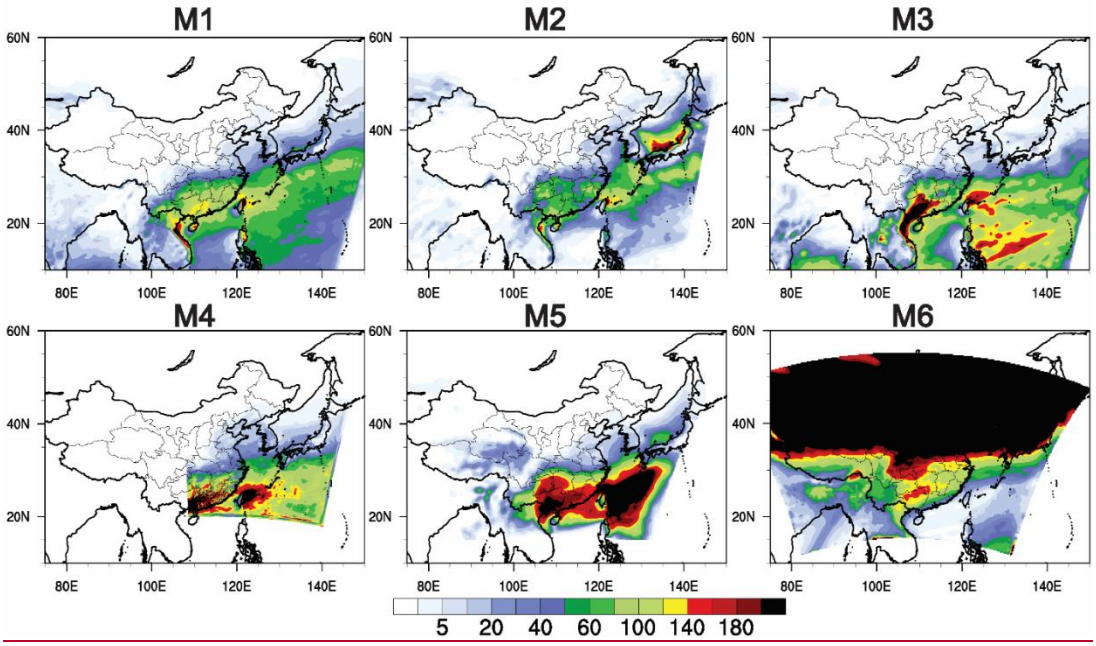
Line 473: What is the reason for the bad performance of M6 and M7? Please check also cloud cover. And why is this huge difference in radiation between M6 and M7 not reflected in T2? Why does M7, which is also WRF, show such a large difference to M1 and M2?

Response: The reason for M7 overestimation of SWDOWN in North China is the exclusion of aerosol-radiation interactions, which tends to overpredict SWDOWN especially in high PM days and areas. T2 looks fine because T2 nudging was used, so T2 is close to measurements in M7. M1 and M2 used very different settings, with aerosol-radiation interactions, and without using meteorology nudging. M6 simulates a lower SWDOWN due to overestimation of cloud integrated liquid water as mentioned above. Sorry for the confusion here. We add the descriptions of setting in Table 1 and text to make it clearer.

Line 480: Why is this the case? (Figure 5e), M6 and M7 show a better consistence with observations than over northern China sites.

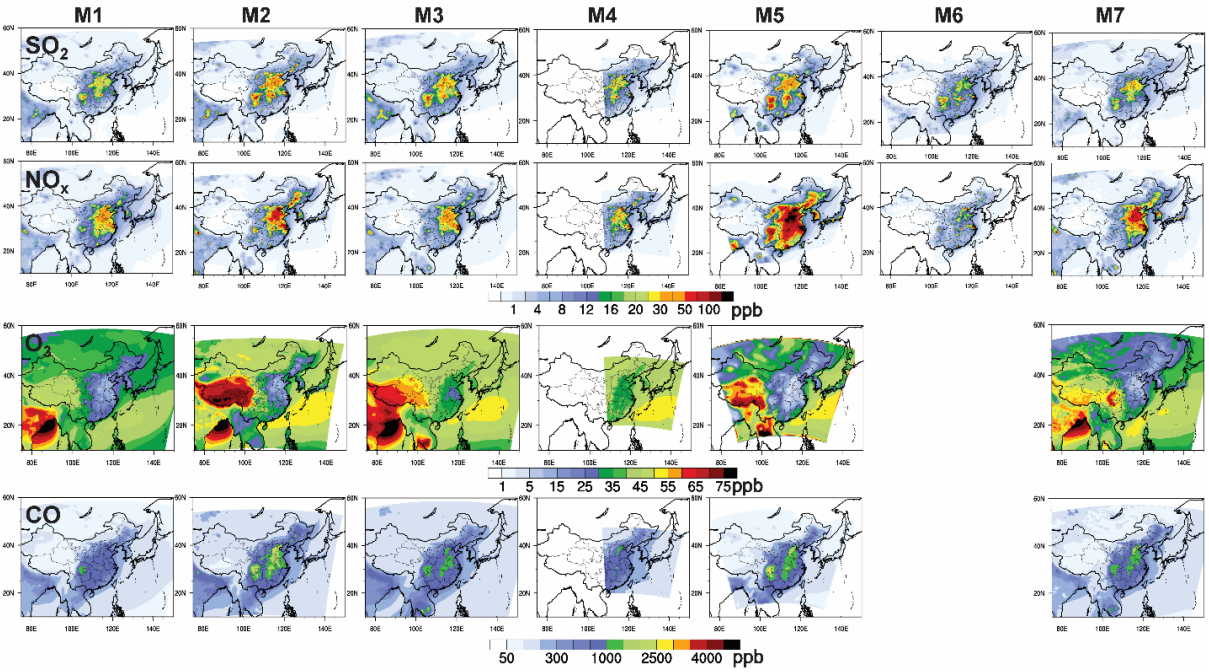
Response: We add plots of integrated liquid water, which may explain a better simulation of

M6 in southern China, and M7 shows a better agreement with observation because relatively lower PM level and weaker aerosol radiative effect in southern China.. We added this point in the revised manuscript.



Section 4.2: Spatial distributions of the gas phase pollutants (similar to figures 8 and 9) could be shown also in the supplement.

Response: Thanks for this suggestion. We have added in the revised manuscript.



Line 498: The ‘only’ is probably placed wrong here?

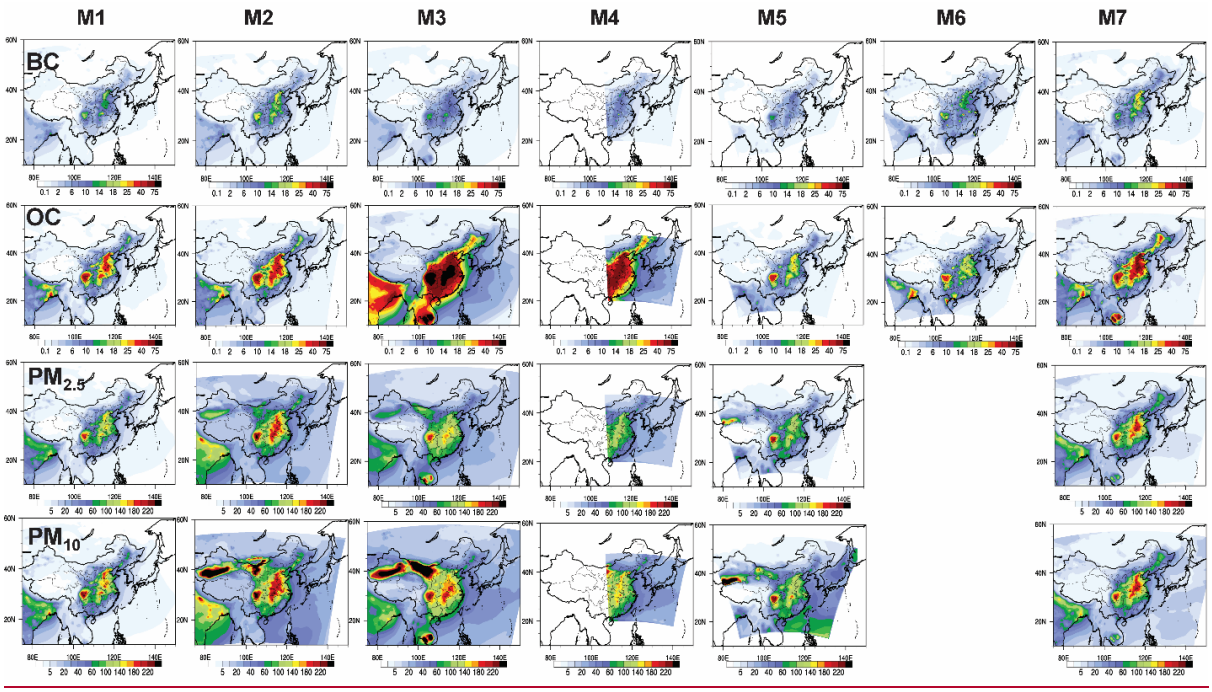
Response: Yes, we have removed it.

Line 504: Please give some evidence for this.

Response: This statement is based on the mean MBE averaged over all used CARE-China sites. It is typo here. We changed Beijing to "CARE-China sites".

Line 521: Please give more evidence for this statement. And what is the contribution of soil dust to PM10? What is the contribution of different of the dust emission parameterizations?

Response: In winter of the north Huabei Plain, soil dust generally contributes about 10% to PM concentration, but there is also primary PM from anthropogenic activity, such as power plant, traffic, construction etc. and this part of PM mostly settles in coarse mode, which may contribute to PM_{10}, but it's not clear if all the models include this emission sector and this sector is of higher uncertainty compared with other anthropogenic emission sectors. The schemes of dust deflation are similar in WRF-Chem series and different in M5, M6 and M7, the discussion on different dust schemes is less important given the relatively small contribution to $PM_{2.5}$. However, we added the dust implementation in Table 1 to provide how it affect PM10, which are shown below. The implementation of wind-blown dust are mostly significant in northwestern regions, and less important in Beijing and surrounding regions.



Line 524 – 526: What is the reason for the negative correlation? On an hourly basis, the diurnal course of ozone should reflect the course of the solar radiation. Or are the correlations just calculated from on daily values? Please clarify. How do the diurnal courses look like for the

individual models? Could the parameterization of dry deposition or differences in the lateral boundary conditions or differences in biogenic VOC emissions explain the overestimation of simulated ozone for M3 and M4? It is surprising, that the overestimated ozone seems not to be related to solar radiation, so there are probably other reasons for this overestimation. How do the ozone profiles look like, can they contribute to an explanation?

Response: We think the major causes for O3 overprediction in M3 and M4 are the combined effects of vertical diffusion and lateral boundary condition. M3 and M4 could predict larger vertical diffusivity coefficients, which leads to stronger vertical mixing, this not only results in lower NOx concentration and weaker titration of O3, but also stronger mixing of O3 from lateral boundary down to near surface, consequently causing higher O3 concentration than others.

Line 530: If the citation does not fit, it should be removed. Knote et al. (2015, Atmospheric Environment, Vol. 115) may probably suit better as this paper includes also a winter episode.

Response: Thanks. We replaced this citation with Knote et al. 2015.

Sections 4.3 and 4.4: It would be nice if soils dust were also included in the discussion.

Response: Thanks. We have added discussion of dust in the revised manuscript.

Lines 550 – 554: These general remarks are not necessary here.

Response: We remove this part.

Line 561: ‘M5 and M6 shows : : :’: Please observe the proper use of singular and plural (not only here but throughout the paper, e.g. line 582).

Response: We have changed these two and checked through the manuscript.

Lines 562 – 578: It is not clear in how far these statements apply to the models which are discussed here.

Response: We think these sentences are important to explain the possible reasons for the consistent underestimation of sulfate concentration from most of the models. Some important chemical processes for sulfate formation during hazy events are not implemented or fully considered in the participating models. M5 and M6 don't show the low biases possibly due to other compensatory processes, such as larger chemical reaction rate or lower deposition in the models.

Line 568 – 572: This sentence is quite hard to understand. Please split it into two or more sentences. Please also split other lengthy convoluted sentences.

Response: We have changed these two and checked through the manuscript.

Line 593: Is this just a general remark or is there some evidence for this? Does M7 really include heterogeneous nitrate formation?

Response: Yes, in default CMAQ, the heterogeneous nitrate related reactions include N2O5 +H2O and 2NO2 + H2O

Line 608 and line 772: How can this be? Emissions were supposed to be the same for all models.

Response: We explain this in response to the same question from another reviewer. Emission should be similar among models, but treatments of deposition and aging process are different, we rewrite the sentence.

Line 609: Was the vertical distribution of the emissions not prescribed? If there are differences in the vertical distributions, they must be described.

Response: All the models release emission at the surface. We have rewritten this sentence.

Line 616: According to line 191, M2 does not include SOA formation

Response: We have removed M2 here.

Line 616 – 617: The statement about SORGAM does not fit here as M1 includes a VBS approach (Ahmadov, R., et al., 2012, J. Geophys. Res.).

Response: Sorry, we double check with the modeler, and they confirm that SORGAM instead of VBS is used. We have changed the description in Table 1 and in the text.

Line 619: 'volatile' seems to be missing here

Response: We changed it to semi-volatile VOCs

Lines 619 – 624: These lines include quite general statements. How are they related to the models discussed in this paper?

Response: We have deleted this sentence.

Lines 642 – 643: This should have been mentioned in section 2.2.

Response: We have added the dust parameterization in section 2.2.

Lines 669 – 672: Please avoid this kind of redundancies (not only here).

Response: We have removed it and checked through the paper.

Lines 677 – 680: What is the contribution of soil dust during these situations?

Line 688: Please split this sentence.

Response: We have splitted.

Line 690 – 691: This statement is true, but unnecessary.

Response: We have removed.

Lines 692 – 698: These are quite general statements, which must be related to the applied models.

Response: We have removed

Line 710: M2 does not have modes

Response: We added (size bins)

Line 712: What are the consequences of this? If necessary, this can be discussed based on the findings by Curci et al. (2015, Atmospheric Environment, Vol. 115).

Response: Thanks for this good suggestion. We added the conclusion from Curci et al. (2015) that external mixing can increase simulated AOD but core-shell assumption is a minor issue.

Figures and figure captions:

Figure captions in general: More detailed descriptions must be given in the figure captions (are daily values or hourly values shown, relevant area, etc., depending on what is shown in the figure).

Response: Thanks for pointing out. We have added these information.

Figure 1: It looks like the labels M3 and M4 in the legend of Figure 1 are mixed up (According to Table 1 and Figures 8 and 9 M4 is the small domain). Furthermore, using the same colors for the model domains as for the curves in Fig. 5 etc. would make the reading a bit easier.

Response: Thanks for pointing out. We noticed it after submission. We have changed it.

Caption of Figure 1: Repeating the model names in the caption (for example 'M1: WRF-Chem, 45 km; M2 WRF-Chem, 50 km, M3 : : :') would make the paper a bit 'reader-friendly'.

Response: Thanks for this good suggestion. We have added: M1: WRF-Chem 45km; M2: WRF-Chem 50km; M3: NU WRF 45km; M4: NU-WRF 15km; M5: RIEMS-IAP 60km; RegCCMS 50km; WRF-CMAQ 45km

Caption of Figure 5: Please mention that this is a spatial average (for a, b and c: over which area) of daily values and explain error bars.

Response: Thanks for this good suggestion. We have added: (spatial daily values are averaged over measurements shown in S4 and S5; the error bars show the standard deviation of values over the measurement sites)

Figure 7: Please show also CO and PM2.5 although no measurements are available.

Response: Thanks for this good suggestion. We have changed.

Figures 8 and 9: Please include also PM10.

Response: Thanks for this good suggestion. We have changed.

Figures 8 and 9: The split into two figures appears quite arbitrary. Perhaps it would look better if the figures are organized differently: One figure with 7 rows (M1 – M7) and 3 columns (PM10, PM2.5, BC) and one figure with 7 rows (M1 – M7) and 4 columns
(SO24, NO3, NH+4, OC) – no obligation to do this, just a suggestion.

