Anonymous Referee #1

General comments: The manuscript entitled "Air Quality and Climate Change, Topic 3 of the Model Inter-Comparison Study for Asia Phase III (MICS-Asia III), Part II: aerosol radiative effects and aerosol feedbacks discussed the estimates of aerosol radiative forcing, aerosol feedbacks and the dominant roles of direct forcing. The possible causes for the differences among the models were also analyzed by sensitivity simulation. Some interesting results have been found. However, the manuscript needs to be improved in writing and logically organization in its structure. I recommend publishing it after major revision.

- Reply: Thanks for the valuable comments. We have revised the manuscript following your comments, which are shown below.

Specific comments:

1. There are a few grammatical errors, please find a native speaker to proofread the paper.

- Reply: We, including a native speaker, have carefully read the manuscript and edited to avoid grammatical errors.

2. The sections of manuscript need to be organized more logically in structure.

- Reply: The manuscript discusses aerosol radiative forcing as it is closely connected with aerosol feedbacks, so we put it in Sect. 3 while aerosol feedbacks in Sect. 4. In Sect. 5, simulations were conducted to figure out the reasons for differences. We added Sect. 2 to describe the design of the experiment to make it easier to read.

3. Lines 61-65: Some important previous work in China from the observational perspective have been ignored, including

Huang et al., Satellite-based assessment of possible dust aerosols semi-direct effect on cloud water path over East Asia, 2006;

Liu et al., Aerosol optical properties and radiative effect determined from sky-radiometer over Loess Plateau of Northwest China, 2011;

Also, Line 82-89: some modeling work have been ignored, including

Chen et al., Modeling the transport and radiative forcing of Taklimakan dust over the Tibetan Plateau, 2013;

Chen et al., Dust modeling over East Asia during the summer of 2010 using the WRF-Chem model, 2018;

Liu et al., Modeling study on the transport of summer dust and anthropogenic aerosols over the Tibetan Plateau, 2015;

Jia et al., Estimation of the aerosol radiative effect over the Tibetan Plateau based on the latest CALIPSO product, 2018.

These researches are highly relevant to the topic investigated here.

- Reply: Thanks for mentioning these important studies. We have added these references in the revised manuscript.
- Huang, J., Lin, B., Minnis, P., Wang, T., Wang, X., Hu, Y., Yi, Y. and Ayers, J.K.: Satellite-based assessment of possible dust aerosols semi-direct effect on cloud water path over East Asia, Geophy. Res. Let., 33(19), https://doi.org/10.1029/2006GL026561, 2006.
- Liu, Y., Huang, J., Shi, G., Takamura, T., Khatri, P., Bi, J., Shi, J., Wang, T., Wang, X. and Zhang, B.: Aerosol optical properties and radiative effect determined from sky-radiometer over Loess Plateau of Northwest China, Atmos. Chem. Phys., 11(22), pp.11455-11463, https://doi.org/10.5194/acp-11-11455-2011, 2011.
- Chen, S., Huang, J., Zhao, C., Qian, Y., Leung, L.R. and Yang, B.: Modeling the transport and radiative forcing of Taklimakan dust over the Tibetan Plateau: A case study in the summer of 2006, Jour. Geophy. Res.: Atmos., 118(2), pp.797-812, https://doi.org/10.1002/jgrd.50122, 2013.
- Chen, S., Yuan, T., Zhang, X., Zhang, G., Feng, T., Zhao, D., Zang, Z., Liao, S., Ma, X., Jiang, N. and Zhang, J.: Dust modeling over East Asia during the summer of 2010 using the WRF-Chem model, Jour. Quan. Spec. Rad. Tran., 213, pp.1-12, https://doi.org/10.1016/j.jqsrt.2018.04.013, 2018.
- Liu, Y., Sato, Y., Jia, R., Xie, Y., Huang, J. and Nakajima, T.: Modeling study on the transport of summer dust and anthropogenic aerosols over the Tibetan Plateau, Atmos. Chem. Phys., 15(21), pp.12581-12594, https://doi.org/10.5194/acp-15-12581-2015, 2015.
- Jia, R., Liu, Y., Hua, S., Zhu, Q. and Shao, T.: Estimation of the aerosol radiative effect over the Tibetan Plateau based on the latest CALIPSO product, Jour. Met. Res., 32(5), pp.707-722, https://doi.org/10.1007/s13351-018-8060-3, 2018.

4. Line 66-69, please cite the previous researches as an illustration basis.

- Reply: We have included these studies in the revised manuscript:
- "Aerosols change weather and climate via the following pathways: they absorb and scatter solar and thermal radiation to alter the radiative balance of the earth-atmosphere system (Liu et al., 2011; Jia et al., 2018)";
- "The suppression of cloud convection induced by direct effects of absorbing aerosols is called the semi-direct effect (Huang et al., 2006; Lohmann and Feichter, 2005)."
- "including WRF-Chem (the Weather Research Forecasting model coupled with Chemistry, Chen et al., 2013, 2018; Gao et al., 2016, 2017; Liu et al., 2015),"

5. Lines 80-81, the meaning of theory and practice of studying aerosol feedbacks over Asia should be illustrated in detail.

- Reply: We have added descriptions of the theory in the revised manuscript: "High concentrations of aerosols would enhance the stability of boundary layer due to reductions in radiation that reach the surface, which in turn cause further increases in PM2.5 concentrations (Ding et al., 2016; Gao et al., 2016)".
- More explanations of related studies can be found in Sect. 3: "These results can be compared to previous studies. The contributions of aerosol-radiation feedback to haze formation in China have been investigated in many previous studies (Ding et al., 2016; Gao et al., 2015; Gao et al., 2016; Liu et al., 2018; J. Wang et al., 2014; Z. Wang et al., 2014; Wang et al., 2015; Wu et al., 2019; Zhang et al., 2015; Zhang et al., 2018; Zhong et al., 2018), but the reported values partly diverge."

6. Please add a section to describe the model and research methodology. Move the illustration in Line 90-101 and Line 157-159 to the new section.

- Reply: Model descriptions, research methodology and model evaluations were provided in a companion paper, part I:
- Gao, M., Han, Z., Liu, Z., Li, M., Xin, J., Tao, Z., Li, J., Kang, J.-E., Huang, K., Dong, X., Zhuang, B., Li, S., Ge, B., Wu, Q., Cheng, Y., Wang, Y., Lee, H.-J., Kim, C.-H., Fu, J. S., Wang, T., Chin, M., Woo, J.-H., Zhang, Q., Wang, Z., and Carmichael, G. R.: Air quality and climate change, Topic 3 of the Model Inter-Comparison Study for Asia Phase III (MICS-Asia III) – Part 1: Overview and model evaluation, Atmos. Chem. Phys., 18, 4859–4884, https://doi.org/10.5194/acp-18-4859-2018, 2018.
- In the revised manuscript, we have added a section to describe the activity.
- "2 Overview of MICS-Asia III Topic 3 The participants were requested to use common emissions to simulate air quality during January 2010 and submit requested model variables. 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: 45 and 15 km horizontal resolutions) of the National Aeronautics and Space Administration (NASA) Unified WRF (NU-WRF; *Peters-Lidard*

et al., 2015) model by the Universities Space Research Association (USRA) and NASA's Goddard Space 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). A new Asian emission inventory was developed for MICS-III by integrating state-of-the-art national or regional inventories to support this model intercomparison study (*Li et al., 2017*), which was provided to all modeling groups, along with biogenic emissions, biomass burning emissions, emissions from air and ship transport, volcano emissions, and dust emissions. Simulations of two global chemical transport models (e.g., GEOS-Chem (The Goddard Earth Observing System Model-Chemistry) and MOZART (Model for OZone And Related chemical Tracers)) were used as boundary conditions for MICS-Asia III. Comprehensive model evaluations suggest that all models could capture the observed near-surface temperature and water vapor mixing ratio, but overestimated near-surface wind speeds to varying degrees. Participating models were able to represent the observed daily maximum downward shortwave radiation, particularly low values during haze days, and the observed variations of air pollutants, including SO_2, NO_x, CO, O_3, $PM_{2.5}$, and PM_{10}. However, large differences in the models were found in the predicted $PM_{2.5}$ chemical compositions."

7. Please give the detailed description when the abbreviation first appears (for example, M1, M2, M3. . .M7).

- Reply: M1, M2…M7 represent different participating models, which were documented in our paper part I. To make it easier to read, we added Table 1 into the revised manuscript: "from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (Table 1:".

8. Please use the box or symbol to show the BTH region, Huabei province and Beijing in Figure 1.

- Reply: BTH and Beijing are small areas in Figure 1 and it is not clear if we mark them in Figure 1. Thus, we add Figure S1 in the supplement to display the BTH region (marked with blue) and Beijing (shown using the green arrow). In the revised manuscript, we change Huabei province to the BTH region.

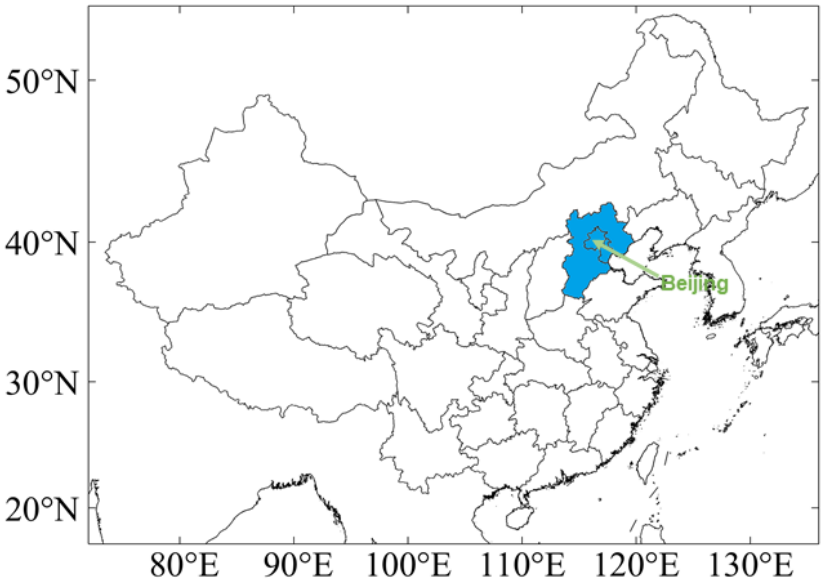

Figure S1. The Beijing-Tianjin-Hebei region is marked with blue and Beijing is shown with green arrow.

9. Line 118-119, the description is inconsistent with Table 1.

- Reply: We have changed it to make it consistent.

10. Line 145-149, why do you only use M4 and M5 to provide direct and indirect aerosol radiative forcing? Why do you only use M5 to study the effects of aerosols mixing state, hygroscopic growth, black carbon and mineral dust. Obviously, the values given by these models are very different. Which should be illustrated.

- Reply: MICS-Asia is a volunteer-based model inter-comparison activity. Only limited model outputs were requested. Most modeling groups did conduct extra numerical experiments to distinguish direct and indirect forcing. Thus, we only include the discussions based on M4 and M5, and the results from these two model applications are consistent that direct forcing dominates the total forcing.
- Please notice that we have updated the results from M5 in the revised manuscript as some mistakes were found for M5 in the calculations of optical properties. The updated results show higher agreements with other models, but M5 still produces the largest (negative)

radiative forcing at TOA and the second largest forcing at the surface in both the BTH region and Beijing.

- It is of great importance to understand the drivers for the differences, and the IAP group (M5) volunteered to conduct additional simulations. We agree that different models would behave differently in the sensitivity simulations. To address this, we added comparison and discussion in the revised manuscript to provide a clearer picture of how these factors would influence the results.
- "Large uncertainties still remain in the estimates of the role of BC in aerosol feedbacks relative to scattering aerosols. Gao et al. (2016) suggested that the impacts of BC on boundary layer height and PM2.5 concentrations can account for as high as 60% of the total aerosol feedbacks in the North China Plain at 2 p.m., although it only accounts for a small share of PM in terms of mass concentration. Qiu et al. (2017) indicated that PM2.5 concentrations averaged over the North China Plain increased by 16.8% and 1.0% due to scattering aerosols and BC, respectively. It should be noted that most participating models, including RIEMS-Chem, tend to underpredict the total mass concentrations of scattering aerosols (inorganic and organic aerosols) by up to a factor of two over the study period, leading to overestimation of the contribution of BC."

11. Line 195- 196, the description is inconsistent with Table 3. M2 (12.9)
    - Reply: We have changed the upper range to 12.9.

12. Please add a discussion about the simulation performance of different models.
    - Reply: We evaluated all models in the companion paper part I: "model evaluations have been archived in Gao et al. (2018a)."
    - We did include discussions in this manuscript:
    - "Model evaluation of PM2.5 composition in Gao et al. (2018a) reveals that M4 overpredicts strong scattering organic carbon, which could be one of the reasons for higher temperature reduction."
    - "As suggested in model evaluation, sulfate and organic aerosol concentrations are generally underestimated by most models in this study, except that M4 overestimate organic aerosol (Gao et al., 2018a). These were attributed to the missing multiphase

oxidation mechanisms of SO2, and different secondary organic aerosol (SOA) formation mechanisms in these models (Gao et al., 2018a)."