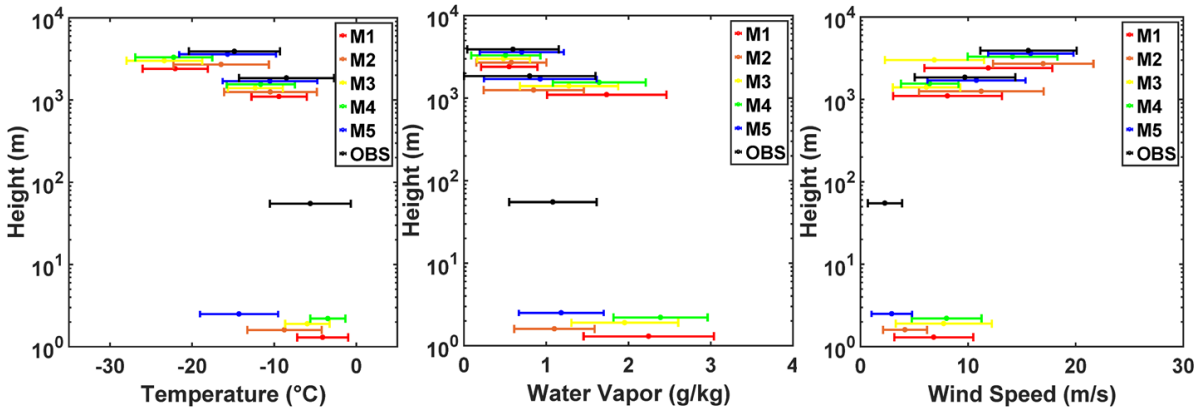
Response: Thanks for this good suggestion. We have added PM10 in Figure 9.

Spatial distributions of the gas phase pollutants (similar to Figures 8 and 9) would be nice in the supplement.

Response: Thanks for this good suggestion. We have added gas pollutants.

Figure S4: This figure seems to be contorted. Please improve the quality. Why are all the lines within the single 'height groups' (e.g. at '1 km') at different heights? Please explain in the figure caption. Also: explain what is shown here (daily values, hourly values, : : :?)

Response: Do you mean Figure S7? I used slight differences to better show there values, otherwise they overlap with each other. The shown values are monthly mean. I have added these information (near surface observation is at 55m and model predictions are at 2m; comparisons are conducted at near surface, 1km and 3km; shifts in heights are made to make it clearer to avoid overlapping) in the revised caption and replotted it to sole the contortion problem.


Height (m)
Temperature (°C)
Water Vapor (g/kg)
Wind Speed (m/s)
M1
M2
M3
M4
M5
OBS

Air Quality and Climate Change, Topic 3 of the Model Inter-Comparison Study for Asia Phase III (MICS-Asia III), Part I: overview and model evaluation

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Abstract

Topic 3 of the Model Inter-Comparison Study for Asia (MICS-Asia) Phase III examines how online coupled air quality models perform in simulating high aerosol pollution in the North China Plain region during wintertime haze events and evaluates the importance of aerosol radiative and microphysical feedbacks. A comprehensive overview of the MICS-ASIA III Topic 3 study design, including descriptions of participating models and model inputs, the experimental designs, and results of model evaluation, are presented. ~~Two winter months (January 2010 and January 2013) were selected as study periods, when severe haze occurred in North China. Simulations were designed to evaluate radiative and microphysical feedbacks, together and separately, relative to simulations without feedbacks.~~ Six modeling groups from China, Korea and the United States submitted results from seven applications of online coupled chemistry-meteorology models. Results are compared to meteorology and air quality measurements, including the Campaign on Atmospheric Aerosol Research Network of China (CARE-China) network, and the Acid Deposition Monitoring Network in East Asia (EANET). ~~The analysis focuses on model evaluations and aerosol effects on meteorology and air quality, and potentially other interesting topics, such as the impacts of model resolutions on aerosol-radiation-weather~~

~~interactions. The model evaluations for January 2010 show that current online-coupled meteorology-chemistry model can generally well reproduced meteorological features and variations of major air pollutants, including aerosol concentrations. The correlation coefficients between multi-model ensemble mean and observed near-surface temperature, water vapor mixing ratio and wind speeds can reach as high as 0.99, 0.99 and 0.98.~~ The correlation coefficients between multi-model ensemble mean and the CARE-China observed near-surface air pollutants range from 0.51 to 0.94 (0.51 for ozone and 0.94 for $PM_{2.5}$) for January 2010. However, large discrepancies exist between simulated aerosol chemical compositions from different models, which is due to different parameterizations of chemical reactions. The coefficient of variation (standard deviation divided by ~~average~~the mean) can reach above 1.3 for sulfate in Beijing, and above 1.6 for nitrate and organic aerosol in coastal regions, indicating these compositions are less consistent from different models. During clean periods, simulated Aerosol Optical Depths (AOD) from different models are ~~consistent~~similar, but peak values differ during severe haze events, which can be explained by the differences in simulated inorganic aerosol concentrations and the hygroscopic growth efficiency (affected by varied RH). These results ~~provide some brief senses of~~present how current online-coupled meteorology-chemistry models reproduce severe haze events, and provide some directions for future model improvements~~.~~, such as new heterogeneous or aqueous pathways for sulfate and nitrate formation under hazy condition, secondary organic aerosol (SOA) formation chemical mechanism with new volatile organic compounds (VOCs) precursors, yield data and approaches, and the dependence of aerosol optical properties on size distribution and mixing state.

1 Introduction

Air pollution in Asia, particularly in China and India, has been an increasing important research topic, and has attracted enormous media coverage since about 60% of the world population live and are exposed to extremely unhealthy air in this region. It is estimated that outdoor air pollution brings about 3.3 million premature deaths per year worldwide ~~but~~ with most deaths occur primarily in Asia (Lelieveld et al., 2015). In addition, the impacts of regional and intercontinental transport of Asian pollutants on air quality and climate change have been frequently reported (Akimoto, 2003; Menon et al., 2002, Ramanathan and Carmichael, 2008). Chemical transport models have been developed and applied to study various air pollution issues in Asia. For example, an Eulerian regional scale acid deposition and photochemical oxidant model was developed in the United States (Carmichael and Peters, 1984; Carmichael et al., 1986; Carmichael et al., 1991) and applied to study long-range transport of sulfur oxides (SO_x), dust and ozone production in East Asia (Carmichael et al., 1998; Xiao et al., 1997~~);~~). ~~a~~ A nested urban and regional scale air quality prediction modeling system was developed and applied to investigate ozone pollution in Taiwan (Wang et al., 2001). Although important advances have taken place in air quality modeling, large uncertainties still remain, which are related to inaccurate and/or incomplete emission inventories, poorly represented initial and boundary conditions and missing or poorly parameterized physical and chemical processes (Carmichael et al., 2008a).

Furthermore, many models used to study air quality in Asia have been developed in other regions (e.g., USA and Europe), and the assumptions and parameterizations included in these models may not be applicable to the Asian environment. In order to develop a common understanding of

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model performance and uncertainties in Asia, and to further develop the models for Asian applications, a model inter-comparison study was initiated, i.e., Model Inter-Comparison Study for Asia Phase I (MICS-Asia I), in 1998 during a workshop on Transport of Air Pollutants in Asia in Austria. The focus of MICS-Asia Phase I was to study long-range transport and deposition of sulfur within Asia in support of on-going acid deposition studies. Eight long-range transport models from six institutes in Korea, Japan, Denmark, the USA, and Sweden participated in MICS-Asia I. Multi-model results of sulfur dioxide (SO_2) and sulfate concentrations, and wet deposition amounts in January and May 1993 were compared with surface observations in East Asia (Carmichael et al., 2002). Source-receptor relationships and how model structure and parameters affect model performance were also discussed during this phase (Carmichael et al., 2002). In 2003, MICS-Asia Phase II was initiated to include more species, including nitrogen compounds, ozone and aerosols. The study period was expanded to cover two different years and three different seasons, and global inflow to the study domain was also considered (Carmichael et al., 2008b). Nine modeling groups from Korea, Hong Kong, Japan, the USA, Sweden, and France participated in this phase. Seven topics (i.e., ozone and related precursors, aerosols, acid deposition, global inflow of pollutants and precursors to Asia, model sensitivities to aerosol parameterization, analysis of emission fields, and detailed analyses of individual models) were discussed and published in a special issue of Atmospheric Environment (Carmichael et al., 2008b).

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In 2010, MICS-Asia phase III was launched and three topics for this phase were decided during the first and second Workshop on Atmospheric Modeling in East Asia. Phase III aims to evaluate strengths and weaknesses of current air quality models and provide techniques to reduce uncertainty in Asia (Topic 1), to develop a reliable anthropogenic emission inventory in Asia

(Topic 2), and to evaluate aerosol-weather-climate interactions (Topic 3). Various multi-scale models participated in this phase and the study periods range from year to month depending on study topics. This phase uses data from the Acid Deposition Monitoring Network in East Asia (EANET), in addition to new observations related to atmospheric chemistry in the region. A detailed overview of MICS-Asia Phase III, including descriptions of different research topics and participating models, will be published in a companion paper. An important advance to this phase is the inclusion of multiple online-coupled chemistry-meteorology models to investigate aerosol-weather-climate interactions, which is the target of topic 3. On-line coupled models are playing important roles in air quality, meteorology and climate applications, but many important research questions remain (Baklanov et al., 2017).

The influences of aerosols on meteorology, e.g., radiation, temperature, boundary layer heights, winds, etc. and $PM_{2.5}$ concentrations have been examined in previous studies using different online coupled models (Forkel et al., 2015; Gao et al., 2016a, 2016b, 2017a, 2017b; Han et al., 2012, 2013; Makar et al., 2015a, 2015b; San Jose et al., 2015; Tao et al., 2015, 2016; Wang et al., 2014; Zhang et al., 2010). In general, there are two ways of online coupling: online integrated coupling (meteorology and chemistry are simulated using the same model grid, and one main time step is used to integrate) and online access coupling (meteorology and chemistry are independent but data are exchanged on a regular basis) (Baklanov et al., 2014). These two different coupling ways can lead to uncertainties in the results of aerosol-weather-climate interactions. Even using the same coupling way, different parameterizations in different online models causes uncertainties as well. Thus, it is important to inter-compare how different online models simulate aerosol-weather-climate interactions~~.~~, particularly in heavily polluted Asian region. Other ongoing related modeling frameworks include the Task Force on Hemispheric

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Transport of Air Pollution (TF HTAP) and the Air Quality Model Evaluation International Initiative (AQMEII). The TF HTAP was initiated to improve knowledge of the intercontinental or hemispheric transport and formation of air pollution, and its impacts on climate, ecosystems and human health (Galmarini et al., 2017; Huang et al., 2017). The AQMEII project specifically focuses on regional modeling domains over Europe and North America (Galmarini et al., 2017), within which aerosol meteorology interactions was studied (Forkel et al., 2015; Makar et al., 2015a, 2015b; San Jose et al., 2015) over Europe and North America.

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This paper presents and overview of the MICS-ASIA III Topic 3, serving as the main repository of the information linked to Topic 3 simulations and comparisons. Specifically, this paper aims to archive the information of participating models, how the experiments, and results of model evaluation. ~~Simulations were designed to evaluate radiative and microphysical feedbacks, together and separately, relative to simulations without feedbacks.~~ The results of the MICS-Asia Topic 3 experiments looking at the direct and indirect effects during heavy haze events will be published in a companion paper, part II. This paper is organized as follows: in Section 2, we provide the inter-comparison framework of Topic 3, including the participating models, emissions, boundary conditions, observational data, and analysis methodology. Section 3 presents the general descriptions of the study periods and Section 4 presents comparisons and discussions focused on the results related to the meteorological and air pollution conditions during the January 2010 heavy haze episode. The results of January 2013 haze episode and detailed analysis of the direct and indirect effects will be presented in a companion paper.

2 Inter-comparison framework

In North China, severe aerosol pollution frequently happens and attracts enormous interests from both public and scientific communities (Cheng et al., 2016; Gao et al., 2015, 2016a, 2016b, 2016c, 2017). Two winter months in which severe haze episodes happened in North China were selected as the study periods for Topic 3. During these two months, maximum hourly $PM_{2.5}$ concentration in urban Beijing reached ~500 μg/m^3 and 1000μg/m^3, respectively. Compared to the China Grade 1 24-h $PM_{2.5}$ standard (35μg/m^3), daily mean $PM_{2.5}$ concentrations in urban Beijing exceeded this standard for 20 days and 27 days within these two months, respectively. The dramatically high aerosol loadings during these two hazy months substantially affected radiation transfer, and provide a good opportunity to study the aerosol effects on weather, air quality and climate. In this study, the participants were required to use common emissions to predict air quality during these two months and submit requested model variables. The emissions were placed on a publicly accessible website. Six modeling groups submitted results for Topic 3. In this section, we briefly describe these models and their configurations, introduce the emission inventories (including anthropogenic, biogenic, biomass burning, air and ship, and volcano emissions), observational datasets, and describe the analysis methodology.

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2.1 Participating models

Table 1 summarizes the characteristics of the participating models. These models include: one application of the Weather Research Forecasting model coupled with Chemistry (WRF-Chem, Fast et al., 2006; Grell et al., 2005) by Pusan National University (PNU) (M1), one application of the WRF-Chem model by the University of Iowa (UIOWA) (M2), two applications (two domains: 45km and 15km horizontal resolutions) of the National Aeronautics and Space Administration (NASA) Unified WRF (NU-WRF, Peters-Lidard et al., 2015; Tao et al., 2013) model by the Universities Space Research Association (USRA) and NASA's Goddard Space

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Flight Center (M3 and M4), one application of the Regional Integrated Environment Modeling System with Chemistry (RIEMS-Chem, Han et al., 2010) by the Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (M5), one application of the coupled Regional Climate Chemistry Modeling System (RegCCMS, Wang et al., 2010) from Nanjing University (M6), and one application of the coupled WRF-CMAQ (Community Multiscale Air Quality) model by the University of Tennessee at Knoxville (UTK) (M7). These models are all online coupled, which enables aerosol-weather-climate interactions. Domain setting of each model application is shown in Figure 1. The domains of M2, M5, and M6 (UIOWA, IAP, and NJU in Figure 1) cover most areas of East Asia, including China, North Korea, South Korea, Japan, Mongolia, and north parts of Southeast Asia. M1, M3 and M7 domains (PNU, NASA D01 and UTK) include more countries in Southeast and South Asia. M4 (NASA D02) covers east China, Korea and Japan. The descriptions of major model settings are listed below. More descriptions including microphysics, radiation, and boundary layer, are listed in Table 1.

(1) Model grids: The horizontal model resolutions of these applications range from 15km to 60km (Table 1). Model vertical resolutions vary from 16 to 60 layers (Table 1) and the set model top pressures range from 100mb to 20mb (Table 1).

(2) Gas phase chemistry: At PNU (M1), the RACM-ESRL (Regional Atmospheric Chemistry Mechanism, Earth System Research Laboratory) gas phase chemistry was used. RACM was developed based on Regional Acid Deposition Model (RADM2) to simulate regional atmospheric chemistry (Stockwell et al., 1997) (including 237 reactions) and the rate coefficients were updated in RACM ESRL version (Kim et al., 2009). At the University of Iowa (M2), CBMZ (Carbon-Bond Mechanism version Z) gas phase chemistry was used. CBMZ (Zaveri and Peters, 1999) extends the original CBM4 mechanism to function properly at larger spatial and

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longer timescales. The augmented CBMZ scheme includes 67 species and 164 reactions. The NU-WRF model (M3 and M4) uses RADM2 for gas phase chemistry. Both the RIEMS-Chem model from IAP (M5) and the RegCCMS model from NJU (M6) used CBM4 to calculate gas phase chemistry (Gery et al., 1989). The CBM4 version incorporated in RIEMS-Chem (M5) includes 37 species and 91 reactions, The version of CBM4 implemented in RegCCMS (M6) consists of 36 reactions (4 photolysis reactions) and 20 species (Wang et al., 2010). M7 applied SAPRC 99 to simulate gas phase chemistry. The SAPRC99 mechanism implanted within the CMAQ model has 88 species and 213 chemical reactions (Carter, 2000a, b).

(3) Aerosol modules: MADE/SORGAM (Modal Aerosol Dynamics Model for Europe/Secondary Organic Aerosol Model) aerosol module was coupled and used in M1. MADE uses 3 log-normal modes (Aitken, accumulation, coarse) and simulates major aerosol compositions, including sulfate, ammonium, nitrate, sea-salt, black carbon (BC), and organic carbon (OC). M2 uses 8 bin MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) aerosol module. MOSAIC considers major aerosol species at urban, regional and global scales, including sulfate, nitrate, ammonium, sodium, chloride, EC, and other unspecified inorganic species (such as inert minerals, trace metals, and silica) (Zaveri et al. 2008). The MOSAIC version used in M2 includes some aqueous reactions but no SOA formation. At NASA, the GOCART aerosol model (Chin et al., 2002) was coupled to RADM2 gas phase chemistry, and incorporated into the NU-WRF model (M3 and M4) to simulate major tropospheric aerosol species, including sulfate, BC, OC, dust, and sea-salt. In this aerosol model, 10% of organic compounds from the volatile organic compounds (VOCs) emission inventory are assumed to be converted to SOA (Chin et al., 2002). Aerosols in RIEMS-Chem include sulfate, nitrate, ammonium, BC, OC, SOA, 5 bins of soil dust, and 5 bins of sea salt (Han et al., 2012).

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ISORROPIA (Nenes et al., 1998) is coupled to RIEMS-Chem to treat thermodynamic equilibrium process and to simulate inorganic aerosols. SOA production from primary anthropogenic and biogenic VOCs is calculated using a bulk aerosol yield method according to Lack et al. (2004). RegCCMS also used ISORROPIA to calculate inorganic aerosols (Wang et al., 2010). For implementation of aerosol effects, sulfate radiative properties were treated following Kiehl and Briegleb (1993). OC were assumed to have the same properties as sulfate, and the wavelength-dependent radiative properties of BC follows Jacobson (2001). AE6 aerosol (the sixth-generation CMAQ aerosol module) mechanism is coupled with WRF. Compared to previous version of CMAQ aerosol modules, AE6 improves SOA treatments, adds a new heterogeneous N_2O_5 hydrolysis parameterization and ads a new gas-to-particle mass transfer for coarse aerosols in sea-salt emissions (Yu et al., 2014). There are seven components including water soluble mass, water insoluble mass, elemental carbon, sea salt, water, diameters and standard deviations passed to WRF to directly change radiation calculations.

(4) Meteorological boundary and initial conditions: M1, M2, M5 and M7 use the National Centers for Environmental Prediction (NCEP) final analysis (FNL) data to drive the model; M3 and M4 use NASA MERRA reanalysis data and M6 uses NCEP-NCAR reanalysis 1 dataset.

(5) Soil dust: M1, M6 and M7 do not include soil dust calculation. M3 and M4 use GOCART dust module, and M2 uses a GOCART version that modified by AFWA (Air Force Weather Agency). M5 uses a dust module that described in Han et al. (2004).

(6) Mixing state: M6 assumes external mixing, while other models use internal mixing treatments.

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Many previous studies have underscored that the choice of gas phase mechanism and aerosol models are of great importance for simulating air pollutants (Knote et al., 2015). The different gas phase chemistry and aerosol modules used in the participating models are expected to yield notable differences in performances, which are shown later in section 4.

~~The horizontal model resolutions of these applications range from 15km to 60km (Table 1). Model vertical resolutions vary from 16 to 60 layers (Table 1) and the set model top pressures range from 100mb to 20mb.~~

~~Gas phase chemistry and aerosol modules are key components of chemical transport models. Although the WRF-Chem and NU-WRF models were applied at three institutes (PNU, UIOWA, and NASA), different gas phase chemistry and aerosol modules were used. At PNU (M1), the RACM-ESRL (Regional Atmospheric Chemistry Mechanism, Earth System Research Laboratory) gas phase chemistry coupled with MADE/VBS (Modal Aerosol Dynamics Model for Europe/Volatility Basis set) aerosol module was used. RACM was developed based on Regional Acid Deposition Model (RADM2) to simulate regional atmospheric chemistry (Stockwell et al., 1997) (including 237 reactions) and the rate coefficients were updated in RACM-ESRL version (Kim et al., 2009). MADE uses 3 log-normal modes (Aitken, accumulation, coarse) and simulates major aerosol compositions, including sulfate, ammonium, nitrate, sea-salt, black carbon (BC), and organic carbon (OC). In addition, the VBS method was implemented to simulate secondary organic aerosols (SOA). At the University of Iowa (M2), CBMZ (Carbon-Bond Mechanism version Z) gas phase chemistry coupled with an 8-bin MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) aerosol module was applied. CBMZ (Zaveri and Peters, 1999) extends the original CBM4 mechanism to function properly at larger spatial and longer timescales. The augmented CBMZ scheme includes 67~~

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~~species and 164 reactions. MOSAIC considers major aerosol species at urban, regional and global scales, including sulfate, nitrate, ammonium, sodium, chloride, EC, and other unspecified inorganic species (such as inert minerals, trace metals, and silica) (Zaveri et al. 2008). MOSAIC includes some aqueous reactions but no SOA formation. At NASA, the GOCART aerosol model (Chin et al., 2002) was coupled to RADM2 gas phase chemistry, and incorporated into the NU-WRF model (M3 and M4) to simulate major tropospheric aerosol species, including sulfate, BC, OC, dust, and sea-salt. In this aerosol model, 10% of organic compounds from the volatile organic compounds (VOCs) emission inventory are assumed to be converted to SOA (Chin et al., 2002). Both the RIEMS-Chem model from IAP (M5) and the RegCCMS model from NJU (M6) used CBM4 to calculate gas phase chemistry (Gery et al., 1989). The CBM4 version incorporated in RIEMS-Chem (M5) includes 37 species and 91 reactions,The version of CBM4 implemented in RegCCMS (M6) consists of 36 reactions (4 photolysis reactions) and 20 species (Wang et al., 2010).M7 applied SAPRC 99 coupled to the sixth-generation CMAQ aerosol module (AE6) to simulate gas phase chemistry and aerosol formation The SAPRC99 mechanism implanted within the CMAQ model has 88 species and 213 chemical reactions (Carter, 2000a,b).At NASA, the GOCART aerosol model (Chin et al., 2002) was coupled to RADM2 gas phase chemistry, and incorporated into the NU-WRF model (M3 and M4) to simulate major tropospheric aerosol species, including sulfate, BC, OC, dust, and sea-salt. In this aerosol model, 10% of organic compounds from the volatile organic compounds (VOCs) emission inventory are assumed to be converted to SOA (Chin et al., 2002).~~

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RIEMS-Chem (M5) includes 37 species and 91 reactions, and aerosols in RIEMS-Chem include sulfate, nitrate, ammonium, BC, OC, SOA, 5 bins of soil dust, and 5 bins of sea salt (Han et al., 2012). ISORROPIA (Nenes et al., 1998) is coupled to RIEMS-Chem to treat thermodynamic equilibrium process and to simulate inorganic aerosols. SOA production from primary anthropogenic and biogenic VOCs is calculated using a bulk aerosol yield method according to Lack et al. (2004). A lognormal size distribution is assumed for inorganic aerosols, BC, and OC, with median radius of 0.07 mm, 0.01 mm, and 0.02 mm, and geometric standard deviation of 2.0, 2.0, and 2.2, respectively. The schemes for soil dust deflation and sea salt generation were from Han et al. (2004), which used 5 size bins (0.1-1.0, 1.0-2.0, 2.0-4.0, 4.0-8.0, 8.0-20.0μm) to represent dust and sea salt size distribution. The refractive indices of aerosol components were mainly derived from the OPAC (Optical Properties of Aerosols and Clouds) database. Aerosol extinction coefficient as well as single scattering albedo and asymmetry factor are calculated by a Mie-theory based parameterization developed by Ghan and Zaveri (2007), which has a high computational efficiency with similar degree of accuracy compared with complete Mie code. An internal mixture of aerosols was assumed in this region of large emissions. A method known as kappa (k) parameterization (Petters and Kreidenweis, 2007) was adopted to represent the aerosol hygroscopic growth.

The version of CBM4 implemented in RegCCMS (M6) consists of 36 reactions (4 photolysis reactions) and 20 species (Wang et al., 2010). RegCCMS also used ISORROPIA to calculate inorganic aerosols (Wang et al., 2010). For implementation of aerosol effects, sulfate radiative properties were treated following Kiehl and Briegleb (1993), OC were assumed to have the same properties as sulfate, and the wavelength-dependent radiative properties of BC follows Jacobson (2001).

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2.2 Emissions

The accuracy of air quality modeling results highly depends on the quality and reliability of emission inventory. Accordingly, a new Asian emission inventory was developed for MICS-III by integrating state-of-the-art national/regional inventories to support this model inter-comparison study (Li et al., 2017). This is the major theme of MICS-ASIA III Topic 2. These emissions, along with biogenic emissions, biomass burning emissions, emissions from air and ship, and volcano emissions were used. This section offers some basic descriptions of these provided emissions.

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2.2.1 Anthropogenic emissions

The state-of-the-art anthropogenic emission inventory for Asia (MIX) was developed by incorporating five inventories, including the REAS inventory for Asia developed at the Japan National Institute for Environmental Studies (NIES), the MEIC inventory for China developed at

Tsinghua University, the high resolution ammonia (NH_3) emission inventory in China developed at Peking University, the Indian emission inventory developed at Argonne National Laboratory in the United States, and the CAPSS Korean emission inventory developed at Konkuk University (Li et al., 2017). This MIX inventory includes emissions for ten species, namely SO_2, nitrogen oxides (NO_x), carbon monoxide (CO), non-methane volatile organic compounds (NMVOC), NH_3, PM_{10}, $PM_{2.5}$, BC, OC, and carbon dioxide (CO_2). NMVOC are provided with CB-05 and SAPRC-99 speciation datasets. Emissions of these species were prepared for years 2008 and 2010 in monthly temporal resolution and 0.25 degree spatial resolution. Weekly/diurnal profiles were also provided. Five sectors were considered, namely industry, power generation, residential sources, transportation and agriculture. Figure 2 shows the spatial maps of these ten species for January 2010. Emissions of most of these species exhibit similar spatial patterns, with enhanced values in east China and lower values in north and south India. Emissions of NH_3 display a different spatial distribution, with pronounced values in India and lower values in north China (Figure 2). More detailed description of this emission inventory is documented in Li et al. (2017).

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2.2.2 Biogenic emissions

Terrestrial ecosystems generate ~~miscellaneous~~ various chemical species, including volatile and semi-volatile compounds, which play important roles in atmospheric chemistry and are the largest contributor to global annual flux of reactive volatile organic compounds (VOCs) (Guenther et al., 2006). For MICS-ASIA III, hourly biogenic emissions were provided for the entire year of 2010 using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.04 (Guenther et al., 2006). The variables that drive MEGAN include land cover information (plant function type, leaf area index) and weather condition, which includes solar

transmission, air temperature, humidity, wind speed, and soil moisture. In the preparation of MEGAN biogenic emissions, land cover information is taken from the NASA MODIS products, and weather condition are calculated using WRF simulations. Figure S1 shows biogenic emissions of some selected species (isoprene and HCHO) for January 2010. High biogenic emissions are found in south Asia during winter, including India, south China, and Southeast Asia, where solar radiation, air temperature and vegetation covers are relatively higher than in northern regions. As shown in Table 1, M1 and M5 use prescribed biogenic VOCs emissions, other models except M6 use internal calculation.

~~Some models used these emissions directly. Others internally calculated the biogenic emissions on-line with the model predicted meteorology using the MEGAN model.~~

2.2.3 Biomass burning emissions

Biomass burning in the tropics is a strong contributor to air pollutants, and extensive biomass burning in Asia, particularly Southeast Asia, exerts a great influence on air quality (Streets et al., 2003). For MICS-ASIA III, biomass burning emissions were processed by re-gridding the Global Fire Emissions Database version 3 (GFEDv3) (0.5 by 0.5 degree). GFED fire emissions are estimated through combining satellite-detected fire activity and vegetation productivity information. Carbon, dry matter, CO_2, CO, CH_4, hydrogen, nitrous oxide, NO_x, NMHC, OC, BC, $PM_{2.5}$, total particulate matter and SO_2 emissions are estimated in monthly temporal resolution. Figure S2 shows the gridded biomass burning emissions for January 2010. Biomass burning activity is highest in Cambodia and some areas of Myanmar and north of Thailand (Figure S2), and the peak emission season is spring. Although it has been concluded that biomass burning could significantly contribute to aerosol concentrations in China, the contribution is limited for

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Topic 3 study since the focused region is North China where biomass burning emissions are negligible during ~~cold~~ winter (Gao et al., 2016a).

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2.2.4 Volcanic SO_2 emissions

Volcanoes are important sources of various sulfur and halogen compounds, which play crucial roles in tropospheric and stratospheric chemistry. It is estimated that SO_2 emitted from volcanoes account for about 9% of the total worldwide annual SO_2 flux (Stoiber et al., 1987). The Asia-Pacific region is one of the most geologically unstable regions in the world where many active volcanoes are located. During MICS-ASIA Phase II, the volcano SO_2 emissions had already been provided for chemical transport models (Carmichael et al. 2008b). Volcano SO_2 emissions were provided, with a daily temporal resolution. In January, some volcanoes in Japan are very active, such as Miyakejima (139.53ºE, 34.08ºN, and 775m above sea level) and Sakurajima (130.65ºE, 31.59ºN, 1117m above sea level).

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2.2.5 Air and Ship emissions

Fuel burning in aircraft and ship engines produces greenhouse gases and air pollutants. The shipping and aircraft emissions used are based on HTAPv2 emission inventory (0.1 by 0.1 degree) for year 2010 (Janssens-Maenhout et al., 2015), provided on an annual basis. Aircraft emissions include three parts: landing and takeoff (LTO), climbing and descent (CDS), and cruise (CRS). Aircraft emission hot spots are mostly located in Japan, and Beijing, Yangtze River Delta (YRD) and Pearl River Delta (PRD) in China (Figure S3). East China Sea, sea around Japan and Singapore exhibit high shipping emissions due to active shipping transportation (Figure S3). It is estimated that international shipping contributed about 10% to

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the global SO_2 emissions, and together with aviation contribute more than 10% of global NO_x emissions (Janssens-Maenhout et al., 2015).

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2.3 Boundary conditions

To predict more realistic spatial and temporal variations of air pollutants, boundary conditions from global chemical transport models are necessary to drive regional chemical transport models (Carmichael et al., 2008b). Simulations of ~~three~~ two global chemical transport models (i.e., ~~CHASER,~~ GEOS-Chem and MOZART) were ~~provided~~ used as boundary conditions for MICS-ASIA III. ~~CHASER was developed in Japan to simulate the O_3-HO_x-NO_x-CH_4-CO photochemical system and its effects on climate (Sudo et al., 2002).~~ GEOS-Chem was developed in the USA to simulate tropospheric chemistry driven by assimilated meteorology (Bey et al., 2001). T~~In addition, t~~he National Center for Atmospheric Research (NCAR) also provides global simulations of atmospheric chemistry (MOZART model) and an interface to convert them to WRF-Chem boundary conditions (Emmons et al., 2010), and NASA provides global aerosol distributions using the global GOCART chemistry model (Chin et al., 2002). GEOS-Chem was run with 2.5ºx2º resolution and 47 vertical layers ~~and CHASER model was run with 2.8ºx2.8º and 32 vertical layers. 3-hourly-average fields of gaseous and aerosols were distributed to all participants~~. The MOZART-4 simulations were ~~also~~ configured at the horizontal resolution of 2.8ºx2.8º, and ~~but~~ with 28 vertical levels. NASA GOCART was configured at the same resolution as GEOS-5 meteorology (1.25ºx1º). As listed in Table 1, M1 used climatological data from the NOAA Aeronomy Lab Regional Oxidant Model (NALROM), while M2 used boundary conditions from the MOZART-4 (provided from the NCAR website). M3 and M4 used MOZART-4 as boundary conditions for gases and used GOCART as boundary conditions for aerosols. M6 also used fixed climatology boundary conditions, and M5 and M7 used GEOS-

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Chem outputs as boundary conditions. The spatial distribution of near surface concentrations of major gases and aerosols from both GEOS-Chem and MOZART are shown in Figure S4. Even though the same global model is used as boundary conditions, the treatments of inputs might differ in details, which might lead to ~~considerable~~ dissimilarities. In MICS-ASIA II, Holloway et al. (2008) discussed the impacts of uncertainties in global models on regional air quality simulations.