- Following your suggestion, we added more discussions of model evaluation in the revised manuscript: "Comprehensive model evaluations suggest that all models could capture the observed near-surface temperature and water vapor mixing ratio, but overestimated near-surface wind speeds to varying degrees. Participating models were able to represent the observed daily maximum downward shortwave radiation, particularly low values during haze days, and the observed variations of air pollutants, including SO2, NOx, CO, O3, PM2.5, and PM10. However, large differences in the models were found in the predicted PM2.5 chemical compositions. "

13. Line 271-272 please give some evidence for that the effect of BC indicates smaller than that of other scattering aerosols. How about the effect of sulfate aerosol?

- Reply: To avoid confusion, we rewrote the sentence in the revised manuscript and added discussions:
- "Two sets of simulations, namely without BC and with doubled BC concentrations, were conducted to examine the influences of BC on aerosol radiative forcing and feedbacks. In the control case, the aerosol induced changes in monthly mean surface air temperature, wind speed and PM2.5 values are -0.47 °C, -0.03 m/s and 1.5 μg/m3 for the BTH region, respectively. When BC is not included (only scattering aerosols and dust), the corresponding aerosol induced changes are -0.37 °C, -0.02 m/s and 0.7 μg/m3, respectively. When BC concentrations are doubled, these values change to -0.52 °C, -0.04 m/s and 2.2 μg/m3, respectively. The comparison between the control case and two additional sensitivity cases indicates that the changes caused by BC are comparable to those by scattering aerosols. The contribution of BC to aerosol feedbacks can reach up to 40~50%. It is also found that the influence of BC on aerosol feedbacks with internal mixing assumption is larger than that with external mixing assumption (Figure not shown).
- Large uncertainties still remain in the estimates of the role of BC in aerosol feedbacks relative to scattering aerosols. Gao et al. (2016) suggested that the impacts of BC on boundary layer height and PM2.5 concentrations can account for as high as 60% of the total aerosol feedbacks in the North China Plain at 2 p.m., although it only accounts for a small share of PM in terms of mass concentration. Qiu et al. (2017) indicated that PM2.5

concentrations averaged over the North China Plain increased by 16.8% and 1.0% due to scattering aerosols and BC, respectively. It should be noted that most participating models, including RIEMS-Chem, tend to underpredict the total mass concentrations of scattering aerosols (inorganic and organic aerosols) by up to a factor of two over the study period, leading to overestimation of the contribution of BC."

- In the companion paper part I, simulations of aerosol components were validated against observations in Beijing. Observations show that concentrations of sulfate (daily mean exceeding 60 μg/m3) are comparable to nitrate, and are about 30% lower than the concentrations of OC during haze days. The hygroscopicity of sulfate and nitrate is larger than that of OC. Most of the models underpredict concentrations of total scattering aerosols (sulfate, nitrate and OC) by up to a factor of two, while models show reasonably good skills for BC. These results imply that the effect of scattering aerosols on aerosol feedbacks might have been underestimated.
- There were no simulations in MICS-Asia to separate the effect of sulfate. In the revised manuscript, we added sentences to acknowledge this limitation: "Huang et al. (2015) separated the contributions of different aerosol components to aerosol direct radiative forcing, highlighting the roles of BC and sulfate. Future studies are also needed to separate the effects of other aerosol components, including sulfate, on aerosol feedbacks. "
- Qiu, Y., Liao, H., Zhang, R. and Hu, J.: Simulated impacts of direct radiative effects of scattering and absorbing aerosols on surface layer aerosol concentrations in China during a heavily polluted event in February 2014, Jour. Geophy. Res.: Atmos., 122(11), pp.5955-5975, https://doi.org/10.1002/2016JD026309, 2017.
- Huang, X., Song, Y., Zhao, C., Cai, X., Zhang, H. and Zhu, T.: Direct radiative effect by multicomponent aerosol over China, Jour. Clim., 28(9), pp.3472-3495, https://doi.org/10.1175/JCLI-D-14-00365.1, 2015.

Technical corrections:

1. Line 110, 'In Sect. 2'??? Please check it.

- Reply: We've changed to Sect. 3.

2. Line 118, Please give the full spelling of BTH when the abbreviation appears at the first time.

- Reply: We've changed accordingly.

3. Line 118, 'reports' should be changed to 'report'.

- Reply: We've changed accordingly.

4. Line 121, Please give the full spelling of AOD.

- Reply: We've changed accordingly.

5. Line 257, 'by dust' should be changed to 'of dust'.

- Reply: We've changed accordingly.

Anonymous Referee #2

General comments

The paper is the second part of two papers discussing the results of the MICS-Asia III model inter-comparison exercise with special focus on the performance of online coupled air quality models in simulating high aerosol pollution in the North China Plain region during wintertime haze events. While the focus of first part is on the description of the design of the modelling exercise and the overall model performance, this paper focuses on the role of aerosol radiative forcing and aerosol meteorology interactions for six different models. By means of case studies with one of the models, the authors investigate the sensitivity of aerosol radiative forcing to different aerosol descriptions. The current paper includes some interesting results and is generally worth to be published. However, some aspects need to be discussed in more detail and the presentation quality must be improved for major parts of the paper. Therefore, I recommend publishing the paper after major revisions.

- Reply: Thanks for the valuable comments. We have revised the manuscript following your comments, especially the presentation quality. Detailed modifications are shown below.

Specific comments

Although the paper is the second of two associated papers, it is necessary to add a section that gives a brief overview of the experimental design and model setup as well as the applied models. Furthermore, the name of the models should be connected to the abbreviations M1, M2, . . . This information is given in Part 1, so this is obviously not a secret. It could be looked up there, but including this information also in this paper (e.g. in Table 1) would enhance the paper's readability considerably.

- Reply: Thanks for the good suggestion. In the revised manuscript, we have added a section to describe the activity.
- We have added Table 1 to connect model names and abbreviations in the revised manuscript.
- "2 Overview of MICS-Asia III Topic 3 The participants were requested to use common emissions to simulate air quality during January 2010 and submit requested model variables. 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: 45 and 15 km horizontal resolutions) of the National Aeronautics and Space Administration (NASA) Unified WRF (NU-WRF; *Peters-Lidard et al., 2015*) model by the Universities Space Research Association (USRA) and NASA's Goddard Space 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). A new Asian emission inventory was developed for MICS-III by integrating state-of-the-art national or regional inventories to support this model intercomparison study (*Li et al., 2017*), which was provided to all modeling groups, along with biogenic emissions, biomass burning emissions, emissions from air and ship transport, volcano emissions, and dust emissions. Simulations of two global chemical transport models (e.g., GEOS-Chem (The Goddard Earth Observing System Model-Chemistry) and

MOZART (Model for OZone And Related chemical Tracers)) were used as boundary conditions for MICS-Asia III. Comprehensive model evaluations suggest that all models could capture the observed near-surface temperature and water vapor mixing ratio, but overestimated near-surface wind speeds to varying degrees. Participating models were able to represent the observed daily maximum downward shortwave radiation, particularly low values during haze days, and the observed variations of air pollutants, including SO_2, NO_x, CO, O_3, $PM_{2.5}$, and PM_{10}. However, large differences in the models were found in the predicted $PM_{2.5}$ chemical compositions."

Please add also some information about the length of the simulated episode and the simulation setup. Was the entire episode covered by one single simulation or was the episode simulated as a sequence of shorter time slices? The way how the simulation is performed can affect the development of semi-direct effects to a certain amount.

- Reply: We have added one sentence to describe this: "The entire month of January 2010 was simulated and covered by one single simulation for each participating model."

Why is model M3 not included? According to part 1 (Gao et al., 2018a) the simulation with WRF-CMAQ (M7) was performed with aerosol–radiation interactions turned off. If this is also the case here, this should be mentioned and eventual implications on the results should be discussed.

- Reply: Nudging of meteorological variables were applied for M3, so the simulated feedbacks are not apparent. We decided not to include in the comparison. In model evaluation shown in Gao et al. (2018a), the results of M7 are from a simulation with aerosol-radiation interactions turned off, but the results from this study are based on online simulation of M7. As the current paper dicussses aerosol feedbacks, in which results must come from online simulations, it will not lead to confusion with whether aerosol-radiation interactions in M7 are on or off.

Line 78-80: Since e.g. Grell et al., 2011 (doi:10.5194/acp-11-5289-2011) and Yang et al. 2012 (https://doi.org/10.5194/acp-12-3045-2012) describe the development and implementation of aerosol-meteorology interactions into WRF-Chem, these papers should also be mentioned here and not only application papers. This holds of course also for the other models.

- Reply: We have added the suggested references in the revised manuscript.

- Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled "online" chemistry within the WRF model, Atmos. Environ. 39, 6957–6975, 2005.
- Saide, P. E., Spak, S. N., Carmichael, G. R., Mena-Carrasco, M. A., Yang, Q., Howell, S., Leon, D. C., Snider, J. R., Bandy, A. R., Collett, J. L., Benedict, K. B., de Szoeke, S. P., Hawkins, L. N., Allen, G., Crawford, I., Crosier, J., and Springston, S. R.: Evaluating WRF-Chem aerosol indirect effects in Southeast Pacific marine stratocumulus during VOCALS-REx, Atmos. Chem. Phys., 12, 3045-3064, https://doi.org/10.5194/acp-12-3045-2012, 2012.
- Yang, Q., W. I. Gustafson Jr., Fast, J. D., Wang, H., Easter, R. C., Morrison, H., Lee, Y.-N., Chapman, E. G., Spak, S. N., and Mena-Carrasco, M. A.: Assessing regional scale predictions of aerosols, marine stratocumulus, and their interactions during VOCALS-REx using WRF-Chem, Atmos. Chem. Phys., 11, 11951–11975, doi:10.5194/acp-11-11951-2011, 2011.

Lines 122-123: Please give a reference here.

- Reply: We have added a reference in the revised manuscript: "M6 also use an external assumption which likely cause weaker absorption and ADRF in the atmosphere (Conant et al., 2003).".
- Conant, W.C., Seinfeld, J.H., Wang, J., Carmichael, G.R., Tang, Y., Uno, I., Flatau, P.J., Markowicz, K.M. and Quinn, P.K.: A model for the radiative forcing during ACE-Asia derived from CIRPAS Twin Otter and R/V Ronald H. Brown data and comparison with observations, Jour. Geophy. Res.: Atmos., 108(D23), https://doi.org/10.1029/2002JD003260, 2003.

Lines 125-126: Please try to explain this behavior.

- Reply: We added explanation in the revised manuscript: "This is related to the strong negative forcing at the surface and predicted high concentrations of sulfate by M5 (Gao et al., 2018a)."

Lines 133-134: What is 'other model treatments'?

- Reply: Other model treatments include parameterization of hygroscopicity, including dust or not, etc. To avoid confusion, we change the sentence to: “while discrepancies among models could be resulted from assumptions for mixing state and other model treatments (parameterization of hygroscopicity, mineral dust, etc.).”

Line 156-159 and caption of Fig. 3: The name of the model would be more helpful here.

- Reply: We have added names of models here and Table 1 to describe these models.
- (Table 1: M1: WRF-Chem, Pusan National University; M2: WRF-Chem, University of Iowa; M4: NU-WRF, NASA; M5: RIEMS-Chem, Institute of Atmospheric Physics; M6: RegCCMS, Nanjing University; M7: WRF-CMAQ, University of Tennessee; Gao et al., 2018a).

Line 181: Why are the results for M6 so different?

- Reply: Most other models use WRF as the meteorological model while M6 uses a very different climate model. In additional, lots of parameterization schemes used in M6 are different. For example, other models use a kappa parameterization to describe aerosol hygroscopic growth, while M6 uses a different hygroscopic growth scheme following Kiehl and Briegleb (1993). M6 produces way too high concentrations of sulfate in Beijing. All these factors make M6 very different.

Lines 228-242: These results should be discussed in relation to the results by Curci et al., 2015 (http://dx.doi.org/10.1016/j.atmosenv.2014.09.009) and similar studies.