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2.4 Observation data

Historically, the lack of reliable air quality measurements in Asia has been a bottleneck in understanding air quality and constraining air quality modeling in Asia. Beginning with MICS-ASIA II, observational data from Acid Deposition Monitoring Network in East Asia (EANET) has been used to evaluate model performance. EANET was launched in 1998 to address acid deposition problems in East Asia, following the model of the Cooperative Program for Monitoring and Evaluation of the Long-range Transmission of Air pollutants in Europe (EMEP). As of 2010, there are 54 wet deposition sites and 46 dry deposition sites in 13 participating countries. Quality assurance and quality control measures are implemented at the national levels and in the Inter-laboratory Comparison Project schemes to guarantee high quality dataset. EANET supported current activities of MICS-ASIA III, and provided measurements in 2010 to all modeling groups. More information about EANET dataset can be found in http://www.eanet.asia/.

In addition to EANET data, measurements of air pollutants and aerosol optical depth (AOD) collected at the Campaign on Atmospheric Aerosol Research network of China (CARE-China) (Xin et al., 2015) network were also used. Previous successful networks in Europe and the

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United States underscored the importance of building comprehensive observational networks of aerosols in China to get better understanding of the physical, chemical and optical properties of atmospheric aerosols across China. As the first comprehensive attempt in China, CARE-China was launched in 2011 by Chinese Academy of Sciences (CAS) (Xin et al., 2015). Before launching this campaign, CAS had already been measuring air pollutants and AOD at some CARE-China sites. Table 2 summaries the locations and characteristics of the CARE-China measurements for January 2010. Air quality measurements include concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, NO, CO, O_3.

In addition, AOD from Aerosol Robotic Network (AERONET) (https://aeronet.gsfc.nasa.gov/) and operational meteorological measurements (near surface temperature, humidity, wind speed and downward shortwave radiation) in China and atmospheric sounding data in Beijing were used. AERONET provides long-term, continuous, readily accessible and globally distributed database of spectral AOD, inversion products and precipitable water. AOD data are calculated for three quality levels: Level 1.0 (unscreened), Level 1.5 (cloud screened), and Level 2.0 (cloud screened and quality assured) (Holben et al., 1998). The locations and characteristics of the AERONET measurements are also summarized in Table 2. In-situ measurements of meteorological data from standard stations in China are operated by China Meteorological Administration (CMA) and different levels of data, including daily, monthly, and annually, are open to the public (http://data.cma.cn/en). The locations of all used observational sites are marked in Figure ~~S4~~S5, Figure ~~S5~~ S6 and Figure ~~S6~~S7.

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The meteorology measurements (locations are shown in Figure ~~S4~~S5) were averaged and compared with model results that averaged across those locations. The radiation measurements were averaged and compared against model results in North China and South China (locations

are shown in Figure ~~S5~~S6), separately. The CARE-China, AERONET and EANET measurements (locations are shown in Figure ~~S5~~ S6 and ~~S6~~S7) were compared against model results site by site, and model ensemble mean values were made by averaging all model results.

2.5 Analysis methodology

All groups participating in Topic 3 were requested to simulate meteorology, air quality, radiative forcing and effects of aerosols over the Beijing-Tianjin-Hebei region of east China during two periods: January 2010 and January 2013. ~~Simulations were designed to evaluate radiative and microphysical feedbacks, together and separately, relative to simulations without feedbacks.~~ Each group was requested to submit the following fields from their simulations.

(1) hourly mean meteorology:

(a) air temperature and water vapor mixing ratio at 2m above ground (T2, Q2), wind speed at 10m above groud (WS10), and shortwave radiation flux (Wm-2) at the surface;

(b) above variables (except shortwave radiation flux) at 1km and 3km above ground.

(2) hourly mean concentrations:

(a) SO_2, NO_x, CO, O3, $PM_{2.5}$, PM_{10} and sulfate, nitrate, ammonium, BC, OC and dust in $PM_{2.5}$;

(b) above variables at 1km and 3km above ground.

(3) hourly mean AOD, aerosol direct radiative forcings at the surface, top of the atmosphere (TOA) and inside the atmosphere (single scattering albedo is an option for participants).

(4) Hourly mean integrated liquid water, cloud optical depth.

(5) Changes in T2, Q2, WS10 and $PM_{2.5}$ concentrations at the surface due to both direct and indirect aerosol's effects.

We calculated multiple model evaluation metrics, including correlation coefficient (r), root mean square error (RMSE), mean bias error (MBE), normalized mean bias (NMB), mean fractional bias (MFB) and mean fractional error (MFE). The equations are presented in supplemental information.

3 General description of meteorology and haze during the study period~~s~~

Winter haze events ~~are frequently happening~~frequently happen in east China, which is partially due to the stagnant weather conditions in winter. Here we present general descriptions of the meteorological conditions during ~~the selected two~~ January ~~months~~ 2010 using the NCEP/NCAR reanalysis products. Figure S8~~3~~ (a~~, b~~) displays the monthly mean T2 (temperature at 2m) and W10 (wind speeds at 10m) ~~for January 2010 and January 2013, respectively~~. ~~For both periods,~~ WS10 were very weak in eastern and central China regions~~.~~, while lower T2 in Mongolia region ~~was relatively higher for January 2013. Historical analyses have shown that cold conditions are usually associated with strengthened Siberian High (Gong and Ho, 2002), and relatively higher T2 and more weakened~~was associated with Siberian High. As shown in Figure S8 (b), ~~(Figure 3 (c, d)) during January 2013 led to weaker winter monsoon winds and higher pollution levels. The relatively weaker Siberian High during January 2013 compared to January 2010 is also shown in the sea level pressures (Figure 3 (c, d)). The~~ the Siberian High center was about ~~1037mb during January 2013, lower than that (~~1040mb, ~~) during January 2010. Figure 3 (c, d) show that~~and there was no significant precipitation in North China and heavy rainfall only occurred in

Southeast Asia regions. During ~~cold~~ winters, northern China burns coal for heating, generating more emissions. Under stagnant weather conditions, haze episodes are easily triggered. ~~It was reported that January 2013 was the haziest month in the past 60 years in Beijing, and instantaneous $PM_{2.5}$ concentration exceeded 1000μg/m^3 in some areas in Beijing. Winter haze also happened from 16 to 19 January in 2010.~~ High concentrations of aerosols during ~~these two study periods~~this month provide great opportunity to study aerosol-radiation-weather interactions.

4 Results and discussions

In this section, we present some major features of model performances in meteorological and chemical variables for the January 2010 period. Detail analysis of feedbacks and radiative forcing are presented in MICS-ASIA III companion papers. Heavy haze occurred over broad regions of East China in January 2010. The plots of observed meteorological variables and $PM_{2.5}$ in Beijing show the general situation (Figure ~~4~~3). Elevated $PM_{2.5}$ occurred during three periods separated in time by roughly one week (January 8, 16 and 26). The major event occurred during January 15-21. The events occurred during periods of low wind speeds, and increasing temperature and relative humidity. The high $PM_{2.5}$ concentrations during January 15-21 also greatly reduce the downward shortwave radiation. Below we evaluate how well the models predict these features.

4.1 Evaluation of meteorological variables

Air quality is affected by not only emissions, but also meteorological conditions. Meteorology affects air quality through altering emissions, chemical reactions, transport, turbulent mixing, and deposition processes (Gao et al., ~~2016b~~2016c). Thus, it is important to assess how well these participating models reproduced meteorological variables. The predicted temperature at 2m high (T2), water vapor mixing ratio at 2m (Q2), wind speed at 10m high (WS10) and daily maximum downward shortwave radiation (SWDOWN) were evaluated against near surface observations at the CMA sites.

Figure ~~5~~ 4 (a-c) shows the comparisons between simulated and observed daily mean T2, Q2 and WS10 averaged over stations in East China (locations are shown in Figure ~~S4~~S5) during January 2010, along with multi-model ensemble mean and observation standard deviation. The calculated correlation coefficients between models and observations are also shown in Figure 5 and other calculated model evaluation metrics are summarized in Table 3. In general, the simulated magnitudes and temporal variations of T2 and Q2 show high order of consistencies with observations, with correlation coefficients ranging from 0.88 to 1. For T2, models tend to have a cool bias; M1 and M2 have the lowest RMSE (0.64 and 0.68), lowest MBE (-0.19 and -0.60) and lowest NMB (-0.07% and -0.22%) values (Table 3). For Q2, most models tend to slightly overestimate; M1 and M2 have the best performance, with the lowest RMSE (0.14 and 0.10), lowest MBE (0.02 and -0.01), and lowest NMB (0.84% and -0.55%) values (Table 3).

Simulated WS10 exhibit larger diversity of results. All models tend to overestimate WS10, with MBE ranging from 0.15m/s to 2.37m/s. Overestimating wind speeds under low wind conditions is a common problem of current weather forecasting models, and many factors, including errors in terrain data and reanalysis data, relatively low horizontal and vertical model resolutions, as well as poorly parameterized urban surface effect, contribute to these overestimations. From the

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554 calculated RMSE, MBE, and NMB listed in Table 3, M2, M5 and M7 show better skills in
555 capturing WS10. In addition, the multi-model ensemble mean show the lowest RMSE for Q2,
556 and also better skills than most models for T2 and WS10. The correlation coefficients between
557 multi-model ensemble mean and observations are 0.99, 0.99 and 0.98 for T2, Q2 and WS10,
558 respectively.

559 The accuracy of radiation predictions is of great significance in evaluating aerosol-radiation-
560 weather interactions. We evaluated simulated daily maximum SWDOWN averaged over sites in
561 northern China and southern China separately in January 2010 against observations. The
562 locations of the radiation sites are shown in Figure ~~S5~~S6. As shown in Figure ~~5~~ 4 (d), over
563 stations in northern China, all models except M6 and M7 reproduce daily maximum SWDOWN
564 well, with correlation coefficients ranging from 0.72 to 0.94. The poor performance of M6 in
565 North China is caused by largely overpredicted liquid water path (LWP) over North China
566 (Figure S9).

567 SWDOWN decreases under conditions of high PM, as shown for example on January 9 and 15-
568 21. This is one of the important reasons for coupled air quality and meteorology modeling, as
569 they can account for this effect of aerosols. It is worth noting that most models predict higher
570 daily maximum SWDOWN compared to observations when severe haze happened in the North
571 China Plain (16-19 January 2010), indicating aerosol effects on radiation might be
572 underestimated. Clouds are also important to alter radiation. To exclude its impacts on the
573 radiation shown here, we calculated the reduction ratio of radiation due to clouds using radiation
574 prediction for clear sky and with clouds from M2 (shown in Figure S10). During the severe haze
575 period (16-19 January 2010), the averaged reduction fraction is 5.9% in north China and 4.2% in
576 south China, suggesting the relatively lower radiation during this period shown in Figure 4(d) is

mainly caused by aerosols, while the lowest radiation on 20 January was caused by clouds (Figure 4(d) and Figure S10). Over southern China sites (Figure ~~5e~~4e), M6 and M7 show a better consistence with observations than over northern China sites. According to the calculated RMSE listed in Table 3, M3 and multi-model ensemble mean exhibit relatively better performance in capturing the observed time series of daily maximum SWDOWN in both northern China and southern China.

The above comparisons show that T2 and Q2 are reproduced well by the participating models, and WS10 is overestimated by all models. Emery et al. (2001) proposed that excellent model performance would be classified as wind speed RMSE smaller than 2 m/s, and wind speed bias smaller than 0.5 m/s. Based on the calculated RMSE and MBE of WS10 shown in Table 3, RMSE values from all models match the proposed RMSE threshold but MBE values are higher than 0.5 m/s. The vertical distributions of temperature, water vapor mixing ratio and wind speeds were also validated against atmospheric sounding data in Beijing at 1km and 3km (Figure ~~S7~~S11, averaged at 00:00 and 12:00 UTC) (http://weather.uwyo.edu/upperair/sounding.html). The magnitudes of temperature, water vapor mixing ratio and wind speeds from different models are generally consistent with each other at 1km and 3km, but variations are larger near the surface.

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4.2 Evaluation of air pollutants

Figure ~~6~~ 5 displays the daily averaged predicted and observed SO_2, NO_x, CO, O_3, $PM_{2.5}$, and PM_{10} at the Beijing station, along with the observation standard deviation (locations are shown in Figure ~~S6~~S7). Comparisons for the Tianjin, Shijiazhuang and Xianghe sites are shown in Figure ~~S8~~S12-~~S10~~S14. M6 only provided SO_2, NO_x concentrations, so it is not ~~only~~ shown in the plots

of CO, O_3, $PM_{2.5}$, and PM_{10}. The observed and predicted primary pollutants and $PM_{2.5}$ and PM_{10} show the same monthly variations with elevated values at roughly weekly intervals, with the largest event occurring during January 15-21. For example, as shown in the comparisons of SO_2 concentration, the temporal variations are reproduced well by all the models, but peak values are overestimated or underestimated by some models. Based on the calculated MBE values shown in Table 4, all models except M2 tend to underestimate SO_2 in ~~Beijing~~the CARE-China sites. M1 shows the highest correlation (0.90) with SO_2 observations in the Beijing site, and most other models show similar good correlations. The multi-model ensemble mean shows a better agreement with observations with a higher correlation of 0.92, and it falls within the range shown with standard deviation error bar. In general, the predictions for NO_x capture the main features in the observations, with slightly less skill than for the SO_2 prediction. The calculated correlation coefficients for NO_x from different models are close to each other, ranging from 0.63 to 0.88. M2 and M5 predict higher NO_x concentrations than observations and other models (MBE in Table 4). All models overestimate NO_x concentration in Shijiazhuang (Figure ~~S8~~S14), suggesting NO_x emissions in Shijiazhuang might be overestimated in the MIX emission inventory. All models produce similar CO predictions~~All models are consistent with each in CO predictions~~.

$PM_{2.5}$ concentrations are well modelled, with high correlation coefficients ranging from 0.87 to 0.90 in Beijing, from 0.83 to 0.93 in Tianjin, and from 0.74 to 0.91 in Xianghe. The correlation coefficient of the multi-model ensemble mean for $PM_{2.5}$ reaches 0.94 (Table 4), better than any individual model. The performances of all participating models in reproducing PM_{10} variations are not as good as reproducing $PM_{2.5}$. M1 and M2 overestimate PM_{10} concentrations, and other

models underestimate PM_{10} concentrations (MBE in Table 4). These biases are probably related to different treatments of primary aerosols and anthropogenic dust in the models.

The models showed the poorest skill in predicting ozone. All models exhibit different performances in simulating ozone concentrations, and the correlation coefficients between models and observations can reach negative values (Figure ~~S8~~S12). M3 and M4 tend to overestimate ozone concentrations, M2 slightly overestimates it, and M1, M5, and M7 slightly underestimate it (MBE in Table 4). According to the calculated RMSE in Table 4, M1 and M7 shows relatively better performance in modeling ozone variations. Although WRF-Chem and NU-WRF models were applied at three institutions, different gas phase chemistry schemes were used, which leads to these diversities among predicted ozone concentrations. The impacts of gas phase chemical mechanisms on ozone simulations have been investigated in ~~Zhang~~ Knote et al. (2015~~2~~). ~~; but under high photochemical conditions. The results presented here winter conditions with slower photochemistry in general and where hazy conditions further reduce photochemistry through diming effects.~~

Figure ~~7~~ 6 shows the comparisons between modeled and observed ground level daily averaged concentrations of SO_2, NO_x, O_3 and PM_{10} during January 2010 at the Rishiri site in Japan from EANET. The locations of ~~used~~ EANET sites are marked in Figure ~~S6~~S7. Comparisons at other EANET sites are shown in Figure ~~S11~~S15-~~S14~~S18. The models are able to predict the major features in the observations. For example, low values of most pollutants are observed (and predicted) during the first half of the month, followed by elevated values, which peak on January 21. For SO_2, most models show similar capability in producing the temporal variations in observations with slight underestimation (MBE in Table 5). According to the calculated RMSE averaged over all the EANET sites, M2 and the multi-model ensemble mean performed the best. For NO_x, the multi-model ensemble mean shows lower RMSE than any individual model (Table 5). Similar to the comparisons over CARE-China sites, large discrepancies exist in ozone predictions, but the model ensemble mean still shows the lowest RMSE for ozone predictions.