- Reply: In the revised manuscript, we added comparison with Curci et al., 2015 and other similar studies:
- “In the control case, a homogeneous mixture of inorganic aerosols and BC is assumed. The refractive index of this mixture is estimated using the volume-weighted average of the refractive index of individual component. The size of the mixture is prescribed to be the maximum size of the mixed aerosol components. For example, the size of the mixture of

sulfate and BC is set to be equal to sulfate, assuming a small BC particle sticking to a larger sulfate particle. An additional simulation was conducted with the assumption of external mixing, and the corresponding results are displayed in Fig. 7-9. For external mixing assumption, each aerosol component is considered individually, and the total AOD is calculated as the sum of extinction by each aerosol component. Compared with internal mixing assumption, results from external mixing assumption generally exhibit a weaker (negative) ADRF at the surface (~15%), a stronger (negative) ADRF at TOA (~50%) and a decreased (positive) ADRF in the atmosphere (~30%) (Fig. 9a, 9f, 9k). These reponses of ADRF to aerosol mixing state inferred by this study are consistent with those from Conant et al. (2003). Curci et al. (2015) reported lower AOD with internal mixing assumption than external mixing assumption, because aerosol mass was distributed more to larger particles. As a result, fewer scattering agents are estimated, leading to lower AOD. These differences also suggest that the effects of mixing state on radiative forcing may differ under different treatments. With external mixing assumption, M5 predicts smaller aerosol feedbacks (changes in surface meteorological variables and PM2.5 concentrations, Fig. 8a, 8d, 8g, and 8j) than the estimates with internal mixing assumption. The monthly averaged changes in surface air temperature, wind speed and PM2.5 values are -0.47 °C, -0.03 m/s and 1.5 μg/m3 for the BTH region with internal mixing assumption, while the corresponding values change to -0.46 °C, -0.02 m/s and 1.2 μg/m3 with external mixing assumption. These differences emphasize the important influences of aerosol mixing state on ADRF and aerosol feedbacks. Aerosol mixing states can vary with time and location. Measurements in North China suggest that aerosols are partially internally mixed, and the fraction of internal mixing increased from clean to haze periods (Li et al., 2014)."

- Conant, W.C., Seinfeld, J.H., Wang, J., Carmichael, G.R., Tang, Y., Uno, I., Flatau, P.J., Markowicz, K.M. and Quinn, P.K.: A model for the radiative forcing during ACE-Asia derived from CIRPAS Twin Otter and R/V Ronald H. Brown data and comparison with observations, Jour. Geophy. Res.: Atmos., 108(D23), https://doi.org/10.1029/2002JD003260, 2003.

Lines 253-254: Model names in addition to M5 and M5 would be helpful. Does M1 (which is also WRF-Chem) definitely not include any soil dust? It is possible to use MADE-Sorgam in combination with a dust option. Please clarify this in the paper.

- Reply: All models have the options to include dust, but in some applications, the modelers did not turn this option on. We have changed the sentence to make it clearer: "M5 (RIEMS-Chem) includes all anthropogenic aerosols and dust, sea salt, while the other models except M2 (WRF-Chem, University of Iowa) do not consider natural dust in their model settings."

Lines 285 (Language quality): 'Previous paper': Which previous paper?

- Reply: We have changed "previous paper" to Gao et al. (2018a).

Line 291: What is the reason for this?

- Reply: ADRF at the TOA is the sum of ADRF at the surface (negative) and ADRF inside the atmosphere (positive). the ADRF at the TOA can be either positive and negative, depending on the relative magnitudes of ADRF at the surface and inside the atmosphere. Most of the model results show alternating sign of positive and negative values in the distribution of ADRF at the TOA, in contrast to the consistent negative and positive values of ADRF at the surface and in the atmosphere. This is related to the distribution of predicted relative importance of scattering and absorbing aerosols.

Line 299: Which studies?

- Reply: We have changed the sentence to "Our findings differ from previous studies (Ding et al., 2016; Gao et al., 2015; Gao et al., 2016; Liu et al., 2018; J. Wang et al., 2014; Z. Wang et al., 2014; Wang et al., 2015; Wu et al., 2019; Zhang et al., 2015; Zhang et al., 2018; Zhong et al., 2018)."
-

Caption of Fig. 2: Which month?

- Reply: To avoid confusion, we have added "(January 2010)" into all captions of figures.

Caption of Table 1 and 2: To which area do the results given in the tables refer to?

- Reply: We add Figure S1 in the supplement to display the BTH region (marked with blue) and Beijing (shown using the green arrow). These regions are defined with political boundaries. We also add "(areas marked in Fig. S1)" in captions of Tables

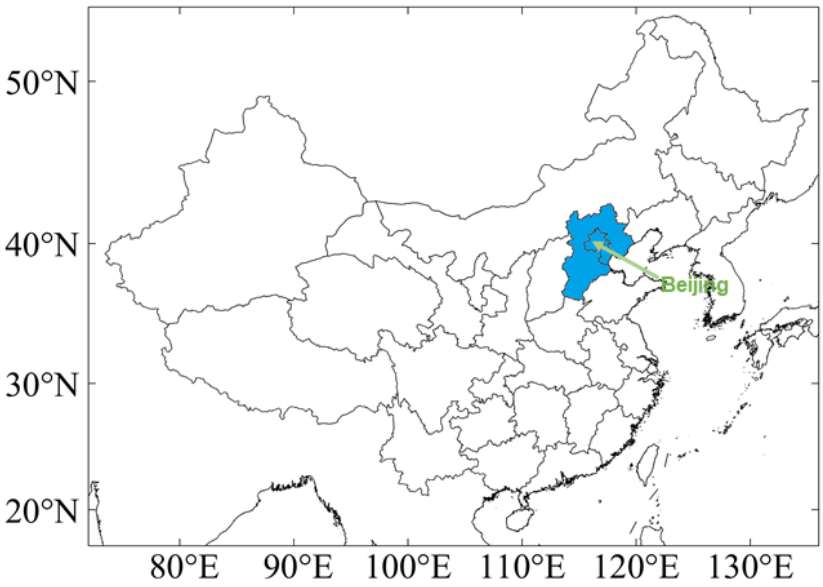

Figure S1. The Beijing-Tianjin-Hebei region is marked with blue and Beijing is shown with green arrow.

The language quality must be improved by consulting a native speaker or a language editing service.

- Reply: We (including a native speaker) have carefully checked the language and improved the quality.

Minor points

Line 101, 175, 188: Start a new paragraph here (and everywhere else, where you start to discuss a new topic).

- Reply: We have changed accordingly.

Lines 104-105, 122, 133 (could be resulted), 216-220, 291, 303-304 and many other lines: Odd language

- Reply: We have changed these sentences to:
- 104-105: "how do aerosol feedbacks change meteorological variables? and how do current models differ in estimating these changes?"
- 122: "It is noticed that M6 predicts lower aerosol optical depth (AOD) than M7 (Gao et al., 2018a), which could partly explain the weaker ADRF estimated by M6. M6 uses an external assumption of aerosol mixing state which likely cause weaker absorption and ADRF in the atmosphere (Conant et al., 2015)."
- 133: "among models could be due to assumptions"
- 216-220: "Concentrations of sulfate and organic aerosol are generally underestimated by most of the participating models, and M4 overestimates the concentrations of organic aerosols (Gao et al., 2018a). These model errors were attributed to the missing multiphase oxidation mechanisms of SO2, and different secondary organic aerosol (SOA) formation mechanisms in these models (Gao et al., 2018a)."
- 291: "The spatial distributions of ADRF at the surface and inside the atmosphere inferred from multiple models are generally consistent, but the spatial distributions of ADRF at the TOA estimiated by these models greatly differ."
- 303-304: "The results indicate the important effect of aerosol mixing state on the estimates of ADRF and aerosol feedbacks, and BC exhibits large contribution to atmospheric heating although it accounts for a small share of mass concentration of PM2.5"

Line 122: 'an external assumption': About what?

- Reply: external assumption of aerosol mixing state. We have changed it in the revised manuscript.

Line 122: Why 'also'? Which other models use also the assumption of external mixing?

- Reply: We have deleted “also” here

Line 169: A word seems to missing here.

- Reply: We have changed the sentence to “Model evaluation of PM2.5 composition in Gao et al. (2018a) reveals that M4 overpredicts the concentrations of organic carbon, which could be one of the reasons for the higher estimated temperature reduction due to aerosols.”

Line 225: A reference should be given here.

- Reply: We have added the following reference: RIEMS-Chem model (M5) (Han et al., 2010)
- Han, Z.: Direct radiative effect of aerosols over East Asia with a regional coupled climate/chemistry model, Meteorologische Zeitschrift, 19(3), pp.287-298, https://doi.org/10.1127/0941-2948/2010/0461, 2010.

Line 282: A reference should be given here.

- Reply: We have added the reference “Topic 3 of MICS-Asia III (Gao et al., 2018a) focuses on understanding how current online coupled air quality models perform in capturing extreme aerosol pollution event in northern China and how aerosols interact with radiation and weather.”

References in Figure captions: 2018a or 2018b? Better mention the models instead of the institutions (eventually not necessary for all figures)

- Reply: We have changed 2018 to 2018a. We have added model names: “(M1: WRF-Chem, Pusan National University; M2: WRF-Chem, University of Iowa; M4: NU-WRF, NASA; M5: RIEMS-Chem, Institute of Atmospheric Physics; M6: RegCCMS, Nanjing University; M7: WRF-CMAQ, University of Tennessee; Gao et al., 2018a). ”

Line 450: Use either complete list of authors (Forkel, R., Balzarini, A., Baró, R., Bianconi, R., Curci, G., Jiménez-Guerrero, P., Hirtl, M., Honzak, L., Lorenz, C., Im, U., Pérez, J. L., Pirovano, G., José, R. S., Tuccella, P., Werhahn, J., and Zabkar, R.: Analysis of the WRF-Chem contributions to AQMEII phase2 with respect to aerosol radiative feedbacks on meteorology and pollutant distributions, Atmos. Environ., 115, 630–645, 2015.) or 'et al' after the third author.

- Reply: Thank you for mentioning. This list was generated automatically with Mendeley. We have updated.

Air Quality and Climate Change, Topic 3 of the Model Inter-Comparison Study for Asia Phase III (MICS-Asia III), Part II: aerosol radiative effects and aerosol feedbacks

Meng Gao[1], Zhiwei Han[2,3], Zhining Tao[4,5], Jiawei Li[2,3], Jeong-Eon Kang[6], Kan Huang[7], Xinyi Dong[8], Bingliang Zhuang[9], Shu Li[9], Baozhu Ge[10], Qizhong Wu[11], Hyo-Jung Lee[6], Cheol-Hee Kim[6], Joshua S. Fu[8], Tijian Wang[9], Mian Chin[5], Meng Li[12], Jung-Hun Woo[13], Qiang Zhang[14], Yafang Cheng[12], Zifa ~~Wang~~[4]Wang[3,10], Gregory R. Carmichael[15]

1 Department of Geography, Hong Kong Baptist University, Hong Kong SAR, China
2 Key Laboratory of Regional Climate-Environment for Temperate East Asia, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China
3 University of Chinese Academy of Sciences, Beijing 100049, China
4 Universities Space Research Association, Columbia, MD, USA
5 NASA Goddard Space Flight Center, Greenbelt, MD, USA
6 Department of Atmospheric Sciences, Pusan National University, Busan, South Korea
7 Department of Environmental Science and Engineering, Fudan University, Shanghai, China
8 Department of Civil and Environmental Engineering, University of Tennessee, Knoxville, TN, USA
9 School of Atmospheric Sciences, Nanjing University, Nanjing, China
10 State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China
11 College of Global Change and Earth System Science, Beijing Normal University, Beijing, China
12 Multiphase Chemistry Department, Max Planck Institute for Chemistry, Mainz, Germany
13 Department of Advanced Technology Fusion, Konkuk University, Seoul, South Korea
14 Ministry of Education Key Laboratory for Earth System Modeling, Center for Earth System Science, Tsinghua 15 University, Beijing, China
15 Center for Global and Regional Environmental Research, University of Iowa, Iowa City, IA, USA
Correspondence to: M. Gao (mmgao2@hkbu.edu.hk), Z. Han (hzw@mail.iap.ac.cn), and G. R. Carmichael (gcarmich@engineering.uiowa.edu)

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 wintertime haze events ~~high aerosol pollution~~ in the North China Plain region ~~during wintertime haze events~~ and evaluates the importance of aerosol radiative and microphysical feedbacks. This paper discusses the estimates of aerosol radiative forcing, aerosol feedbacks, and possible causes for the differences among the participating models. Over the Beijing-Tianjin-Hebei (BTH) region, the ensemble mean of estimated aerosol direct radiative forcing (ADRF) at the top of atmosphere, inside the atmosphere and at the surface are -1.~~9~~1, 7.7~~8.4~~ and -8.8~~10.3~~ W/m^2, respectively. Subdivisions of direct and indirect aerosol radiative forcing confirm the dominant role~~s~~ of direct forcing. During severe haze days (January 17-19, 2010), the averaged reduction in near surface temperature for the BTH region can reach 0.3~~-3.0~~1.6 ºC. The responses of wind speeds at 10 m (WS10) inferred from different models show consistent declines in eastern China. For the BTH region, aerosol-radiation feedback induced daytime changes in $PM_{2.5}$ concentrations during severe haze days range from 6.0 to ~~8.8~~12.9 μg/m^3 (< 6~~.6~~%). Sensitivity simulations indicate the most sensitive parameter for aerosol radiative forcing and feedbacks is the aerosol mixing state, and BC exhibits large contribution to atmospheric heating although it accounts for a small share of mass concentration of $PM_{2.5}$.

1 Introduction

Aerosols change weather and climate via the following pathways: they absorb and scatter solar and thermal radiation to alter the radiative balance of the earth-atmosphere system (*Gao et al., 2019b; Liu et al., 2011; Jia et al., 2018*), which is referred to as direct effects; and, they serve as cloud condensation nuclei (CCN) and/or ice nuclei (IN) to modify cloud properties, which is referred to as indirect effects (*Haywood and Boucher, 2000*). The suppression of cloud convection induced by direct effects of absorbing aerosols is ~~called~~ known as the semi-direct effect (*Huang et al., 2006; Lohmann and Feichter, 2005*). Increases in cloud droplet number can increase cloud albedo for a constant liquid water path (LWP), which is further classified as the first indirect effect or Twomey effect (*Twomey, 1991*). More but smaller cloud droplets

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reduce precipitation intensity but increase cloud lifetime, which is ~~called~~ known as the cloud lifetime or second indirect aerosol effect (*Albrecht, 1989*). In turn, changes in the radiative balance can alter meteorological variables (e.g. temperature, relative humidity, photolysis rate, etc.) and further the transport, diffusion and chemical conversion of trace gases and aerosols, while changes in clouds can affect in-cloud aqueous-phase chemistry and wet deposition of gases and aerosols.

The impacts of meteorology on chemistry have been explicitly treated in chemical transport models (CTMs). For example, temperature modulates chemical reaction and photolysis rates, affects volatility of chemical species, and biogenic emissions, wind speed and direction determine transport and mixing, and precipitation influences wet deposition (*Baklanov et al., 2014*). However, due to the complexity of these processes and lack of computational resources, the influences of atmospheric compositions on weather and climate have been generally ignored in previous CTMs (*Baklanov et al., 2014*). Studies examining how aerosols interact with weather/climate remain uncertain and limited. ~~Until r~~ Recently, with the rapid development of coupled meteorology and chemistry models, many new studies have been conducted to investigate the aerosol direct and indirect effects and feedbacks (*Baklanov et al., 2017; Forkel et al., 2015; Gao et al., 2016, 2017*; *Grell et al., 2005; Han et al., 2010; Huang et al., 2016; Jacobson et al., 2007; Saide et al., 2012; Wang et al., 2014; Yang et al., 2011; Zhang et al., 2010*). In highly polluted regions like Asia, aerosol feedbacks can be particularly important (*Gao et al., 2016, 2017*). High concentrations of aerosols would enhance the stability of boundary layer due to reductions in radiation that reach the surface, which in turn can cause further increases in $PM_{2.5}$ concentrations (*Ding et al., 2016; Gao et al., 2016*).

Aerosol feedbacks during haze events in China have been explored using multiple online coupled meteorology-chemistry models, including WRF-Chem (the Weather Research Forecasting model coupled with Chemistry, *Chen et al., 2013, 2018; Gao et al., 2016, 2017, 2019a; Liu et al., 2015*), WRF-CMAQ (Community Multiscale Air Quality, *Wang et al., 2014*). Nevertheless, large uncertainties remain in the modelling of these processes, due to the lack of direct observational constraints and challenges in predicting aerosol compositions. Thus, the inter-comparison of coupled meteorology-chemistry models is of great significance to better understand the differences, causes, and uncertainties within these processes.

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Topic 3: air quality and climate change within the Model Inter-Comparison Study for Asia Phase III (MICS-Asia phase III) was initialized to address these issues (*Gao et al., 2018a*). Results from seven applications of fully online coupled meteorology-chemistry models using harmonized emission and chemical boundary conditions were submitted to this topic (*Gao et al., 2018a*). These model applications include two applications of WRF-Chem by different institutions, two applications of the National Aeronautics and Space Administration (NASA) Unified WRF (NU-WRF) model with different model resolutions, one application of the Regional Integrated Environment Modeling System with Chemistry (RIEMS-Chem, *Han et al., 2010*), one application of the coupled Regional Climate Chemistry Modeling System (RegCCMS), and one application of the coupled WRF-CMAQ model (*Gao et al., 2018a*). More detailed information of the participating models, and information about how the experiments were designed and how models perform~~l evaluations~~ have been archived in *Gao et al. (2018a)*. In this paper, we analyze the results from the participating models to address the following questions: (1) how large is the aerosol radiative forcing during winter haze in China and how differently are models estimating it? (2) ~~to what extend do~~how do aerosol feedbacks change meteorological variables? and how ~~differently are~~do current models differ in estimating these changes? (3) ~~to what extent~~how do aerosol feedbacks contribute to the evolution of high aerosol concentrations during winter haze episodes? and what are the best estimates from different models? And (4) what are the major causes of the differences among the models? Sect. 2 describes briefly how the experiments were designed and how models perform. Sect. 3~~2~~ presents the estimates of aerosol direct radiative forcing inferred from multiple models, including the separation of direct and indirect effects. In Sect. ~~2~~4, we discuss the impacts of aerosol-radiation feedbacks on meteorological variables and $PM_{2.5}$ concentrations. Sect. ~~4~~ 5 illustrates the sensitivity of aerosol forcing to different processes in the model, and the summary is presented in Sect. ~~5~~6.

2 Overview of MICS-Asia III Topic 3

The participants were requested to use common emissions to simulate air quality during January 2010 and submit requested model variables. The participating models include one

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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: 45 and 15 km horizontal resolutions) of the National Aeronautics and Space Administration (NASA) Unified WRF (NU-WRF; *Peters-Lidard et al., 2015*) model by the Universities Space Research Association (USRA) and NASA's Goddard Space 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) (Table 1). A new Asian emission inventory was developed for MICS-Asia III by integrating state-of-the-art national or regional inventories (*Li et al., 2017*), which was provided to all modeling groups, along with biogenic emissions, biomass burning emissions, etc. Simulations from two global chemical transport models (e.g., GEOS-Chem (The Goddard Earth Observing System Model-Chemistry) and MOZART (Model for OZone And Related chemical Tracers)) were provided as boundary conditions for MICS-Asia III. The entire month of January 2010 was simulated and covered by one single simulation for each participating model. Comprehensive model evaluations indicate that all models could capture the observed near-surface temperature and water vapor mixing ratio, but overestimated near-surface wind speeds. These models were able to represent the observed daily maximum downward shortwave radiation, particularly low values during haze days. The observed variations of air pollutants, including SO_2, NO_x, CO, O_3, $PM_{2.5}$, and PM_{10}, were reproduced by these models. However, large differences in the models were found in the predicted $PM_{2.5}$ chemical compositions.

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~~2~~ 3 Aerosol Direct and Indirect Forcing

Fig. 1 shows the monthly mean all-sky aerosol direct radiative forcing (ADRF) over China. The spatial distributions of ADRF at the surface and inside the atmosphere inferred from

multiple models are generally consistent, with the largest values in eastern and southwestern China. Over the Beijing-Tianjin-Hebei (BTH) region (areas marked in Figure S1), ~~M5 and~~ M7 report~~s~~ the highest ADRF at the surface (~~-16.7 and -~~17.0 W/m^2), and the ~~greatest~~ largest ADRF inside the atmosphere (~~10.1 and~~ 14.6 W/m^2) (**Table ~~1~~2**). M6 shows the lowest ADRF both at the surface and inside the atmosphere (-3.6 and 3.6 W/m^2) (**Table ~~1~~2**). It is noticed that M6 predicts lower aerosol optical depth (AOD) than ~~M5 and~~ M7 (*Gao et al., 2018a*), which could partly explain the weaker ADRF estimated by M6.~~,~~ ~~and~~ M6 ~~also~~ uses an external assumption of aerosol mixing states, which is likely to cause weaker absorption and ADRF in the atmosphere (*Curci et al., 2015*). However, the reported ADRF at the top of the atmosphere (TOA) vary widely, and no consensus is reached on whether the forcing is positive or negative. The spatial pattern of ADRF at the TOA inferred from M5 are consistently negative across the modeling domain, while the results inferred from other models are patchy with positive values to the north or to the southwest (**Fig. 1**). Consistent negative ADRF at the TOA estimated by M5 is related to the strong negative forcing at the surface and the predicted high concentrations of sulfate by M5 (*Gao et al., 2018a*). Over the BTH region, ~~suggested~~ simulated ADRF at the TOA range from -2.6~~7.6~~ to 0.2 W/m^2 (**Table ~~2~~1**). *Li et al. (2010)* reported observation-based estimates of aerosol radiative forcing across China to be 0.3±1.6 at the TOA. *Chung et al. (2005)* and *Chung et al. (2010)* estimated the forcing over south Asia to be -2.9 W/m^2 and -3.6 W/m^2 at the TOA, respectively. The magnitudes of the model ~~modelled~~ estimated aerosol radiative forcing values are generally in line with these estimates inferred from observations, while discrepancies among models could be ~~resulted from~~due to assumptions ~~for~~ of aerosol mixing state~~s~~ and other model treatments (parameterization of hygroscopicity, soil dust, etc.). The discussions on how different model treatments affect the results of ADRF is provided in Sect. ~~4~~5.

Fig. 2 exhibits the ensemble mean of monthly averaged ADRF at the TOA, inside the atmosphere and at the surface. Elevated forcing inside the atmosphere and at the surface are mainly located in east China. However, the ensemble mean of forcing at the TOA over the ocean is slightly higher than that over the land. Over the BTH region, the ensemble mean of ADRF at the TOA, inside the atmosphere and at the surface are -1.~~9~~1, ~~8.4~~7.7 and ~~-8~~10.~~8~~3 W/m^2, respectively. ~~When only haze days are considered, these values increase to -2.4, 19.0 and -21.4~~

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~~W/m^2, respectively.~~ In winter, the aerosol radiative forcing in China is largely contributed by the power sector and residential sector, but with different signs of the contribution (*Gao et al., 2018b*).

M4 and M5 further provide subdivision of direct and indirect aerosol radiative forcing. As listed in **Table ~~2~~3**, although the magnitudes of forcing estimated by M4 and M5 differ from each other, the dominant roles of direct forcing are consistent. Over ~~north~~ North China and during wintertime, aerosol indirect forcing is negligible due to the lack of water vapor and the stable weather conditions.

~~3~~ 4 Impact of aerosol feedbacks on meteorological variables and $PM_{2.5}$ concentrations

~~Here we analyze results for simulations on the time period, January 2010, of a heavy haze event.~~ When extreme haze events happen, high aerosol loadings can reduce significantly the shortwave radiation reaching the surface, modifying near-surface temperature (*Gao et al., 2017*). **Fig. 3** displays the aerosol-radiation feedback induced changes in temperature at 2 m (T2) from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (Table 1: M1: WRF-Chem, Pusan National University; M2: WRF-Chem, University of Iowa; M4: NU-WRF, NASA; M5: RIEMS-Chem, Institute of Atmospheric Physics; M6: RegCCMS, Nanjing University; M7: WRF-CMAQ, University of Tennessee; *Gao et al., 2018a*). ~~All~~ The participating models show different degrees of reductions in T2~~, but the magnitudes differ~~. M5 exhibits the ~~largest areas where T2 is reduced~~most widespread areas with reductions, which include ~~northeastern~~ Northeastern China~~,~~. However,~~while~~ significant reductions in T2 inferred from other models are mainly concentrated in southern China (**Fig. 3**). In Beijing (areas marked in Figure S1), the monthly averaged reductions in T2 from multiple models range from 0 to~~-~~ 0.7~~1~~ ºC, with the greatest changes calculated from ~~M5~~ M4 (**Table ~~1~~2**). In the Beijing-Tianjin-Hebei (BTH) region, similar magnitudes (0~~-1.3~~0.8 ºC) are found. When only severe haze days (January 17-19) are considered, the averaged reductions in T2 for Beijing (0.1~~-3.2~~1.7 ºC) and the BTH region (0.3-1.6~~3.0~~ ºC) are further enhanced (**Table ~~3~~4**). In terms of aerosol-radiation feedback induced temperature reduction, M1 and M2 generally report similar magnitudes, which are

lower than M4, M5 and M7. Model evaluations of $PM_{2.5}$ composition in *Gao et al. (2018a)* reveals that M4 overpredicts ~~strong scattering~~the concentrations of organic carbon, which could be one of the reasons for the higher estimated ~~temperature~~ reductions in T2 due to aerosols.

Pronounced decreases in water vapor at 2 m (Q2) are mostly located in southern China (**Fig. 4**), where water vapor is more abundant due to the proximity to the sea. During extreme haze days, the aerosol-radiation feedback induced decreases in Q2 in the BTH region from multiple models range from 0.07 to 0.~~5~~29 g/kg, with the lowest estimate from M1 and the highest from ~~M6~~ M4 (**Table ~~3~~4**).

The responses of wind speeds at 10 m (WS10) inferred from different models are generally consistent, displaying decreases in eastern China except M6 **(Fig. 5)**. In the BTH region, the monthly mean aerosol-radiation feedback induced decreases in WS10 range from 0.02 to 0.09 m/s (**Table ~~1~~2**), and more pronounced reductions are suggested by M4~~, M5~~ and M7 (**Fig. 5**).