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PM_{10} concentrations are largely underestimated by M1 (largest negative MBE: -21.03ug/m^3) and overestimated by M5 (highest positive MBE: 3.77ug/m^3) (Table 5), which could also be related to differences in the way sea-salt emissions are treated in the various models. Spatial distributions of monthly near surface concentrations of SO_2, NO_x, O_3 and CO for January 2010 from all participating models are shown in Figure S19. The aerosol spatial distributions are discussed in the following section.

4.3 $PM_{2.5}$ and $PM_{2.5}$ chemical composition distribution

~~Haze pollution is characterized by high loadings of $PM_{2.5}$, thus accurately predicting $PM_{2.5}$ and its chemical compositions are crucial to understand haze pollution and to provide insightful implications for controlling haze in China. The accuracy of predicting $PM_{2.5}$ chemical composition is also of great importance in estimating aerosol-radiation interactions. For example, black carbon absorbs shortwave radiation, whereas sulfate and organic carbon mostly scatter radiation.~~ Due to different implementations of chemical reactions in the models, predicted $PM_{2.5}$ chemical compositions from participating models differ largely. Figure ~~8~~ 7 and Figure ~~9~~ 8 show the predicted monthly mean concentrations of sulfate, nitrate, ammonium, BC and OC in $PM_{2.5}$ from all participating models for January 2010.

M1, M2, M3, M4 and M7 all predict quite low sulfate concentrations in east China, but with considerably enhanced sulfate in southwest areas of China and west areas of India. M5 and M6 show~~s~~ similar spatial patterns of sulfate except that M6 produces higher concentrations. The chemical production of sulfate is mainly from gas-phase oxidation of SO_2 by OH radicals and aqueous-phase pathways in cloud water. In cloud water, dissolved SO_2 can be oxidized by O_3,

H_2O_2, Fe(III), Mn(II), and NO_2 (Seinfeld and Pandis, 2016). Most chemical transport models have included the above gas phase oxidation of SO_2 by OH and oxidation of SO_2 by O_3 and H_2O_2 in aqueous phase. Under hazy conditions, radiation is largely reduced due to aerosol dimming effects, and sulfate formation from gas phase and aqueous phase oxidation processes are slowed down, which tend to reduce sulfate concentration. However, field observations exhibit an increase in sulfate concentration during haze episode (Zheng et al., 2015~~),~~). ~~and~~ Cheng et al. (2016) proposed that the reactive nitrogen chemistry in aerosol water could contribute significantly to the sulfate increase due to enhanced sulfate production rates of NO_2 reaction pathway under high aerosol pH and elevated NO_2 concentrations in the North China Plain (NCP) ~~during haze periods~~. Wang et al. (2016) also pointed out the aqueous oxidation of SO_2 by NO_2 is key to efficient sulfate formation on fine aerosols with high relative humidity and NH_3 neutralization or under cloudy conditions. Besides, Zheng et al. (2015) suggested that heterogeneous chemistry on primary aerosols could play an important role in sulfate production and lead to increasing sulfate simulation during haze episodes. X. Huang et al. (2014) found including natural and anthropogenic mineral aerosols can enhance sulfate production through aqueous-phase oxidation of dissolved SO_2 by O_3, NO_2, H_2O_2 and transition metal. Gao et al. (2016), Wang et al. (2014), and Zhang et al. (2015) also emphasized the importance of multiphase oxidation in winter sulfate production. However, ~~T~~these ~~above aqueous and heterogeneous~~ processes are currently not incorporated in the participating models for this study, which might be responsible for the apparent under-predictions of sulfate concentration (Figure ~~10~~9). M5 ~~also~~ incorporated heterogeneous chemical reactions on aerosol surface (Li and Han, 2010), which enhances total sulfate production.

M1 and M5 predict relatively small nitrate and ammonium concentrations; while M2, M6 and M7 produce~~s~~ similar magnitudes and spatial patterns of nitrate. Nitrate formation involves both

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daytime and nighttime chemistry. During daytime, NO_2 can be oxidized by OH to form nitric acid (HNO_3), and by ozone to form NO_3. HNO_3 is easily removed by dry or wet deposition, but NO_3 is easily photolyzed back to NO_2. During nighttime, NO_3 is the major oxidant, which oxides NO_2 to form dinitrogen pentoxide (N_2O_5). Homogenous reaction of N_2O_5 with water vapor is possible but very slow while heterogeneous uptake of N_2O_5 onto aerosol particles has been identified as a major sink of N_2O_5 and an important contributor to particulate nitrate (Kim et al., 2014). The MOSAIC aerosol module (Zaveri et al., 2008) coupled with CBMZ gas phase chemistry in WRF-Chem already includes heterogeneous uptake of N_2O_5 since version v3.5.1 (Archer-Nicholls et al., 2014), which is the version used by M2, leading to the high production of nitrate. An et al. (2013) incorporated photoexcited nitrogen dioxide molecules, heterogeneous reactions on aerosol surfaces, and direct nitrous acid (HONO) emissions into the WRF-Chem model and found these additional HONO sources can improve simulations of HONO and nitrate in north China. M7 also predict high nitrate concentrations (N_2O_5 and NO_2 gases react with liquid water, Zheng et al., 2015), and the predicted lower nitrate concentrations from other models are probably due to missing aqueous phase and heterogeneous chemistry, or the implementations of different gas phase oxidation in these models. Many studies have been conducted regarding sulfate formation issues. Nitrate also account for a large mass fraction in $PM_{2.5}$ during winter haze events in north China, yet less attention was attracted to fully understand its formation. It is worth furtherly digging into the details about how different processes contribute to high nitrate concentrations in future studies. M3 and M4 do not include the explicit nitrate and ammonium treatment but ammonium is implicitly considered in total $PM_{2.5}$ mass estimate.

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The predicted ammonium concentrations are associated with the amounts of sulfate and nitrate, as shown by its similar spatial distribution to sulfate and nitrate. NH_3 neutralizes H_2SO_4 and HNO_3 to form aerosol, so its amount can affect the formation of sulfate, nitrate and ammonium. Since the same emission inventory was used, the amount of ammonia available for neutralizing will not vary greatly among these models. Thus, the rates of H_2SO_4 and HNO_3 production determines the amounts of ammonium. For example, the produced ammonium concentrations are small in M1, similar to their sulfate and nitrate productions. High ammonium concentrations are predicted from M6, due to high productions of nitrate and sulfate (Figure ~~8~~7).

The spatial distributions and magnitudes of predicted BC from all participating models are similar to each other as BC is a primary pollutant whose mass as BC is not impacted by chemical reactions. The concentrations of BC in the atmosphere are mainly influenced by PBL mixing and diffusion, aging, deposition (dry deposition and wet scavenging) and advection. Predicted BC from M2 and M7 are higher than from other models, which might be caused by ~~different treatments of emission inventory (for example, how to distribute emissions to different vertical layers), horizontal grid interpolation, and/or different parameterizations for vertical diffusion,~~the treatment of aging and~~,~~ deposition (dry deposition and wet scavenging) ~~and advection~~processes. For example, in the GOCART aerosol model (M3 and M4), 80% of BC are assumed to be hydrophobic and then undergo aging to become hydrophilic in an e-folding time of 1.2 days. Hydrophilic aerosols will go through wet deposition. But in other models like M2 and M7, BC is assumed to be hydrophobic, thus the wet removal is less.

The disparity among predicted OC concentrations is mainly associated with the different treatments of SOA production, given the POC prediction is generally consistent among models using the same emission inventory. The predicted OC concentrations from M1, M2, and M7 are

close to each other. M1 uses SORGAM (Secondary Organic Aerosol Model) to simulate SOA, but M2 and M6 did not include any SOA formation mechanism. The similar magnitudes of OC from M1 ~~and M2~~ suggests that SORGAM in M1 does not produce appreciable amounts of SOA, which is consistent with the findings in Gao et al. (2016a). Although SOA formation is implemented in M5, the production is relatively weak compared to M3 and M4. In the atmosphere, SOA is mainly formed from the condensation of semi-volatile VOCs from oxidation of primary VOCs. An empirical 2-produt model (Odum et al., 1996) is often used to simulate SOA formation, but this method was reported to significantly underestimate measured SOA mass concentrations (Heald et al., 2008). Later, the volatility basis-set approach (Donahue et al., 2006) was developed to represent more realistically the wide range of volatility of organic compounds and more complex processes, and it was found to increase SOA production and to reduce observation-simulation biases in many regions with high emissions (Tsimpidi et al., 2010) including east China (Han et al., 2016). It was also suggested that primary organic aerosols (POA) are semi-volatile and can evaporate to become SOA precursors, which promotes the understanding and improvements of SOA modeling (~~Li et al., 2011~~Kanakidou et al., 2005). In M5, the SOA production is calculated using a bulk yield method via Lack et al. (2004), in which the amount of SOA able to be produced from a unit of reacted VOC from anthropogenic and biogenic origins are used to represent SOA yields. However, the SOA concentration is highly dependent on the yield data. During haze episodes, photochemistry is reduced due to the aerosol dimming effect, thus aqueous reaction processes on aerosol water and cloud/fog water could become much more important in producing SOA. ~~as suggested in Cheng et al. (2016).~~ R. Huang et al. (2014) also suggested that low temperature does not significantly reduce SOA formation rates of biomass burning emissions. Most models over-simplified SOA formation.

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~~The missing representation of such process in the participating models may partly account for the low values in the simulated SOA.~~ In M3 and M4, SOA is treated by assuming that 10% of VOCs from terrestrial source are converted to OC (Chin et al., 2002), and these models produce high OC concentrations, with a major contribution from SOA. The 10% yield rate could be unrealistically high during hazy days because solar radiation was much reduced. Zhao et al. (2015) comprehensively assessed the effect of organic aerosol aging and intermediate-volatile emissions on OA formation and confirmed their significant roles. All these results suggest more complicated SOA scheme are needed to improve organic aerosol simulations during haze events.

The different predictions of $PM_{2.5}$ chemical components lead to differences in $PM_{2.5}$ ~~co~~and PM_{10} concentrations for January 2010, which are shown in the last row of Figure ~~9~~8. Although spatial distributions of $PM_{2.5}$ from these models are similar, the underlying causes are different. M2, M3 and M5 simulated higher $PM_{2.5}$ levels in deserts of west China, which are contributed by wind-blown dust ~~deflation~~. M1 and M7 fail to produce high $PM_{2.5}$ concentrations in the deserts of west China, due to omission of dust emissions. M4 presented results in a smaller domain excluding west China. The enhanced $PM_{2.5}$ concentrations in Central China from M2 and M7 are caused by large nitrate production, as shown in Figure ~~8~~7.

The differences in the predictions of aerosols composition discussed above can be seen clearly in the comparisons at the Beijing site on 13-23 January when a haze event occurred in the NCP (Figure ~~10~~9). Also shown are the observed values. Most models fail to produce the observed high sulfate concentrations. Only the sulfate predictions from M5 are close to the observed values. Sulfate is much lower than observed for all other models, except M6 which is too high. M2 and M7 predict reasonable nitrate concentrations. M3 and M4 overpredict OC during the haze period, but other models underpredict OC concentrations.~~Only M5 prediction is close to~~

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~~observation, and M6 predicts higher sulfate level. M2 and M7 predict reasonable nitrate concentrations. M3 and M4 largely overpredict OC during haze period, but other models tend to underpredict OC concentrations~~.

Figure ~~11~~ 10 and 11~~2~~ show the ensemble mean monthly averaged near-surface $PM_{2.5}$, $PM_{2.5}$ composition, along with the spatial distribution of the coefficient of variation. The coefficient of variation is defined as the standard deviation divided by the average (Carmichael et al., 2008b), and larger values indicate lower consistency among models. Mean concentrations of $PM_{2.5}$ and $PM_{2.5}$ chemical compositions are high in Sichuan Basin and east China. High coefficient of variation are shown in North China for sulfate, and in most areas for nitrate and OC. The diversity in predictions of these species are caused by complexity of secondary formation and different treatments in models as discussed earlier. Higher consistency is shown for model BC with coefficient of variations less than 0.3 in most areas (Figure 10 ~~(~~h)). Coefficient of variations for $PM_{2.5}$ are also low in North China region, which is consistent with good performance of $PM_{2.5}$ predictions shown in above comparisons. However, the coefficient of variation can reach above 1.6 in northwestern regions, partially due to discrepancies in dust predictions.

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4.4 Evaluation of AOD

AOD is an indication of aerosol pollution, which tells us how much sunlight is blocked from reaching the surface by suspended aerosols. We used the measurements of AOD at AERONET and CARE-China sites to evaluate how participating models perform in simulating AOD. ~~In WRF-Chem, AOD is usually calculated at 300, 400, 600 and 999nm, which can be converted to AOD at other wavelengths based on Angstrom exponent relation (Schuster et al., 2006).~~ The submitted AOD from all models except M6 are at 550nm, and AOD from M6 are at 495nm. We

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used Angstrom exponent relation (Schuster et al., 2006) to convert AOD from M6 at 495nm to 550nm, and all used AERONET and CARE-China AOD data to 550nm. The locations of AERONET and CARE-China AOD measurement sites are shown in Figure S5. Daytime mean AOD are calculated in pairwise manner and the comparisons and performance statistics are shown in Figure ~~13~~12, ~~14~~13, and Table 6. On some days, data are missing because AOD cannot be retrieved under serious pollution and cloudy conditions (Gao et al., 2016a). On days with data, the variations of AOD are captured well by all models. However, large disparities exist among models in the simulated peak AOD values (factor of 2) at monitoring stations during the severe haze episode on 15-20 January 2010 (Figure ~~13~~ 12 and Figure ~~14~~13). The participating models exhibit various skill in simulating AOD temporal variation at different sites.

At CARE-China sites, M7 produces the best correlation coefficient R (0.83) among models at Baoding and Beijing forest sites, M2 produces the highest R (0.86) at Cangzhou site, whereas M5 shows the highest R (0.93) at the Beijing city site. At AERONET sites, M7 shows the highest R (0.81) at Beijing, whereas M2 and M5 produce R as high as 0.91 at Xianghe site, which is about 60km southeast of downtown Beijing. In terms of AOD magnitude, ~~it's~~ it is interesting to note that during the severest haze days around 19 January 2010, M2 consistently simulates the highest AOD among models, followed by M5 and M7~~,~~. ~~with the lowest AOD from~~ M6 simulates the lowest, and other models in the middle at the sites (Baoding, Beijing City, Beijing Forest, Cangzhou, Beijing, Xianghe) in the north China plain (NCP). ~~It is important to explore the causes for the disparities in AOD predicitons.~~

~~AOD is calculated as the vertical integration of extinction coefficient, which is a function of particle mass extinction efficiency (extinction cross section) and mass concentration. The extinction efficiency is determined by particle size, refractive index and wave length. Aerosol size~~

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~~can grow bigger as ambient relative humidity increases, which is known as aerosol hygroscopic growth. The overall extinction coefficient of all aerosols also depends on mixing state among aerosols. Therefore, AOD is closely related a series of affecting factors from both aerosol physical properties, mass concentration and meteorological conditions.~~

In M1, M5, ~~M6~~ and M7, particle size distribution is described by a lognormal function with a geometric mean radius and a geometric standard deviation basically based on OPAC (Optical properties of aerosols and clouds) database (Hess et al. 1998). In M3 and M4, sulfate, BC and OC are parameterized in bulk mode, and a sectional scheme is used for sea-salt and dust aerosols. M2 uses an 8 bins sectional aerosol scheme with size sections ranging from 39nm to 10μm. The refractive index of various aerosol components in the models are mainly taken from d'Almeida et al. (1991) or OPAC database. All models except M6 use a kappa (κ) parameterization (Petters and Kreidenweis, 2007), in which the aerosol hygroscopicity κ largely varies among different aerosol chemical components, such as κ=0 for black carbon, and κ>0.6 for inorganic aerosols, but the prescribed κ values could be different in the above models. M6 uses a different hygroscopic growth scheme following Kiehl and Briegleb (1993). WRF-Chem models assume internally mixing among aerosols within each mode (or size bin) and externally mixing between modes (or size bins), M5 assumes inorganic and carbonaceous aerosols are internally mixed and externally mixed with soil dust and sea-salt. M6 uses an external mixture assumption among aerosols except for hydrophilic BC, which is internally mixed with other aerosols in a core-shell way.