Because of aerosol-radiation feedback, most models report that surface $PM_{2.5}$ concentrations are ~~generally~~ enhanced in China, with the exception of M6 (**Fig. 6**). It is also noteworthy that $PM_{2.5}$ concentrations decrease in the Gobi desert and Taklimakan desert of western China in M5 and M2, which is caused by the decreased wind speed near the surface due to the weakened downward transport of momentum from upper layer above boundary layer to the surface (*Han et al., 2013*). The~~For M6 the increases~~ changes of $PM_{2.5}$ concentrations suggested by M6 are patchy over east China, with decreases to the north and to the southwest. The monthly mean $PM_{2.5}$ are enhanced by 0.1-1.6~~4~~ μg/m^3 for Beijing, and by 0.8-2.2~~4.4~~ μg/m^3 for the BTH region. The enhancement fractions are generally below 2~~.7~~% for Beijing, and below 4~~7.8~~% for the BTH region (**Table ~~1~~2**).

To further understand how aerosol-radiation feedback contributes to the formation of haze event, we calculate the mean increase ~~for~~ during extreme haze days (January 17-19). For the BTH region, the contribution of aerosol-radiation feedback to $PM_{2.5}$ concentrations are lower than ~~6~~4%, and the enhancement are below 8.5 μg/m^3. *Gao et al. (2017)* demonstrates that the aerosol-radiation feedback induced changes in $PM_{2.5}$ are negligible during nighttime, so we further calculate daytime mean changes, as listed in **Table ~~3~~4**. For the BTH region, M2 reports

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the ~~highest~~ largest enhancement (12.9 μg/m^3) of $PM_{2.5}$ concentrations during daytime. ~~Except M6, o~~Other models, except M6, report similar magnitudes of ~~the~~ enhancement, ranging from ~~6.0~~5.3 to ~~8.8~~6.6 μg/m^3. The enhancement fraction ~~is still not more~~remains less than 6.~~6~~% for the BTH region, and below 8.3% for Beijing. **Table ~~3~~ 4** also displays the maximum enhancement of $PM_{2.5}$ during haze days over~~for~~ the BTH region. M7 suggests the largest $PM_{2.5}$ enhancement (up to 60.9 μg/m^3), followed by M2 (up to 55.4 μg/m^3)~~, and M5 (41.2 μg/m^3)~~. Other three models, M1, M4, M5, and M6 indicate the aerosol-radiation induced increase in $PM_{2.5}$ can reach up to more than 20 μg/m^3 in the BTH region (**Table ~~3~~4**).

~~These results can be compared to previous studies.~~ The contributions of aerosol-radiation feedback to haze formation in China have been investigated in many previous studies (*Ding et al., 2016; Gao et al., 2015; Gao et al., 2016; Liu et al., 2018; J. Wang et al., 2014; Z. Wang et al., 2014; Wang et al., 2015; Wu et al., 2019; Zhang et al., 2015; Zhang et al., 2018; Zhong et al., 2018*), but the reported values ~~partly~~ diverge. *Ding et al. (2016)*, *J. Wang et al. (2014)* and *Zhong et al. (2018)* indicate that the aerosol radiative effects can increase $PM_{2.5}$ by more than 100 μg/m^3 or +70%. *Gao et al. (2015), Z. Wang et al. (2014), Wang et al. (2015),* and *Zhang et al. (2018)* suggest that the contributions are generally within the range of 10-30%. These ~~studies~~ reports are different from this study~~ours~~ in terms of study~~time~~ periods, region, ~~emissions and resulting aerosol~~and pollution levels. Most of previous reports focused on the January 2013 haze episodes (*J. Wang et al., 2014*), ~~For example,~~while the monthly mean concentrations of $PM_{2.5}$ ~~level~~ in January 2010 are ~~about~~ nearly 50% lower than that ~~in~~ of January 2013. ~~The above studies also differed in the assumptions and treatments for aerosol properties and mixing state.~~ According to the findings~~results from multiple models~~ in this study, the contribution of aerosol-radiation feedback to haze formation during ~~this time period~~January 2010 are generally below 10%.~~–~~ Uncertainties still remain as suggested by~~resulting from~~ the errors in the simulated chemical compositions (*Gao et al., 2018a*). ~~As suggested in model evaluation,~~ Concentrations of ~~s~~sulfate and organic aerosol ~~concentrations~~ are generally underestimated by most of the participating models ~~in this study~~, ~~except that~~and M4 overestimates the concentrations of organic aerosols (*Gao et al., 2018a*). These model errors were attributed to the missing multiphase oxidation mechanisms of sulfate ~~SO$_2$~~, and different treatments of secondary organic aerosol (SOA) formation ~~mechanisms~~ in these models (*Gao*

273 *et al., 2018a*).

274

275 ## 4 5 Sensitivity to Different Processes

276 To ~~further~~ explore the potential causes for the differences among models, and the major~~in~~
277 factors that influence~~influencing the~~ aerosol-radiation feedback, several sensitivity simulations
278 were conducted with~~using~~ the RIEMS-Chem model (M5) (*Han et al., 2010*). These simulations
279 aim to examine the effects of ~~focusing on the effects of aerosols~~ mixing states of aerosols,
280 hygroscopic growth, black carbon and ~~mineral~~ soil dust.

281

282 4 5.1 Aerosol mixing states

283 In the control simulation, inorganic aerosols and BC are assumed to be internally mixed to form
284 a homogeneous mixture. The refractive index of this mixture is estimated using the volume-
285 weighted average of the refractive index of individual component. The size of the mixture is
286 prescribed to be the maximum size of the mixed aerosol components. For example, the size of
287 the mixture of sulfate and BC is set to be equal to the size of sulfate, assuming a small BC
288 particle sticking to a larger sulfate particle.
289 An additional simulation was conducted with the aerosols were treated as externally mixed,
290 and the corresponding results are displayed in **Fig. 7-9**. For external mixing assumption, each
291 aerosol component is considered individually, and the total AOD is calculated as the sum of
292 extinction by each aerosol component. Compared with the results with internal mixing
293 assumption, results with external mixing assumption generally exhibit a weaker (negative)
294 ADRF at the surface (~15%), a stronger (negative) ADRF at TOA (~50%) and a decreased
295 (positive) ADRF in the atmosphere (~30%) (**Fig. 9a, 9f, 9k**). These responses of ADRF to the
296 assumption of aerosol mixing states are consistent with *Conant et al. (2003)*. However, *Curci*
297 *et al. (2015)* reported lower AOD with internal mixing assumption than with external mixing
298 assumption. In *Curci et al. (2015)*, aerosol mass was distributed more to larger particles. As a
299 result, fewer scattering agents and lower AOD were estimated.
300 Aerosol feedbacks estimated by M5 also tend to be weaker with external mixing assumption
301 than with internal mixing assumption (changes in surface meteorological variables and $PM_{2.5}$

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concentrations, **Fig. 8a, 8d, 8g, and 8j**). The monthly averaged changes in T2, WS10 and $PM_{2.5}$ are -0.6 °C, -0.04 m/s and 2.2 μg/m^3 for the BTH region with internal mixing assumption, while the corresponding values change to -0.6 °C, -0.03 m/s and 1.8 μg/m^3, with external mixing assumption. These differences emphasize the important influences of aerosol mixing states on the estimates of ADRF and aerosol feedbacks. However, aerosol mixing states are also varying with time and location. Measurements in North China suggest that aerosols are partially internally mixed, and the fraction of internal mixing increased from clean to haze periods (*Li et al., 2014*).

~~A simulation was run with the assumption of external mixing (results discussed above applied the assumption of internal mixing), and the corresponding results are displayed in **Fig. 7-9**. The simulation with the assumption of external mixing shows weaker (30% smaller) ADRF at the surface, TOA and in the atmosphere (**Fig. 9a, 9f and 9k**), resulting in smaller changes in surface meteorological variables and $PM_{2.5}$ concentrations (**Fig. 8a, 8d, 8g, and 8j**). For example, the monthly mean maximum changes in air temperature, relative humidity, wind speed and $PM_{2.5}$ values are -2.7°C, +3%, -0.24m/s and 16 μg m^{-3}, respectively in the southern Huabei province from the simulation with internal mixing, whereas the corresponding changes from external mixing assumption are -1.4°C, +2%, -0.12m/s and 8 μg m^{-3}, respectively. These differences demonstrate the significant impact of aerosol mixing state on the ADRF and the aerosol-radiation feedback. It should also be emphasized that the aerosol mixing state can vary with time and location. Some previous measurements in the Huabei Plain exhibit that aerosols are partially internally mixed and the fraction of internal mixing could be increasing from clean to haze period (*Li et al., 2014*).~~

45.2 Hygroscopic growth

Given the appreciable~~ important~~ effect of aerosol hygroscopic growth on ADRF (*Li et al., 2014*), another simulation was conducted with decreased relative humidity (RH). In this simulation, ~~by~~ FNL nudging was applied above boundary layer to reduce RH~~. Such~~ This perturbation of RH was based on the fact that M5 overestimates~~predicted higher~~ relative humidity (water vapor mixing ratio) ~~than the observations~~ (*Gao et al., 2017*). ~~The simulation~~With reduced RH, ~~with reduced RH produces~~ lower values of AOD (**Fig. 7f**) and

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weaker ADRF at the surface and TOA (**Fig. 9e, 9j, and 9o**, about ~~10~~15% lower) are found, mainly because of ~~the decreased relative humidity and weaker~~suppressed hygroscopic growth under lower relative humidity.

~~4~~5.3 Soil dust and sea salt

M5 (RIEMS-Chem) includes ~~all anthropogenic~~naturally emitted ~~aerosols and~~soil dust and~~,~~ sea salt, while the other models except M2 (WRF-Chem, University of Iowa) do not consider soil~~natural~~ dust in their model settings. In an additional sensitivity simulation, soil dust and sea salt emissions were turned off in M5 to examine the influence on ADRF and aerosol feedbacks (**Fig. 9d, 9l and 9n**). In January 2010, significant amounts of soil dust were emitted from the Taklimakan desert, influencing wide areas of China. M5 estimates that the monthly mean ADRF at the surface due to dust and sea salt is about -12 W/m^2 over the Taklimakan desert, -4~-6 W/m^2 in the middle reaches of the Yellow River and the Yangtze River Delta, and about -2~-4 W/m^2 over the BTH region. Over the BTH region, the contribution of dust and sea salt aerosols to total ADRF can reach 5~10%. Table 2 illustrates that M5 predicts the largest (negative) radiative forcing at TOA over the BTH region. The above analyses with reduced relative humidity and perturbations in dust and sea salt suggest that the inclusion of dust and overprediction of relative humidity by M5 are important reasons.

~~A simulation with dust aerosol excluded was conducted and the results show the dust aerosol contributes to total $PM_{2.5}$ concentration and ADRF in parts of central and northeast China, especially in the middle reaches of the Yellow River with the ADRF by dust at the surface contribute up to -6 W m^{-2} in terms of monthly mean (**Fig. 9d, 9l, and 9n**), which indicates the nonnegligible role of dust even in winter. Both the overprediction of relative humidity (water vapor) and the inclusion of mineral dust can partly explain the relatively stronger ADRF from M5 compared with other models.~~

~~4~~5.4 The effect of BC

Two sets of simulations, namely without BC and with doubled BC concentrations, were conducted to examine the influences of BC on aerosol radiative forcing and feedbacks. In the control simulation, the aerosol induced changes in monthly T2, WS10 and $PM_{2.5}$ are -0.6 °C, -

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0.04 m/s and 2.2 μg/m^3 for the BTH region, respectively. When BC is not included (only scattering aerosols and dust), the corresponding aerosol induced changes are -0.5 °C, -0.02 m/s and 1.0 μg/m^3, respectively. When BC concentrations are doubled, these values change to -0.7 °C, -0.05 m/s and 3.2 μg/m^3, respectively. The comparison between the control case and two additional sensitivity cases indicates that the changes caused by BC are comparable to those by scattering aerosols. The contribution of BC to aerosol feedbacks can reach up to 40~50%. It is also found that the influence of BC on aerosol feedbacks with internal mixing assumption is larger than that with external mixing assumption (Figure not shown).

Large uncertainties still remain in the estimates of the role of BC in aerosol feedbacks relative to scattering aerosols. *Gao et al. (2016)* suggested that the impacts of BC on boundary layer height and $PM_{2.5}$ concentrations can account for as high as 60% of the total aerosol feedbacks in the North China Plain at 2 p.m., although it only accounts for a small share of PM in terms of mass concentration. *Qiu et al. (2017)* indicated that $PM_{2.5}$ concentrations averaged over the North China Plain increased by 16.8% and 1.0% due to scattering aerosols and BC, respectively. It should be noted that most participating models, including RIEMS-Chem, tend to underpredict the total mass concentrations of scattering aerosols (inorganic and organic aerosols) by up to a factor of two over the study period, leading to overestimation of the contribution of BC.