We first look at the mass concentrations of different aerosol components because of their important roles in determining optical properties. The observed total inorganic aerosol concentration in Beijing on 19 January 2010 is about 130μg/m^3, with sulfate and nitrate being about 50 and 65μg/m^3,

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respectively (Figure ~~10~~9). The models generally predict a much lower sulfate concentration except that the prediction from M5, which is close to observations, and M6, which shows an overprediction. Most models predict lower nitrate concentration, in contrast to the overprediction by M2. In terms of inorganic aerosols, which have a similar optical properties, the total concentration (the sum of sulfate, nitrate and ammonium) from M2 (175μg/m^3) is higher than observation and other models, and this can explain the largest simulated AOD by M2. M6 simulates a similar level of inorganic aerosols to M2, but the simulated AOD is lower than other models, which could be due to a weak hygroscopicity or lower simulated RH ~~simulation~~ (see Figure ~~S14~~S20). For example, high RH on January 19 are captured by M2 and M6, but underpredicted by M6 (Figure S14a). Although M3 and M4 largely overpredict OC concentrations, their simulated AOD are lower than M1 and M5 because their simulated inorganic aerosol concentrations are much lower and OC has a smaller (mass) extinction coefficient than inorganic aerosols. M1 predicts about three times larger BC concentration than the observations, although the mass extinction coefficient of BC is even larger than inorganic aerosols, the mass concentration and hygroscopicity of BC are much smaller and weaker than that of inorganic aerosols, leading to relatively lower AOD from M1 simulation. M5 and M7 predict a similar level of inorganic aerosol concentrations (80~90μg/m^3) and use a similar hygroscopic growth scheme, and this can help explain their consistency in the simulated AOD magnitude.

As listed in Table 1, only M6 uses external mixing for aerosols, and internal mixing is assumed by other models for major aerosol compositions. Curci et al. (2015) discussed the impacts of mixing state on simulated AOD and found that external mixing state assumption significantly increase simulated AOD. M6 used external mixing but shows a relative lower AOD because of ignorance of other aerosol species like dust, sea-salt, etc. In general, it appears the magnitude of inorganic

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aerosol concentrations and the hygroscopic growth efficiency (affected by varied RH) can account for or explain the simulated variations and magnitudes of AOD in Beijing during the severe haze event, given the aerosol size ~~distribution~~ lognormal treatments and mixing state are alike among models.

Table 6 shows the statistics for AOD simulation at NCP sites and at all sites. In the NCP region, R ranges from 0.36~0.74 for all the models. M2, M5 and M7 produce R around 0.7, indicating a better simulation of AOD variations. M2 and M7 exhibit the best R (0.65) for all sites. It is~~'s~~ noteworthy that R values at the sites in NCP are larger than that at all sites, indicating the larger reliability of model inputs (emissions and boundary conditions) and meteorological simulations. In terms of magnitudes, all models tend to underpredict AOD in the whole domain, with NMB of -2.7 to -71% in the NCP, and larger biases (NMB of -21~-75%) at all sites. M7 shows the smallest MBE (-0.05) and NMB (-2.7%) and M2 produces the smallest RMSE. It is interesting to note that the simulated AOD from the WRF-Chem models differed largely (-12 to -71%) between M1 and M3 at the NCP sites, and the WRF-Chem model using finer grid size (M4) can produced slightly smaller NMB compared with the same model using larger grid size (M3). However, as grid size becomes finer, R and RMSE from M4 may become worse, although AOD magnitude improved. The effect of grid resolution will be a topic of future paper.

5 Summary

The MICS-Asia Phase III Topic 3 examines how current online coupled air quality models perform in reproducing extreme aerosol pollution episodes in North China, and how high aerosol loadings during these episodes interact with radiation and weather. ~~Two hazy winter months,~~

~~namely January 2010 and January 2013, were studied by six modeling groups from China, Korea and the United States.~~ Predicted meteorological variables and air pollutants from these modeling groups were compared against each other, and measurements as well. A new anthropogenic emission inventory was developed for this phase (Li et al., 2017), and this inventory along with biogenic, biomass burning, air and ship, and volcano emissions were provided to all modeling groups. All modelling groups were required to submit results based on the analysis methodology that documented in this paper. This paper focused on the evaluation of the predictions of meteorological parameters and the predictions of aerosol mass, composition and optical depth. These factors play important roles in feedbacks impacting weather and climate through radiative and microphysical processes.

Comparisons against daily meteorological variables demonstrated that all models can capture the observed near surface temperature and water vapor mixing ratio, but near surface wind speeds ~~are~~ were overestimated by all models to varying degrees. The observed daily maximum downward shortwave radiation, particularly low values during haze days, were represented in the participating models. Comparisons with measurements of air pollutants, including SO_2, NO_x, CO, O_3, $PM_{2.5}$, and PM_{10}, from CARE-China and EANET networks showed that the main features of accumulations of air pollutants are represented in current generation of online coupled air quality models. The variations in observed AOD from CARE-China and AERONET networks were also reproduced by the participating models. Differences were found ~~exist~~ between simulated air pollutants, particularly ozone~~,~~. While winter time ozone levels are typically low (below 40 ppb) as photochemical pathways are slow, the models captured the synoptic variability but differed in the absolute magnitudes of near surface concentrations. The role of dry deposition and the boundary conditions play important roles.~~which are probably~~

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~~related to different treatments of emission inventory, different meteorological and chemical parameterizations, and uncertainties in interpolations from original emission inventory to model grids might also contribute to these differences.~~

~~Manifo~~Large differences in the models~~ld diversities~~ were found in the predicted $PM_{2.5}$ chemical compositions, especially secondary inorganic ~~aerosol~~s and organic carbon. During winter haze events, the production from gas phase chemistry is inhibited, and whether including other aerosol formation pathways (such as aqueous phase chemistry), or how the~~se~~ chemistry is parametrized leads to the large difference between simulated concentrations of secondary inorganic aerosols. In addition, differences in treatments of SOA also lead to large discrepancies between simulated OC concentrations. Differences in the simulated variations and magnitudes of AOD in Beijing during the severe haze event could be explained by the differences in simulated inorganic aerosol concentrations and the hygroscopic growth efficiency (affected by varied RH).

Results of this intercomparison show that there remain important issues with current coupled models in predicting winter haze episodes. Low wind speeds play an important role in haze episodes. Current models can predict the low wind speed - high haze relationship, but overestimate the low wind speeds. This contributed to the underestimation of $PM_{2.5}$. The models also underestimate the production of secondary inorganic aerosols. There is currently a great deal of research focused on inorganic aerosol production under winter haze conditions and new pathways need to be included in the models to improve prediction skills. Furthermore, current models have various treatments of SOA production and these lead to wide differences in the contribution of SOA to winter haze episodes.

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However, it was also found that using the ensemble mean of the models produced the best prediction skill. While this has been shown for other conditions (for example prediction of high ozone events in the US (Mckeen et al., 2004), this is to our knowledge the first time it has been shown for heavy haze events.

The uncertainties in predictions in aerosols mass and composition will impact estimates of the aerosol direct and indirect effects during haze event (Gao et al., 2017a, 2017b, 2017c). The results of the MICS-Asia Topic 3 experiments looking at the direct and indirect effects during these heavy haze events is the subject of companion papers.

~~These results provide some directions for future model improvements, and underscore the importance of accurately predicting aerosol concentration and compositions. Differences in the simulated variations and magnitudes of AOD in Beijing during the severe haze event could be explained by the differences in simulated inorganic aerosol concentrations and the hygroscopic growth efficiency (affected by varied RH).~~

~~Previous studies have studied radiative forcing during haze event (Gao et al., 2017), but there are large uncertainties in aerosol modeling during haze events and in estimating its interactions with weather and climate. The uncertainties come from model inputs (land use data, model initial and boundary conditions, etc.), physical and chemical mechanisms, and particularly parameterizations of aerosol-radiation-weather interactions. Other companion papers with respect to MICS-Asia Topic 3 will provide more insights into current knowledge of aerosol-weather interactions under heavy pollution conditions.~~

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Models	M1: WRF-Chem	M2: WRF-Chem	M3: NU-WRF	M4: NU-WRF	M5: RIEMS-Chem	M6: RegCCMS	M7: WRF-CMAQ
Modelling Group	Pusan National University	University of Iowa	USRA/NASA	USRA/NASA	Institute of Atmospheric Physics	Nanjing University	University of Tennessee
Grid Resolution	45km	50km	45km	15km	60km	50km	45km

Vertical Layers	40 layers to 50mb	27 layers to 50mb	60 layers to 20mb	60 layers to 20mb	16 layers to 100mb	18 layers to 50mb	
Gas phase chemistry	RACM-ESRL	CBMZ	RADM2	RADM2	CBM4	CBM4	SAPRC99
Aerosols	MADE/SORGAM modal scheme	MOSAIC-8bin	GOCART bulk scheme	GOCART bulk scheme	Sulfate, nitrate, ammonium, BC, OC, SOA, 5 bins of soil dust, and 5 bins of sea salt modal scheme	Sulfate, nitrate, ammonium, BC and POC bulk scheme	AE06 modal scheme
Chemical Boundary Conditions	Climatological data from NALROM	MOZART	MOZART GOCART	MOZART GOCART	GEOS-Chem	Climatological data	GEOS-Chem
Meteorological Boundary Conditions	NCEP FNL	NCEP FNL	NASA MERRA	NASA MERRA	NCEP FNL	NCEP-NCAR	NCEP FNL
BVOC emissions	prescribed	Internal calculation	Internal calculation	Internal calculation	prescribed	No BVOC	Internal calculation
Dust	NA	EROD data Opt 3	GOCART dust	GOCART dust	Han et al. (2004)	NA	No
Microphysics	Lin scheme	Morrison double-moment	GCE (Goddard Cumulus Ensemble)	GCE	Reisner mixed phase		
Longwave radiation	RRTMG	RRTMG	Goddard	Goddard	CCM3		RRTM
Shortwave radiation	RRTMG	RRTMG	Goddard	Goddard	Revised CCM3		Goddard
Boundary Layer	Yonsei University	Yonsei University	YSU	YSU	MRF	BATS	
Cu physics	Grell 3D	Grell 3D	Grell 3D	Grell 3D	Grell 3D		Grell 3D
Surface physics	Thermal diffusion	Unified Noah	Unified Noah	Unified Noah	BATS		
Aerosol-radiation	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Aerosol-microphysics	Yes	Yes	Yes	Yes	Yes	Yes	No
Mixing state	Internal mixing	Internal mixing	Internal mixing	Internal mixing	Internal mixing among inorganic aerosols and BC and OC, and external mixing between dust, sea-salt and other aerosols	External mixing	Internal mixing

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Table 1 Participating models in Topic 3

Models	M1: WRF-Chem1	M2: WRF-Chem2	M3: NU-WRF1	M4: NU-WRF2	M5: RIEMS-Chem	M6: RegCCMS	M7: WRF-CMAQ
Modelling Group	Pusan National University	University of Iowa	USRA/NASA	USRA/NASA	Institute of Atmospheric Physics	Nanjing University	University of Tennessee
Grid Resolution	45km	50km	45km	15km	60km	50km	45km
Vertical Layers	40 layers to 50mb	27 layers to 50mb	60 layers to 20mb	60 layers to 20mb	16 layers to 100mb	18 layers to 50mb	
Gas phase chemistry	RACM	CBMZ	RADM2	RADM2	CBM4	CBM4	SAPRC99
Aerosols	MADE	MOSAIC-8bin	GOCART	GOCART	Sulfate, nitrate, ammonium, BC, OC, SOA, 5 bins of soil dust, and 5 bins of sea salt	Sulfate, nitrate, ammonium, BC and POC	AE06
Chemical Boundary Conditions	Climatological data from NALROM	MOZART	MOZART GOCART	MOZART GOCART	GEOS-Chem	Climatological data	GEOS-Chem

Table 1

Participating models in Topic 3

M1: WRF-Chem v3.7.1; M2: WRF-Chem v3.5.1; M7: WRFv3.4.1&CMAQv5.0.2 NU-WRF v7lis7-3.5.1-p3

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Table 2 CARE-Chine network sites

ID	Site name	Characteristics	Longitude	Latitude
1	Beijing	AOD	116.37	39.97
2	Tianjin	Air quality*	117.21	39.08
3	Shijiazhuang	Air quality	114.53	38.03
4	Xianghe	Air quality	116.96	39.75
5	Xinglong	Air quality	117.58	40.39
6	Beijing Forest	AOD	115.43	39.97
7	Baoding	AOD	115.51	38.87
8	Cangzhou	AOD	116.80	38.28
9	Shenyang	AOD	123.63	41.52
10	Jiaozhou Bay	AOD	120.18	35.90

*Air quality: surface $PM_{2.5}$, PM_{10}, SO_2, NO_x, CO, O_3

Table 3 Performance Statistics of Meteorology Variables (RMSE and MBE units: degree for T2; g/kg for Q2; m/s for WS10; W/m^2 for SWDOWN)

Metrics	Models	T2	Q2	WS10	SWDOWN South	SWDOWN North
RMSE	M1	0.64	0.14	2.04	86.32	69.39
	M2	0.68	0.10	0.95	96.71	72.76
	M3	2.34	0.16	1.16	60.34	59.56
	M4	2.90	0.43	1.44	100.34	74.89
	M5	2.97	0.46	0.91	91.06	65.27
	M6	3.57	0.76	2.48	85.63	222.00
	M7	2.05	0.17	0.22	158.10	218.67
	Ensemble	1.81	0.10	1.28	81.96	62.51
MBE	M1	-0.19	0.02	2.01	66.58	59.94
	M2	-0.60	-0.01	0.91	83.88	62.38
	M3	-2.18	-0.04	1.11	36.44	47.74
	M4	-2.09	0.11	1.40	26.78	33.59
	M5	-2.73	0.43	0.74	49.06	51.00
	M6	-3.06	-0.56	2.37	-0.49	-202.26
	M7	-2.02	-0.12	0.15	145.24	159.02
	Ensemble	-1.71	-0.02	1.25	65.54	36.37
NMB (%)	M1	-0.07%	0.19%	17.58%	14.61%	13.34%
	M2	-0.21%	-0.12%	7.94%	18.41%	13.88%
	M3	-0.79%	-0.34%	9.73%	8.00%	10.63%
	M4	-0.75%	0.95%	12.26%	5.88%	7.48%
	M5	-0.98%	3.65%	6.45%	10.77%	11.35%
	M6	-1.10%	-4.77%	20.73%	-0.11%	-45.02%
	M7	-0.72%	-1.05%	1.31%	31.88%	35.39%
	Ensemble	-0.61%	-0.14%	10.98%	14.38%	8.10%

Table 4 Performance Statistics of Air Pollutants at the CARE-China sites (RMSE and MBE units: ppbv for gases and $\mu g/m^3$ for PM)