~~To identify the effect of BC, two simulations without BC and with doubled BC concentrations were conducted. When BC is not included, the ADRF in the atmosphere decreases largely (**Fig. 9g**), indicating the strong absorbing effect of BC. The ADRF at the surface changes by about 10% (**Fig. 9b**). The monthly mean maximum changes in air temperature, relative humidity, wind speed and $PM_{2.5}$ values in this case are -2.2°C, +3.5%, -0.18m/s and 10 μg m^{-3}, respectively, in the southern Huabei region. When BC concentrations are doubled, the corresponding values are -3.0°C, +2.0%, -0.27m/s and 18 μg m^{-3}, respectively. The comparison with the changes in the base case (the corresponding values are -2.7°C, +3%, -0.24m/s and 16 μg m^{-3}, respectively) indicates that the effect of BC is smaller than that due to other scattering aerosols (inorganic and organic aerosols), and the percentage contribution by BC to the total feedback could be in a range of 20-30%. It is also found that the effect of BC under internal~~

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~~mixing condition is larger than that under external mixing. Gao et al. (2016) demonstrates that the impacts of BC on meteorology and $PM_{2.5}$ can account for as high as 60% of the total aerosol feedbacks, although it is not of great significance in terms of mass concentration.~~

~~The above sensitivity simulations suggest the importance of mixing state assumption for ADRF and feedback and the potentially dominant role of scattering aerosols over absorbing aerosols in aerosol radiative effect during haze periods.~~

~~5~~ 6 Summary

Topic 3 of MICS-Asia III (*Gao et al., 2018a*) focuses on understanding how current online coupled air quality models perform in capturing extreme aerosol pollution event in ~~northern~~ North China and how aerosols interact with radiation and weather. Seven applications of different online coupled meteorology-chemistry models were involved in this activity. ~~Previous paper,~~*Gao et al. (2018a)* has demonstrated that main features of the accumulation of air pollutants are generally well represented, while large differences in the models were found in the predicted $PM_{2.5}$ chemical compositions ~~(*Gao et al., 2018a*)~~. These inconsistencies~~y~~ would lead to differences in estimated ADRF and aerosol feedbacks.

The spatial distributions of ADRF at the surface and inside the atmosphere inferred from multiple models are generally consistent, ~~while~~ but the spatial ~~pattern~~ distributions of ADRF at the TOA estimated by these models greatly differ. Over the BTH region, the ensemble mean of ADRF at the TOA, inside the atmosphere and at the surface are -1.~~9~~1, 7.7~~8.4~~ and -~~8~~10.~~8~~3 W/m^2, respectively. Subdivisions of direct and indirect aerosol radiative forcing confirm the dominant roles of direct forcing.

During severe haze days (January 17-19), the averaged reduction in T2 for the BTH region can reach 0.3-1.6~~3.0~~ ºC. The responses of wind speeds at 10 m (WS10) inferred from different models show consistent declines in eastern China. For the BTH region, aerosol-radiation feedback induced changes in daytime $PM_{2.5}$ range from 5.3~~6.0~~ to ~~8.8~~12.9 μg/m^3 (< 6.~~6~~%). Our findings differ from previous studies~~–~~ (*Ding et al., 2016; Gao et al., 2015; Gao et al., 2016; Liu et al., 2018; J. Wang et al., 2014; Z. Wang et al., 2014; Wang et al., 2015; Wu et al., 2019; Zhang et al., 2015; Zhang et al., 2018; Zhong et al., 2018*) in terms of study~~time~~ period, region

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and pollution levels ~~emissions,~~. ~~for example,~~ The monthly mean concentrations of $PM_{2.5}$ ~~level~~ in January 2010 (current study period) are about 50% lower than those ~~at~~ in January 2013. Sensitivity simulations were conducted with ~~using~~ the RIEMS-Chem model (M5) to understand the influences of aerosols mixing states, hygroscopic growth, black carbon and ~~mineral~~ soil dust. The results indicate the important effect of aerosol mixing states on the estimates of ADRF and aerosol feedbacks. ~~most sensitive parameter for ADRF and feedback is the aerosol mixing state, and~~ It was also found that BC exhibits large contribution to atmospheric heating, but uncertainties remain in estimating its contribution given the fact that the observed aerosol chemical components were not perfectly simulated. ~~although it accounts for a small share of mass concentration of $PM_{2.5}$,~~ *Huang et al. (2015)* separated the contributions of different aerosol components to aerosol direct radiative forcing, highlighting the roles of BC and sulfate. Future studies are also needed to separate the effects of other aerosol components, including sulfate, on aerosol feedbacks.

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Author Contributions

M.G., Z.H., and G.R.C. designed the study, and M.G. processed and analyzed the data. M.G., Z.H., and G.R.C. wrote the paper with inputs from all other authors.

Data availability

The measurements and model simulations data can be accessed through contacting the corresponding authors.

Competing interests

The authors declare that they have no conflict of interests.

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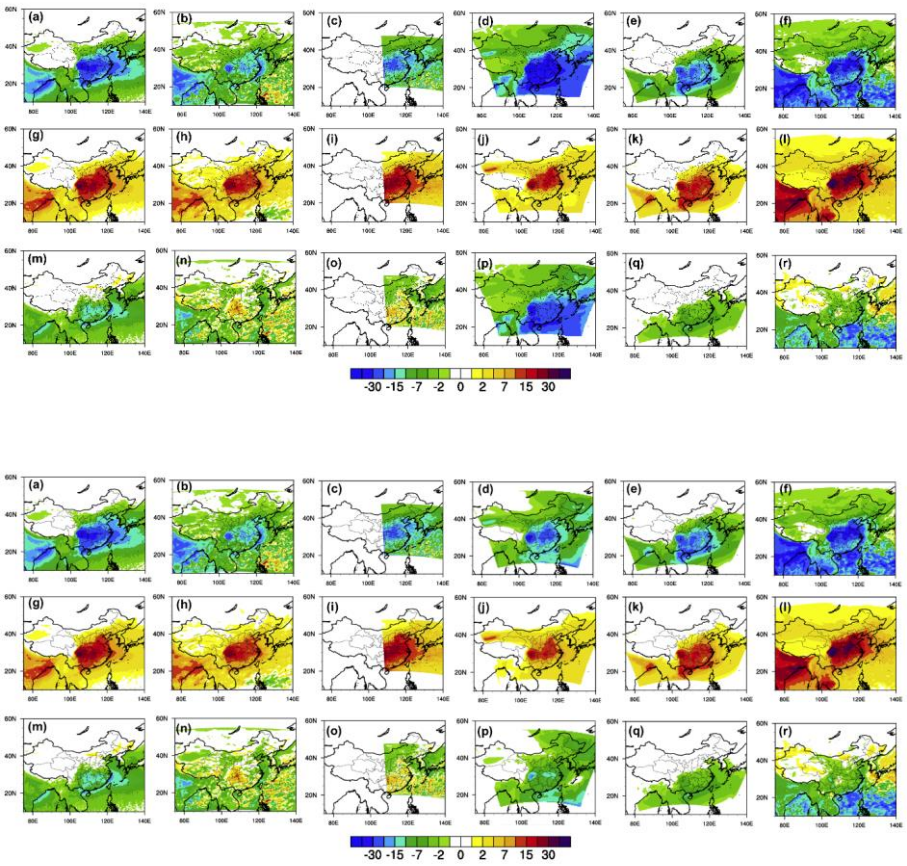

Figure 1. Monthly (January 2010) mean aerosol direct radiative forcing at the surface, inside the atmosphere and at the top of the atmosphere inferred from M1 (a, g, m), M2 (b, h, n), M4 (c, i, o), M5 (d, j, p), M6 (e, k, q), M7 (f, l, r) (M1: WRF-Chem, Pusan National University; M2: WRF-Chem, University of Iowa; M4: NU-WRF, NASA; M5: RIEMS-Chem, Institute of Atmospheric Physics; M6: RegCCMS, Nanjing University; M7: WRF-CMAQ, University of Tennessee; *Gao et al., 2018a*)

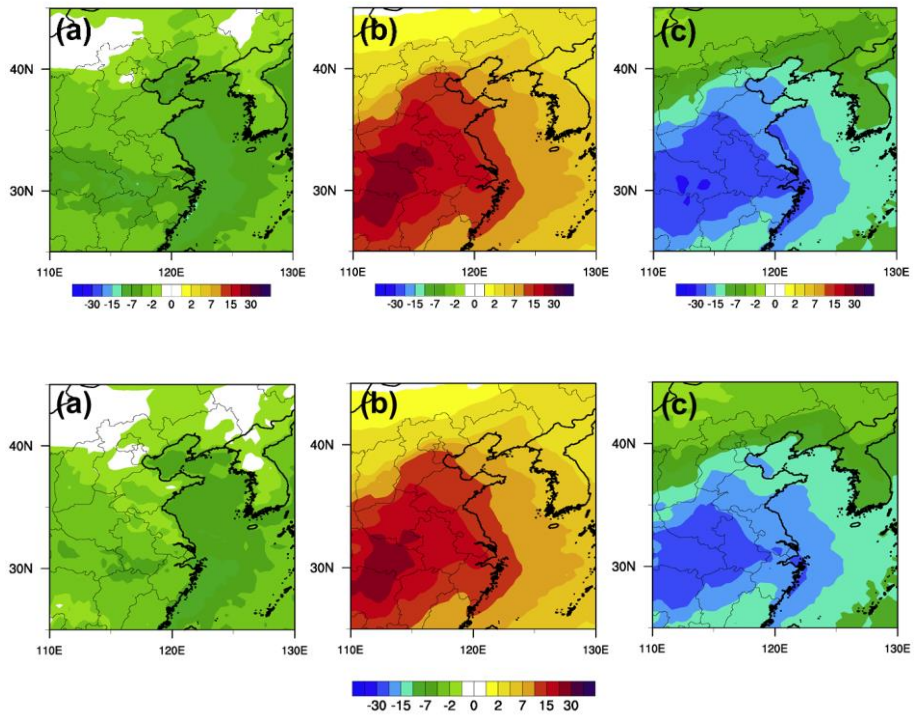

Figure 2. Ensemble mean of monthly (January 2010) mean aerosol direct radiative forcing at the top of the atmosphere (a), inside the atmosphere (b) and at the surface (c)

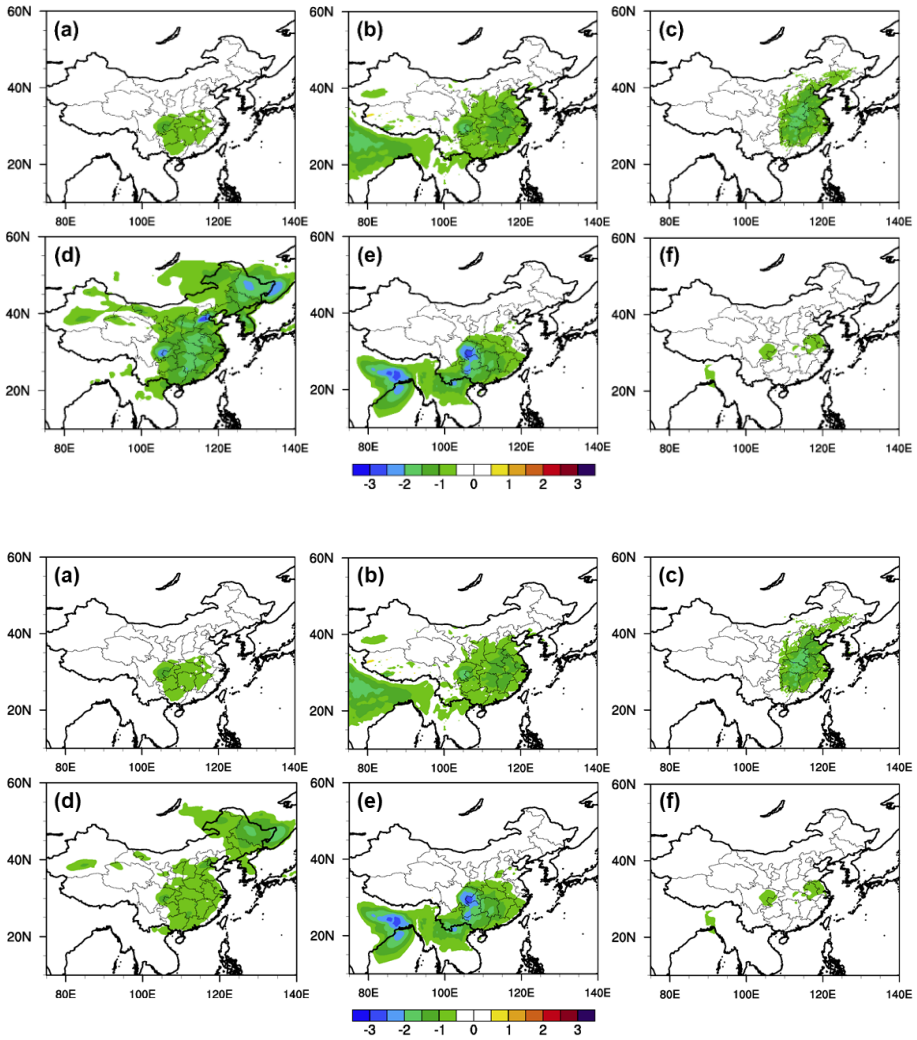

Figure 3. Monthly (January 2010) mean changes in temperature at 2 m (T2, ºC) due to aerosol radiative effects from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (M1: Pusan National University; M2: University of Iowa; M4: NASA; M5: Institute of Atmospheric Physics; M6: Nanjing University; M7: University of Tennessee; *Gao et al., 2018a*)