Metrics	Models	SO_2	NO_x	O_3	$PM_{2.5}$	PM_{10}		SO_2	NO_x	O_3	$PM_{2.5}$	PM_{10}
r	**M1**	0.76	0.60	0.46	0.85	0.76	**MBE**	-17.14	-5.53	-1.54	55.69	30.70
	M2	0.77	0.65	0.48	0.90	0.85		2.10	33.41	2.53	48.44	12.94
	M3	0.69	0.66	0.39	0.85	0.68		-15.89	-8.00	23.93	8.13	-19.92
	M4	0.67	0.61	0.42	0.88	0.73		-9.98	0.28	24.49	23.12	-3.23
	M5	0.72	0.73	0.39	0.91	0.84		-9.69	64.29	-5.30	1.68	-52.49
	M6	0.62	0.48	-	-	-		-27.53	-29.98	-	-	-
	M7	0.57	0.58	0.48	0.82	0.77		-25.56	7.85	-3.09	43.59	-21.00
	Ensemble	0.79	0.71	0.51	0.94	0.87		-14.81	8.90	6.84	30.11	-8.83
RMSE	**M1**	27.63	33.51	6.40	73.37	79.06	**NMB (%)**	-14.05	-5.41	7.37	63.57	18.93
	M2	21.00	66.30	8.15	72.44	80.72		12.13	69.58	39.87	54.07	6.38
	M3	29.50	36.87	24.76	47.20	78.21		-10.44	-6.26	306.33	9.67	-12.41
	M4	26.86	36.10	25.34	49.13	72.25		0.31	4.51	316.99	27.03	-1.78
	M5	32.17	87.48	7.90	45.32	81.00		6.83	127.45	-38.49	0.52	-32.94
	M6	33.95	48.62	-	-	-		-51.28	-48.59	-	-	-
	M7	34.75	35.88	6.89	64.25	70.19		-37.87	18.32	-7.78	48.92	-12.78
	Ensemble	24.10	29.12	8.86	45.25	56.65		-13.48	22.80	104.04	33.96	-5.77
MFB (%)	**M1**	-17.32	5.26	-5.06	64.34	21.98	**MFE (%)**	53.73	43.79	54.54	69.92	41.95
	M2	9.09	32.82	19.88	51.18	3.44		43.18	73.39	60.79	59.87	39.35
	M3	-12.96	4.52	113.60	32.67	-4.62		57.87	46.69	113.60	50.10	36.83
	M4	1.53	15.34	114.35	45.27	6.07		46.30	48.13	114.35	55.03	34.72
	M5	-20.24	67.25	-62.65	16.88	-35.15		63.69	72.07	80.92	48.17	45.09
	M6	-77.13	-56.89	-	-	-		84.21	69.66	-	-	-
	M7	-46.67	21.80	-19.50	57.19	-7.02		72.35	49.18	60.64	66.27	35.83
	Ensemble	-14.17	26.41	62.86	50.61	3.12		43.13	42.94	71.14	55.86	28.05

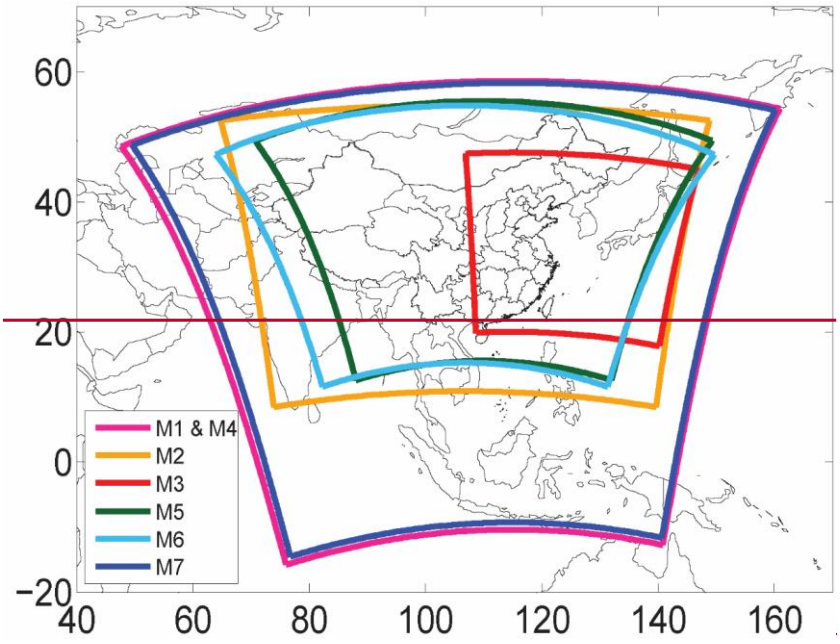
Table 5 Performance Statistics of Air Pollutants at the EANET sites (RMSE and MBE units: ppbv for gases and μg/m^3 for PM)

Metrics	Models	SO_2	NO_x	O_3	PM_{10}		SO_2	NO_x	O_3	PM_{10}
	M1	0.57	0.64	0.14	0.59		-0.68	0.68	-6.16	-21.03
	M2	0.59	0.45	0.30	0.75		-0.45	-0.39	5.50	3.12
	M3	0.50	0.55	0.26	0.51		-0.37	-0.21	3.67	3.55
	M4	0.45	0.55	0.25	0.49		-0.57	-0.61	4.28	2.96
	M5	0.58	0.54	0.01	0.03		-0.57	1.28	4.67	3.77
r	**M6**	0.33	0.24	-	-	**MBE**	0.32	-1.68	-	-
	M7	0.53	0.49	0.38	0.55		-0.03	0.64	-1.89	-15.75
	Ensemble	0.60	0.66	0.32	0.59		-0.34	-0.07	1.68	-3.89
	M1	-46.45	41.49	-15.03	-82.29		1.18	1.37	8.23	23.39
	M2	-29.64	-29.75	13.47	18.90		1.01	1.35	7.29	10.01
	M3	-25.42	-17.75	9.01	19.46		1.02	1.02	6.44	13.71
NMB	**M4**	-39.63	-35.84	10.47	16.95	**RMSE**	1.14	0.97	6.35	13.78
(%)	**M5**	-34.23	38.50	11.38	31.80		1.27	2.75	12.27	23.10
	M6	12.63	-93.57	-	-		1.38	1.85	-	-
	M7	17.42	31.47	-4.71	-56.18		1.04	1.57	6.52	18.76

Ensemble	-20.76	-10.79	4.10	-8.56		0.96	0.79	4.98	11.69

Table 6 Performance Statistics of AOD

Metrics	Models	M1	M2	M3	M4	M5	M6	M7	Ensemble
R	North China	0.63	0.74	0.57	0.51	0.68	0.36	0.71	0.77
	All	0.60	0.65	0.46	0.42	0.53	0.33	0.64	0.75
MBE	North China	-0.25	-0.10	-0.09	-0.07	-0.13	-0.21	-0.05	-0.03
	All	-0.18	-0.02	-0.01	-0.01	-0.01	-0.11	0.00	-0.12
NMB (%)	North China	-71.25	-23.28	-12.63	-9.59	-28.34	-59.19	-2.70	-30.17
	All	-74.94	-30.69	-25.68	-23.64	-28.24	-55.38	-21.12	-28.91
RMSE	North China	0.35	0.20	0.26	0.28	0.24	0.36	0.22	0.22
	All	1.16	1.13	1.15	1.15	1.15	1.17	1.14	0.20


60
40
20
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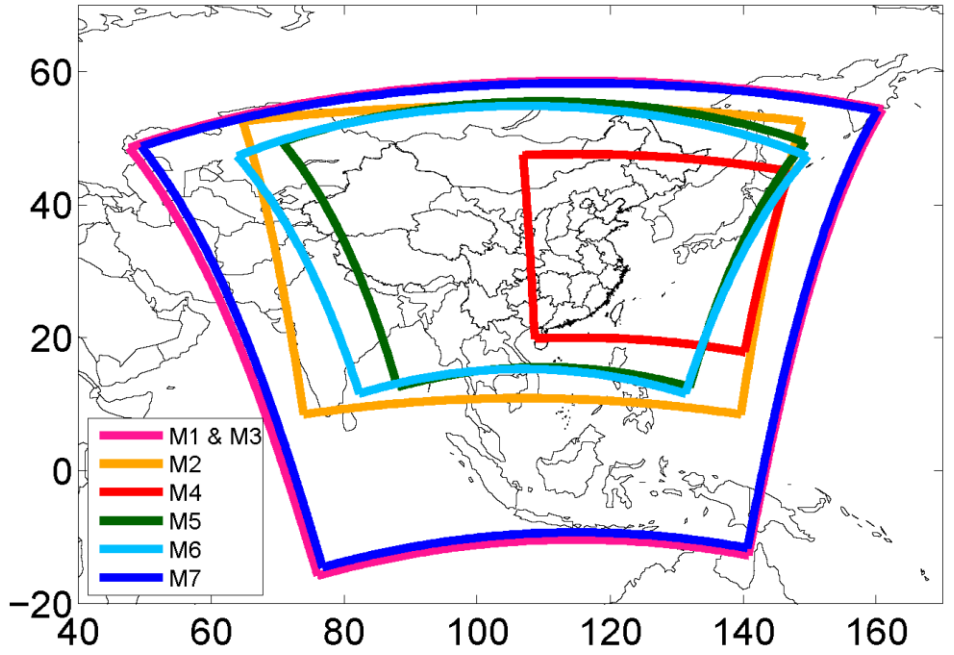
120
140
160
M1 & M4
M2
M3
M5
M6
M7


60
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M1 & M3
M2
M4
M5
M6
M7

Figure 1. MICS-ASIA III Topic 3 modeling domains (descriptions of each model are documented in Table 1) M1: WRF-Chem 45km; M2: WRF-Chem 50km; M3: NU WRF 45km; M4: NU-WRF 15km; M5: RIEMS-IAP 60km; RegCCMS 50km; WRF-CMAQ 45km

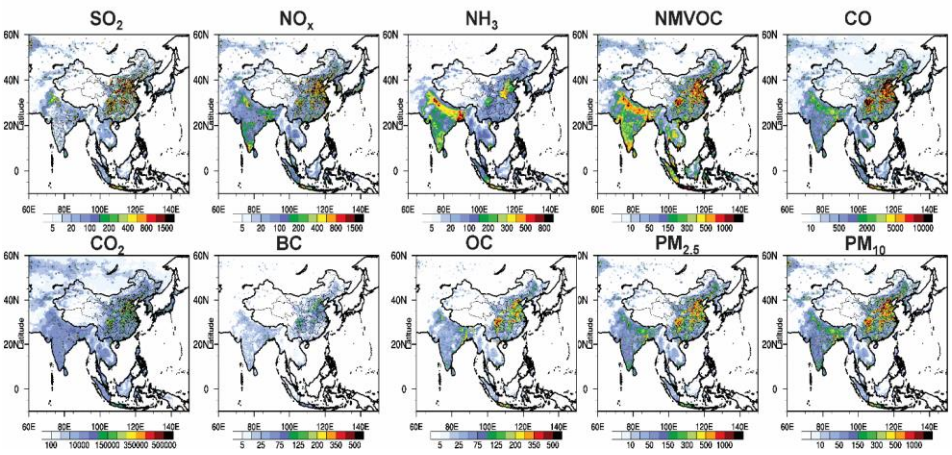
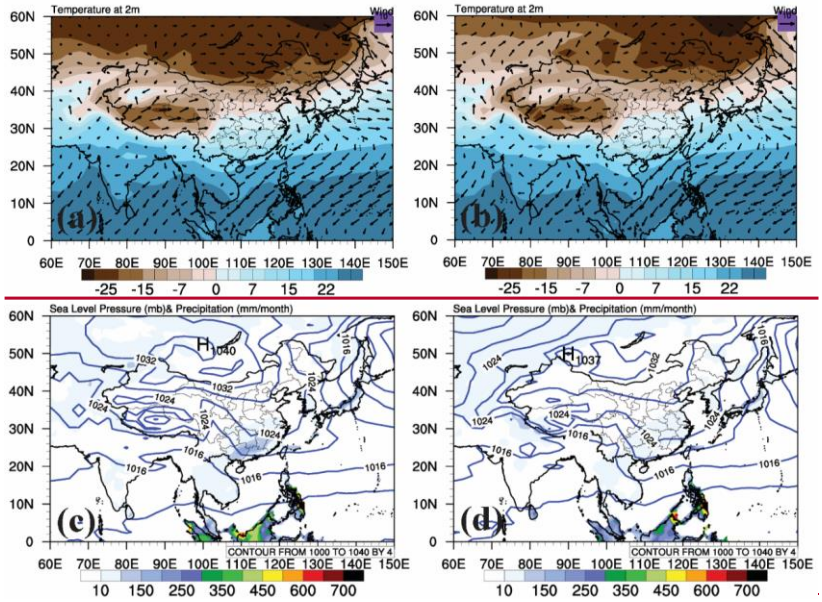


Figure 2. MIX emission inventory for January 2010 (Mg/month/grid)



~~Figure 3. Monthly mean temperature at 2m, winds at 10m, total precipitation and sea level pressure for January 2010 (a,c) and January 2013 (b,d)~~

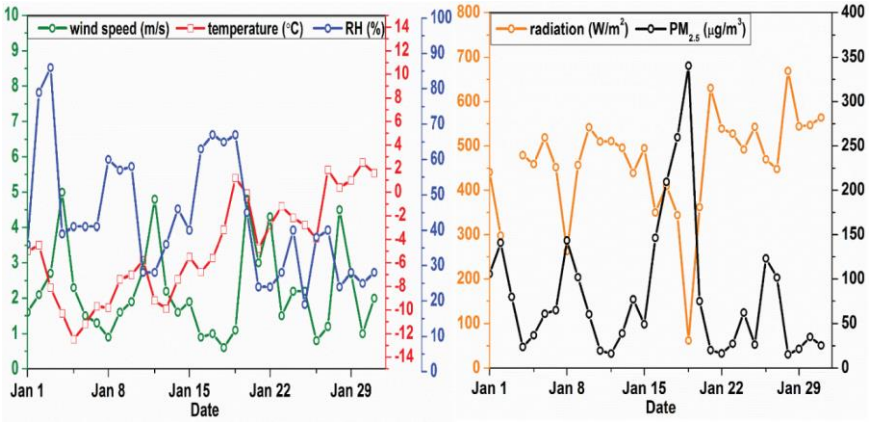


Figure ~~4~~3. Observed near surface daily meteorological variables and $PM_{2.5}$ concentrations in Beijing for January 2010

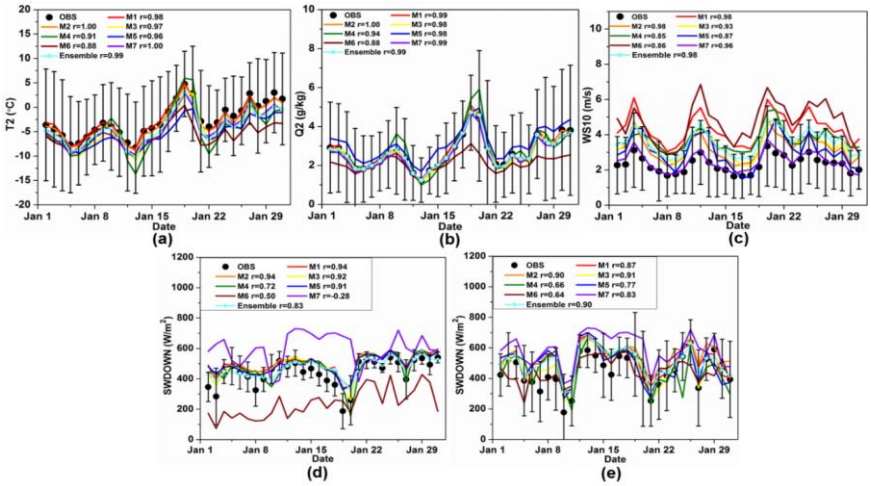


Figure ~~5~~4. Comparisons between simulated and observed near surface temperature (a), water vapor mixing ratio (b), and wind speeds (c) (T2, Q2, and WS10), downward shortwave radiation in North China (d) and South China (e) (spatial daily values are averaged over measurements shown in S4 and S5; the error bars show the standard deviation of values over the measurement sites)

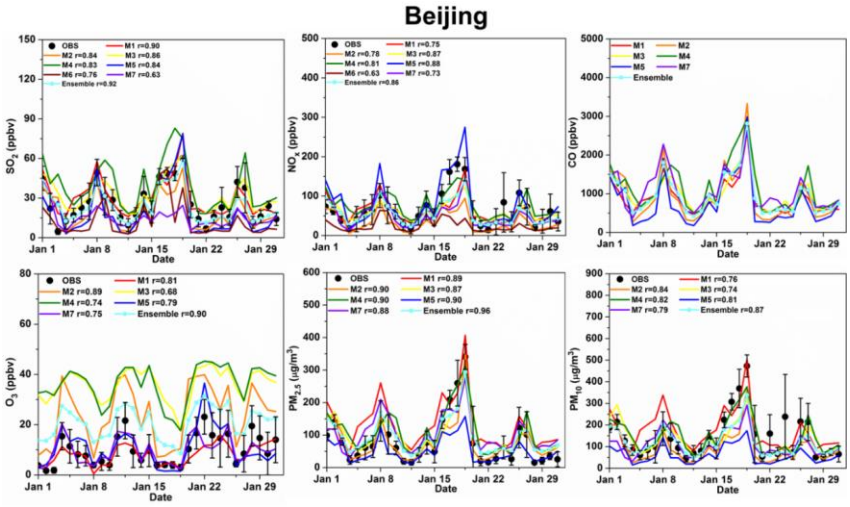


Figure ~~6~~5. Comparisons between simulated and observed daily air pollutants (SO_2, NO_x, CO, O_3, $PM_{2.5}$ and PM_{10}) at the Beijing CARE-China site