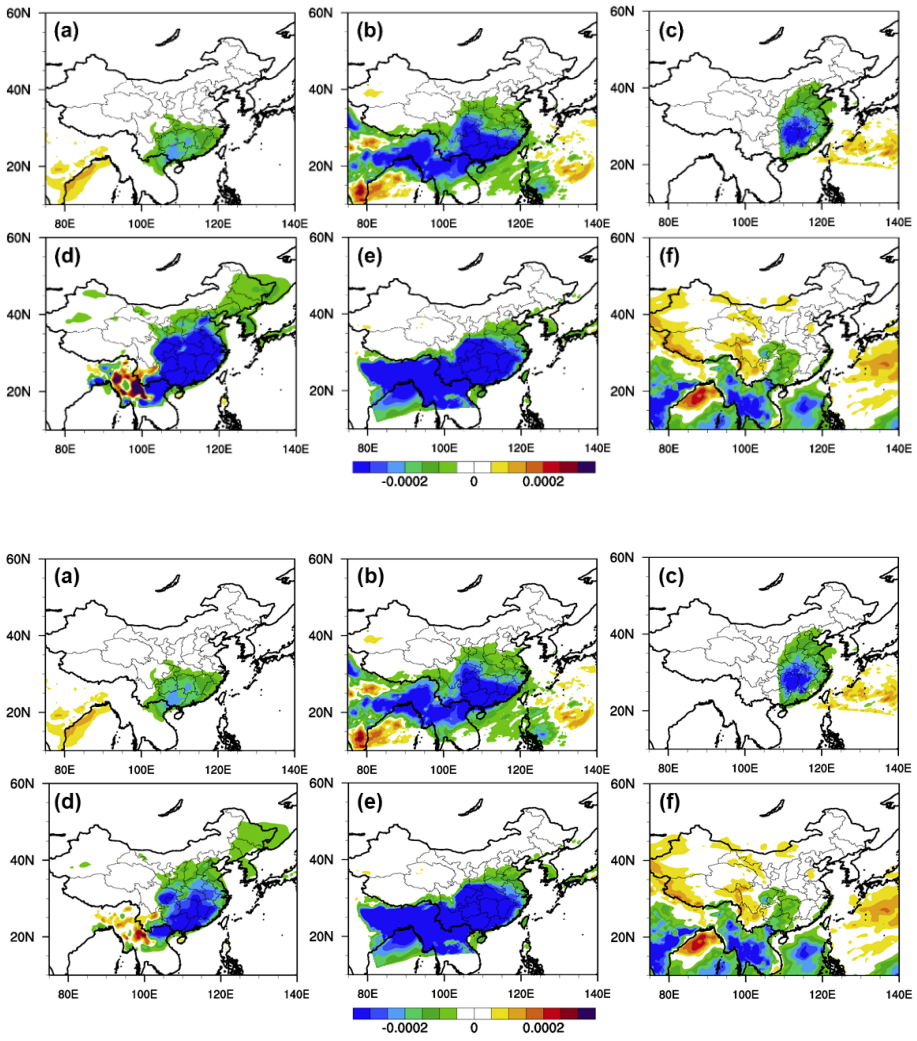

Figure 4. Monthly (January 2010) mean changes in water vapor at 2 m (Q2, kg/kg) due to aerosol radiative effects from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (M1: Pusan National University; M2: University of Iowa; M4: NASA; M5: Institute of Atmospheric Physics; M6: Nanjing University; M7: University of Tennessee; *Gao et al., 2018a*)

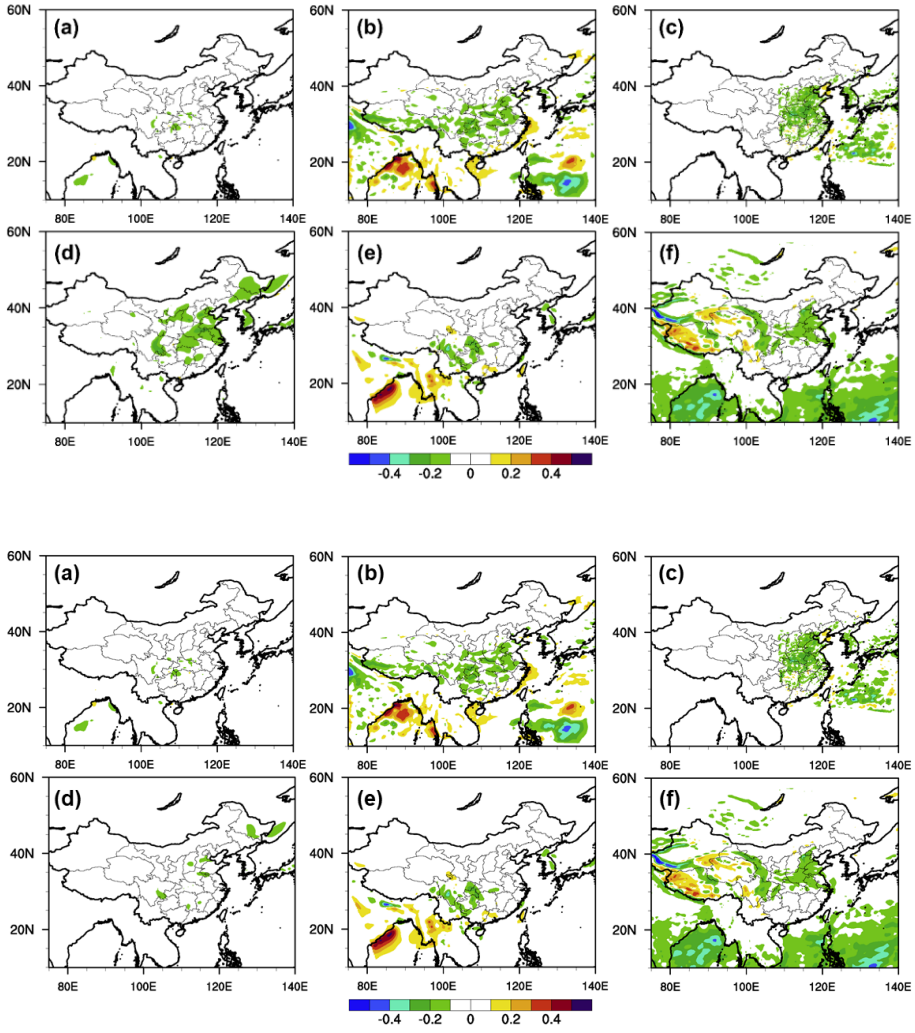

Figure 5. Monthly (January 2010) mean changes in wind speeds at 10 m (WS10, m/s) due to aerosol radiative effects from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (M1: Pusan National University; M2: University of Iowa; M4: NASA; M5: Institute of Atmospheric Physics; M6: Nanjing University; M7: University of Tennessee; *Gao et al., 2018a*)

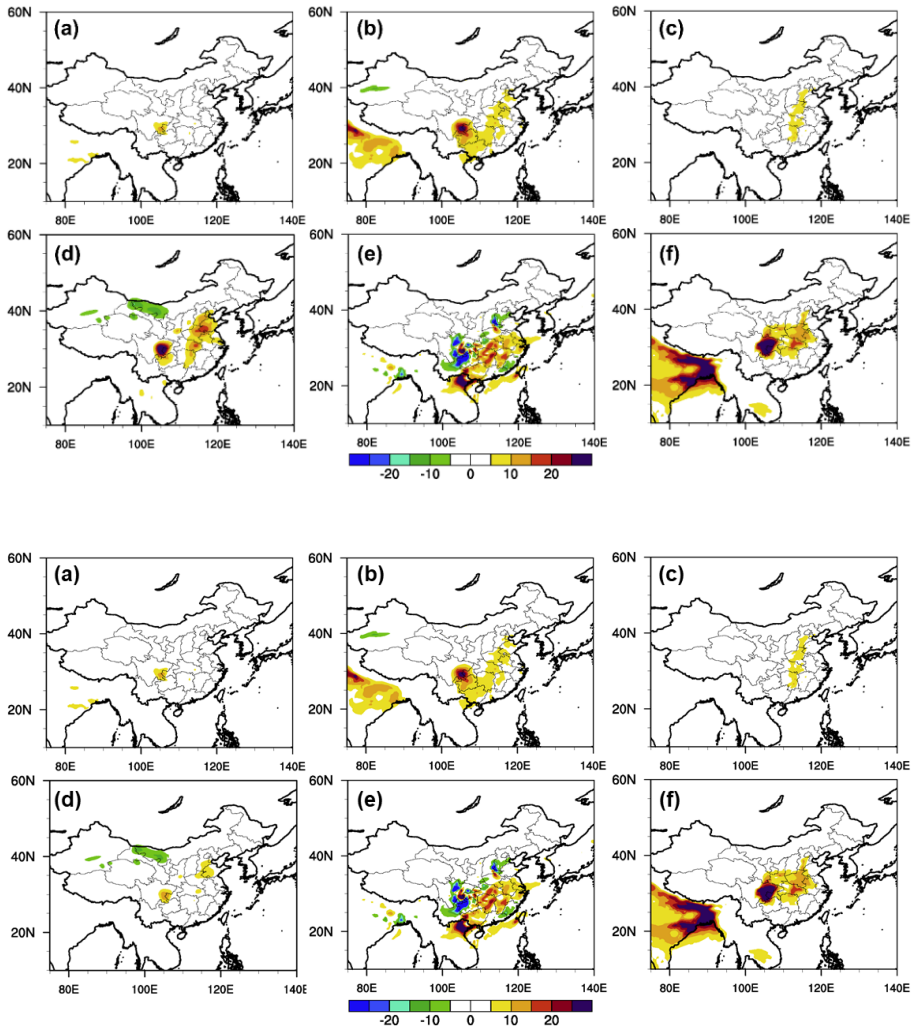

Figure 6. Monthly (January 2010) mean changes in surface $PM_{2.5}$ ($\mu g/m^3$) due to aerosol radiative effects from M1 (a), M2 (b), M4 (c), M5 (d), M6 (e), M7 (f) (M1: Pusan National University; M2: University of Iowa; M4: NASA; M5: Institute of Atmospheric Physics; M6: Nanjing University; M7: University of Tennessee; *Gao et al., 2018a*)