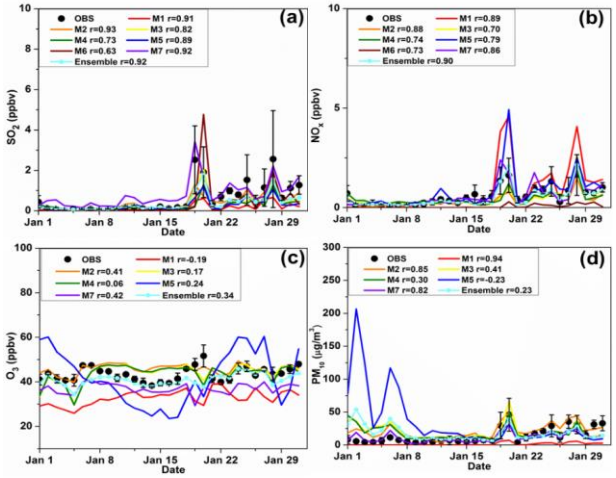


Figure ~~7~~6. Comparisons between simulated and observed daily air pollutants (SO_2, NO_x, O_3, and PM_{10}) at the Rishiri EANET sites

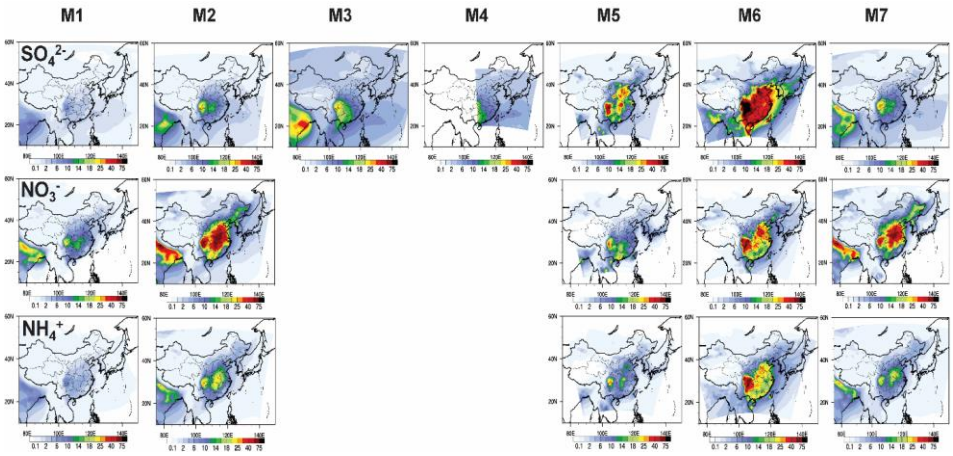


Figure ~~8~~7. Simulated monthly concentrations of major $PM_{2.5}$ components (μg/m^3) for January 2010 from all participating models

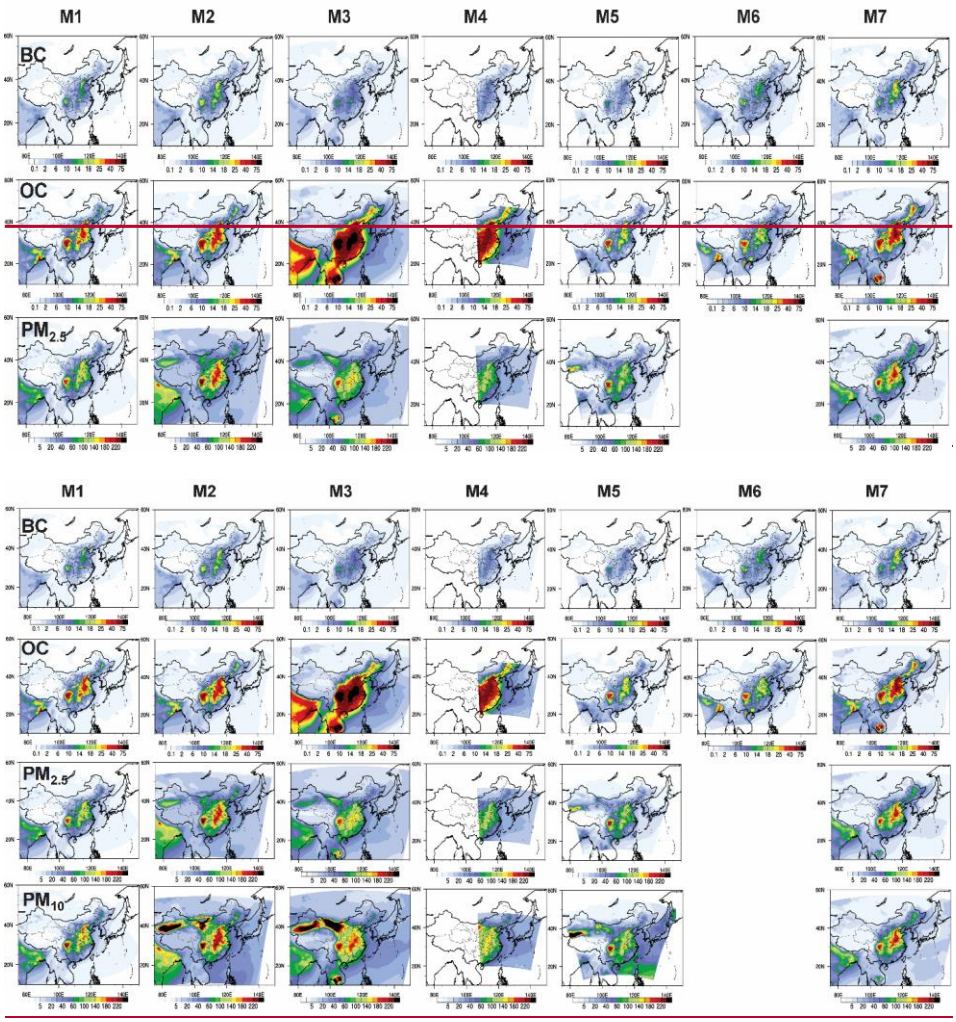


Figure ~~9~~8. Simulated monthly concentrations of $PM_{2.5}$ and major $PM_{2.5}$ components (μg/m^3) for January 2010 from all participating models

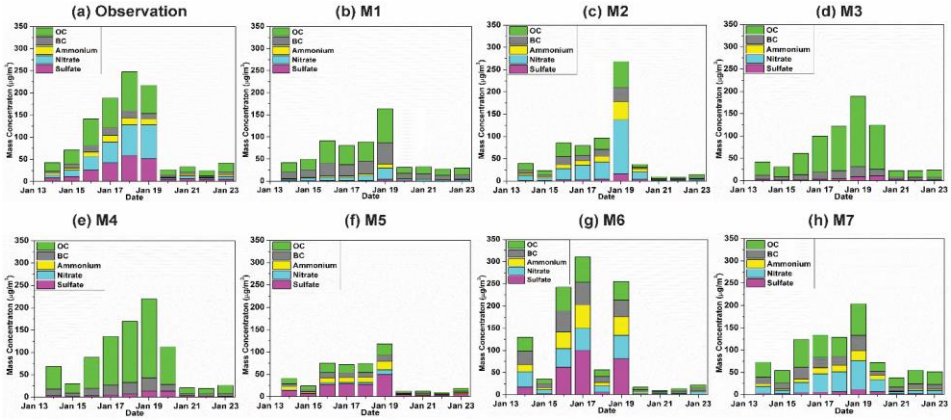


Figure ~~10~~9. Observed and simulated daily mean concentrations of major $PM_{2.5}$ chemical components in the urban Beijing site

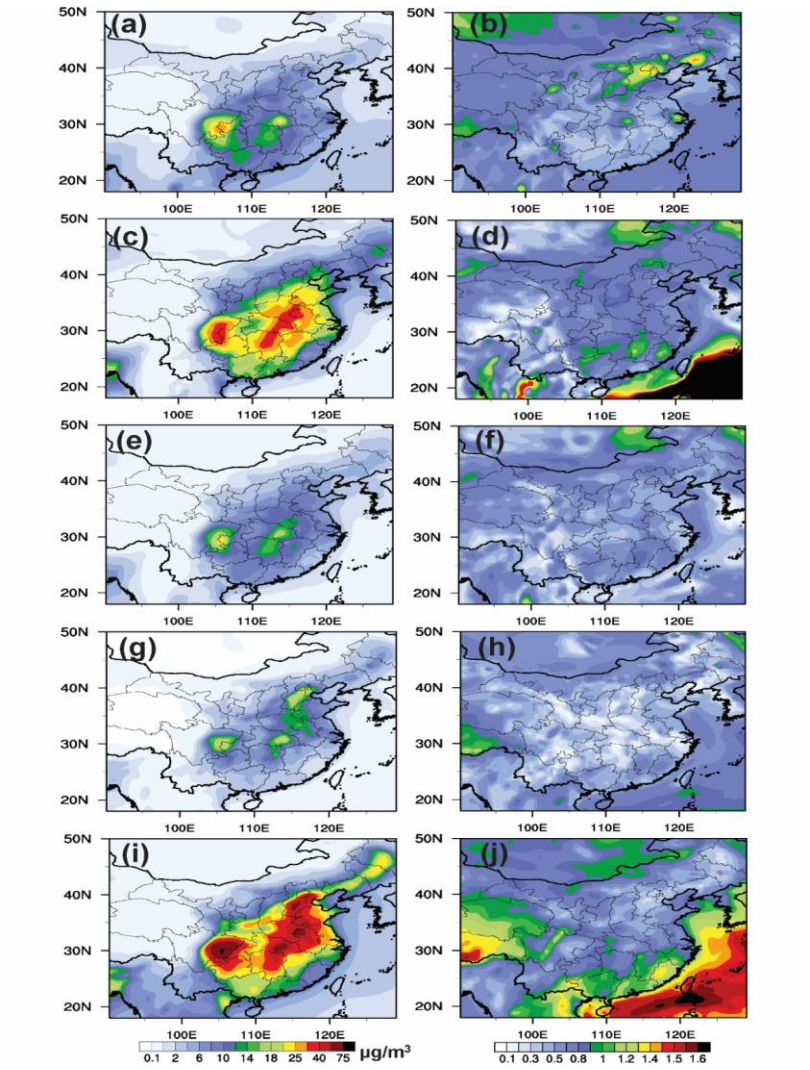


Figure ~~11~~10. The ensemble mean monthly averaged near-surface distributions of $PM_{2.5}$ compositions for January 2010 (sulfate (a), nitrate (c), ammonium (e), BC (g), and OC (i)), along with the spatial distribution of the coefficient of variation ((b), (d), (f), (h), and (j), standard deviation divided by the average)

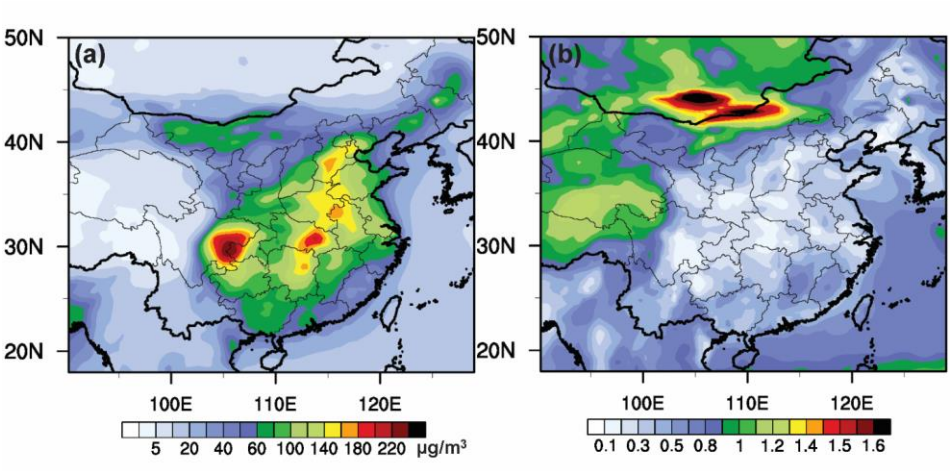


Figure ~~12~~11. The ensemble mean monthly averaged near-surface distributions of $PM_{2.5}$ for January 2010 (a), along with the spatial distribution of the coefficient of variation (b, standard deviation divided by the average)

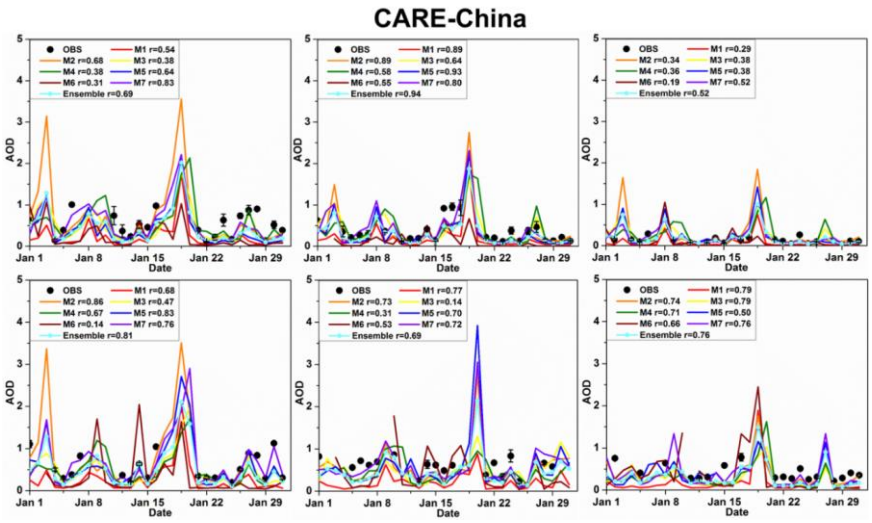


Figure ~~13~~12. Comparisons between simulated and observed daily (daytime) mean AOD at the CARE-China sites (Baoding, Beijing City, Beijing Forest, Cangzhou, Jiaozhou, Shenyang,)

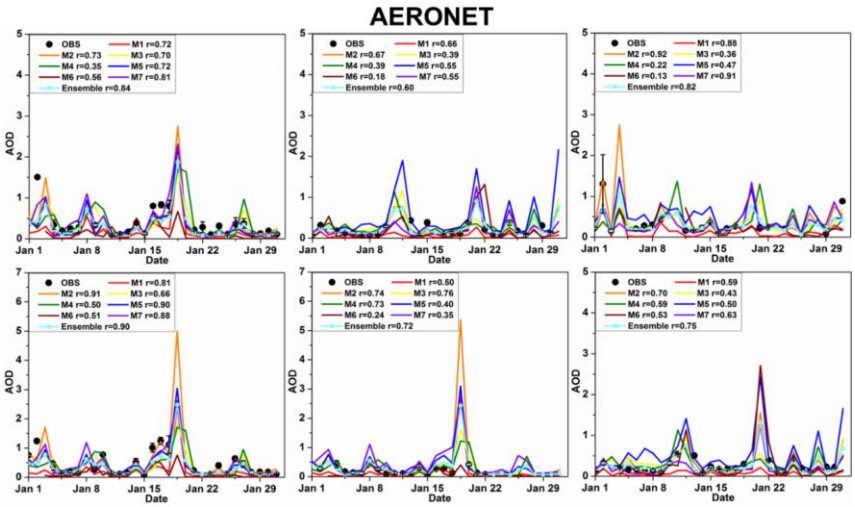


Figure ~~14~~13. Comparisons between simulated and observed daily (daytime) mean AOD at the AERONET sites (Beijing, Shirahama, GIST, Xianghe, Xinglong, Osaka)