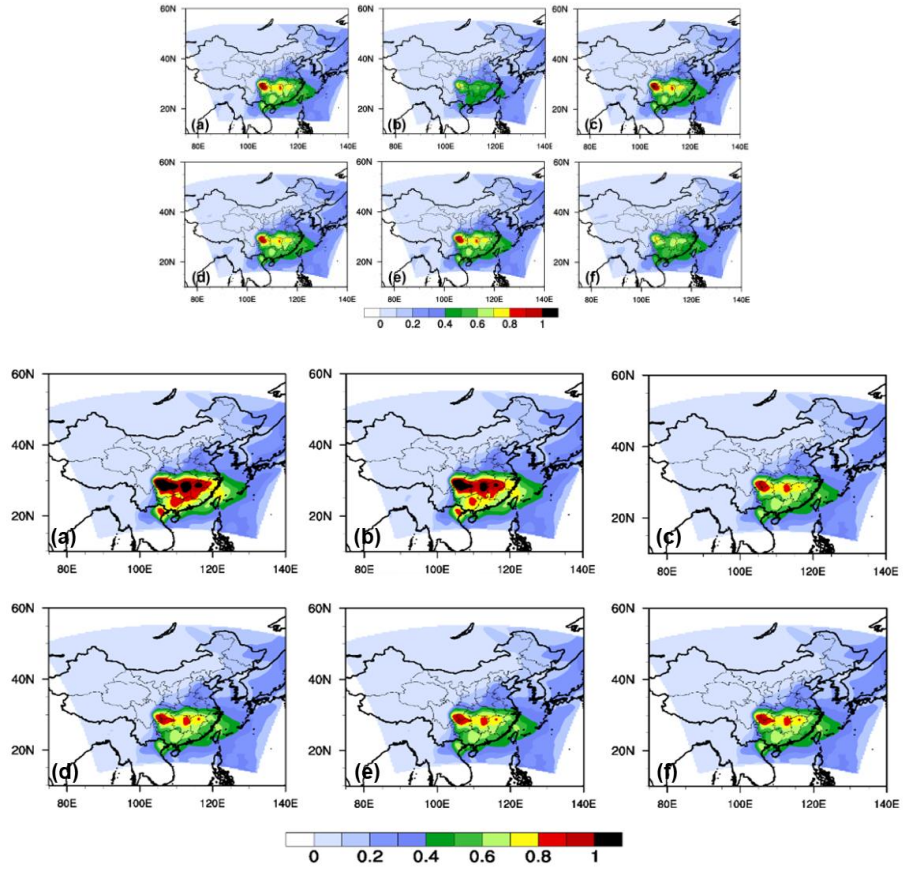

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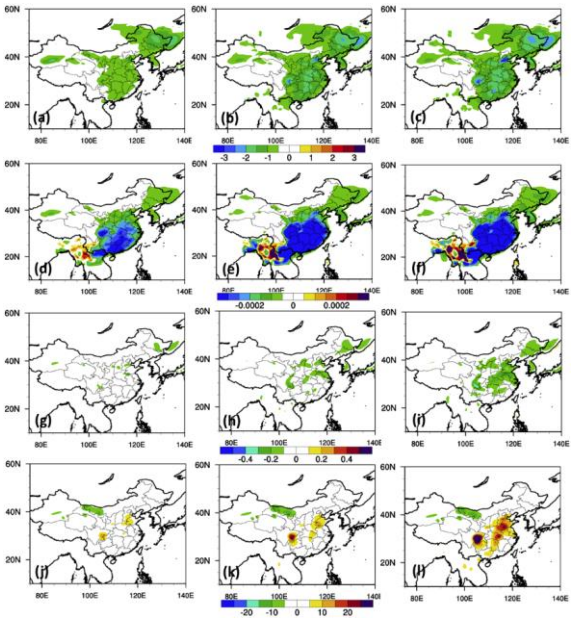
Figure 7. Monthly (January 2010) mean RIEMS-Chem modeled AOD from different simulations: control run (default simulation with internal mixing assumption) (a), external mixing assumption (b), internal mixing assumption but without BC (c), internal mixing assumption but with doubled BC (d), without dust and sea-salt (e), and reduced RH (f)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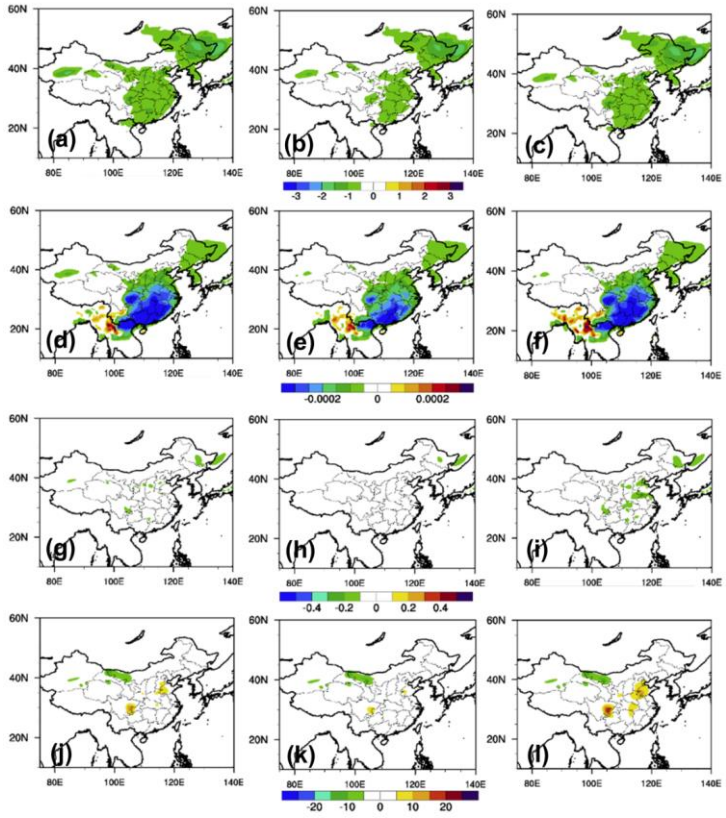

Figure 8. Monthly (January 2010) mean RIEMS-Chem modeled changes in T2 (ºC), Q2 (kg/kg), WS10 (m/s) and $PM_{2.5}$ ($\mu g/m^3$) from different simulations: external mixing assumption (first column), internal mixing assumption but without BC (second column) and internal mixing assumption but with doubled BC (third column)

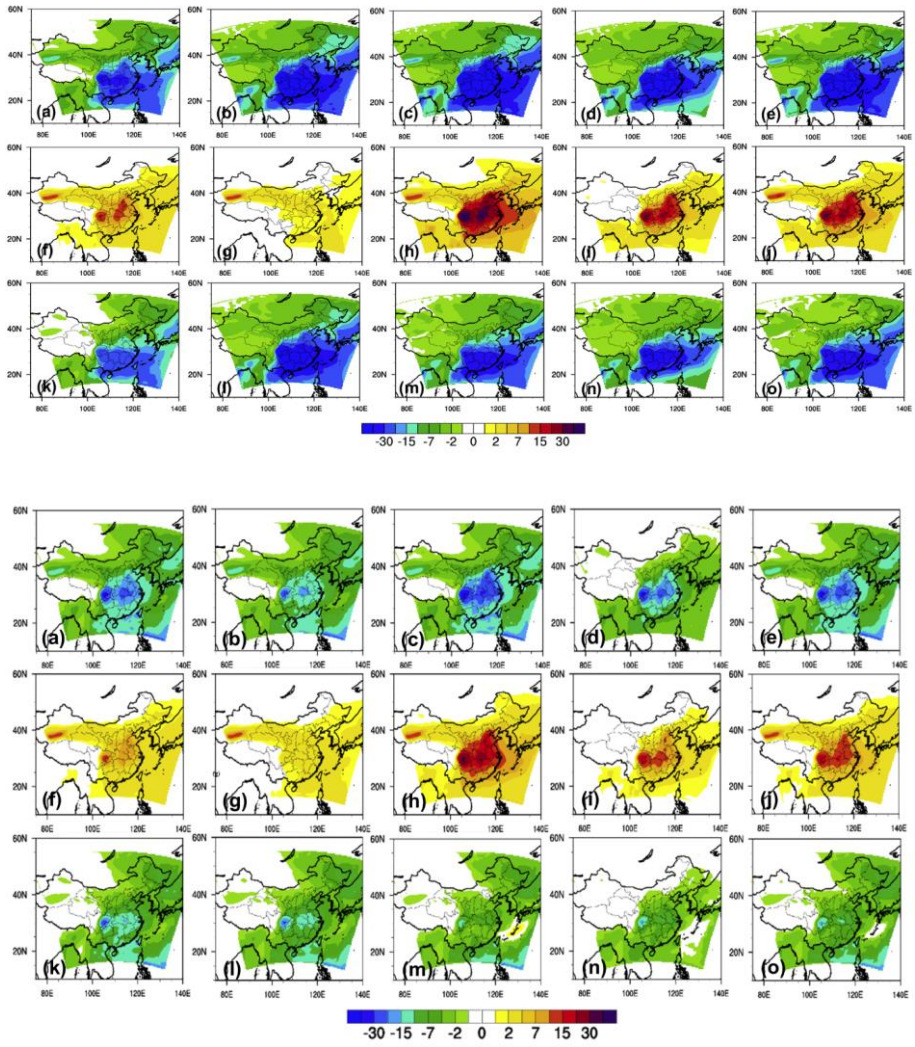

Figure 9. Monthly (January 2010) mean RIEMS-Chem modeled aerosol direct radiative forcing at the surface (a-e), inside the atmosphere (f-j) and at the top of the atmosphere (k-o) from different simulations: external mixing assumption (first column), internal mixing assumption but without BC (second column), internal mixing assumption but with doubled BC (third column), without dust and sea-salt (fourth column), and reduced RH (fifth column)

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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

~~Table1~~ Table 2 Monthly Mean (January 2010) Aerosol Direct Radiative Forcing (W/m^2) and Changes in T2 (ºC), Q2 (g/kg), WS10 (0.1 m/s), and $PM_{2.5}$ ($\mu g/m^3$) for Beijing and Beijing-Tianjin-Hebei region (areas marked in Fig. S1)

Beijing	M1 PNU	M2 UIOWA	M4 NASA	M5 IAP	M6 NJU	M7 UTK
ADRF TOA	-0.6	-2.2	-0.8	-1.45~~.1~~	-0.1	-2.5
ADRF ATM	5.8	4.3	9.3	~~7~~5.1	2.4	11.6
ADRF SFC	-6.4	-6.5	-10.1	~~-12.2~~6.5	-2.5	-14.1
T2	-0.1	-0.3	-0.7	-0.5~~1.0~~	-0.1	0.0
Q2	-1.2E-2	-2.3E-2	-6.4E-2	-5~~1~~.8~~1~~E-~~1~~2	-5.8E-3	2.1E-2
WS10	-0.2	-0.2	-0.6	-0.3~~3~~2	0.0	-1.2
$PM_{2.5}$	0.1 (0.2%)	1.4 (1.6%)	1.1 (1.7%)	~~1.2~~0.6 (1.4~~2.7~~%)	-1.2 (-2.2%)	1.0 (1.4%)
BTH						
ADRF TOA	0.2	-1.4	-0.3	-2.6~~7.6~~	0.0	-2.4
ADRF ATM	7.3	5.4	10.1	6.3~~9.1~~	3.6	14.6
ADRF SFC	-7.1	-6.8	-10.4	-8.9~~16.7~~	-3.6	-17.0
T2	-0.2	-0.4	-0.8	-0.6~~1.3~~	-0.2	0.0
Q2	-1.0E-2	-2.5E-2	-8.1E-2	-7.6~~1.6~~E-~~1~~2	-2.9E-2	2.5E-2
WS10	-0.2	-0.2	-0.9	-0.7~~4~~	0.1	-0.9
$PM_{2.5}$	0.8 (1.4%)	1.8 (1.8%)	2.2 (3.2 %)	~~4.4~~2.2 (3.9~~7.8~~%)	-4.2 (-5.7%)	2.2 (2.4%)

Table ~~2~~ 3 Monthly Mean (January 2010) Aerosol Direct Radiative Forcing and indirect Radiative Forcing (W/m^2) at the top of the atmosphere inferred from M4 and M5 (areas marked in Fig. S1)

Beijing	direct	Indirect
M4	-0.77	-0.15
M5	-1.43~~5.05~~	-0.01
BTH		
M4	-0.28	0.1
M5	~~-7.63~~2.63	-0.04

Table 3 4 Mean Aerosol (January 2010) Direct Radiative Forcing (W/m^2) and Changes in T2 (ºC), Q2 (g/kg), WS10 (0.1 m/s), and $PM_{2.5}$ (μg/m^3) for Beijing and Beijing-Tianjin-Hebei (BTH) region averaged over January 17-19 2010 (areas marked in Fig. S1)

Beijing	M1 PNU	M2 UIOWA	M4 NASA	M5 IAP	M6 NJU	M7 UTK
ADRF TOA	2.6	-1.4	1.8	-3.011.9	-0.6	-3.3
ADRF ATM	18.6	9.8	21.5	13.39.0	7.3	32.3
ADRF SFC	-16.0	-11.2	-19.7	-30.816.3	-7.9	-35.6
T2	-0.5	-0.5	-1.7	-1.33.2	-0.1	-1.5
Q2	-7.4E-2	-6.2E-2	-2.6E-1	-1.84.5E-1	-1.3E-2	-9.2E-2
WS10	-0.1	0.2	-2.3	0.41.7	0.5	-0.8
$PM_{2.5}$	-1.1 (-0.9%)	3.8 (1.7%)	6.3 (3.8%)	-1.02.6 (-02.81%)	-7.9 (-4.7%)	1.3 (1.1%)
BTH						
ADRF TOA	1.4	0.1	4.9	-4.616.0	-0.7	-3.8
ADRF ATM	18.3	12.0	19.1	13.28.7	10.0	36.1
ADRF SFC	-16.9	-11.9	-14.2	-1734.86	-10.7	-39.9
T2	-0.6	-0.7	-1.6	-1.23.0	-0.3	-1.5
Q2	-7.1E-2	-8.2E-2	-2.9E-1	-25.0E-1	-1.2E-1	-8.9E-2
WS10	-0.3	-0.4	-2.5	0.05	0.3	-0.9
$PM_{2.5}$	2.9 (2.3%)	8.5 (3.7%)	5.3 (3.9%)	5.37.9 (35.9%)	-10.5 (-6.2%)	5.1 (2.7%)
Daytime $PM_{2.5}$						
Beijing	2.4 (2.0%)	8.5 (3.9%)	8.4 (5.5%)	-0.72.1 (-0.61.8%)	-4.2 (-3.2%)	10.7 (8.3%)
BTH	6.0 (4.9%)	12.9 (5.9%)	6.6 (5.2%)	58.38 (46.06%)	-6.2 (-3.8%)	6.4 (3.8%)
	Up to 26.4	Up to 55.4	Up to 26.5	Up to 4121.12	Up to 22.8	Up to 60.9

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