Anonymous Referee #3

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General Comments: The manuscript examines the spatial and temporal variability in aerosol concentration and composition simulated by 12 regional models. The model predictions are evaluated with measurements from the different monitoring networks. It also quantifies the ensemble mean bias through several processes, namely model inputs, gas to particle conversion, the impact of sea-salt and dust emissions parameterization, as well as deposition processes. The topic and overall approach fit with ACP. However, I feel that there are some aspects need to be discussed in more detail and the paper needs some improvement before being published in ACP.

As Reviewer #2 already mentioned, the supplementary material contains a very large number of figures that could easily be added in the main text. In the current version, the readers have to go back and forth from the main text to the supplementary material.

It would be nice to have a more in-depth investigation of possible causes that could explain the substantial differences between simulations. As an example, there are large differences between simulations performed using the same model (see Figs.2, S9, S12 and S13). The M4 and M5 models seem to share similar setup and use the same input data, except the organic chemistry treatment. Then, how could the authors explain the different modelled spatial distribution of black carbon, SOR or $C(NO_2)$, as well as the different modelled concentrations of NO_2 and NO_3 ?

The logical connection between the manuscript and the SM needs to be improved. For example, the discussion associated with Fig. S8 appears after the one associated with Fig. S12.

Response: We would like to thank the reviewer for the valuable comments. Following are the responses to the comments.

Specific comments

3Q1: Lines 104-105: it repeats information already provided in the introduction Response: We have revised the manuscript and deleted the redundant information. 3Q2: Line 115: use "IC/BC" instead "BC"

Response: We have changed it.

3Q3: Lines 164-165: Why the online calculation of natural emissions of dust and sea salt particles fully embedded within the WRF-Chem is not included?

Response: We have confirmed with the modelling groups that the two WRF-Chem models did not include dust and sea-salt emissions in the simulations.

3Q4: Line 185: move this information into the main text

Response: We updated the model performance on aerosols in our companion paper (Chen et al., 2019). We removed the section of model evaluation from this manuscript and focused more on the inter-model comparison.

We also moved the figures and tables from the supplementary material to the main text to make it easier for the readers.

Reference: Chen, L., Gao, Y., Zhang, M., Fu, J. S., Zhu, J., Liao, H., Li, J., Huang, K., Ge, B., Wang, X., Lam, Y. F., Lin, C.-Y., Itahashi, S., Nagashima, T., Kajino, M., Yamaji, K., Wang, Z., and Kurokawa, J.: MICS-Asia III: multi-model comparison and evaluation of aerosol over East Asia, Atmos. Chem. Phys., 19, 11911–11937, https://doi.org/10.5194/acp-19-11911-2019, 2019.

3Q5: Lines 185-186: briefly describe the main findings of Chen et al., 2019

Response: We added the description on the main findings of Chen et al., 2019 as follows:

Lines 176-188: "Evaluation of model performance on aerosols can be found in our companion paper (Chen et al., 2019). The following are the main findings: PM_{10} concentrations were generally underestimated over the simulation domain. $PM_{2.5}$ concentrations were also underestimated over Eastern EA but were well simulated in Central EA. Models failed to reproduce the high peaks of SO_4^{2-} concentration in Central EA, probably due to missing SO_4^{2-} formation mechanisms (such as

heterogeneous SO_4^{2-} chemistry), which has been reported as an important formation pathway of SO_4^{2-} in China. NO_3^{-} concentrations were overpredicted by most models over the simulation domain and were associated with the underestimation of SO_4^{2-} . M7 and M8 models produced significantly lower NO_3^{-} concentrations than observations and other models, due to model bias in simulating the NH_3 concentrations and missing the N_2O_5 heterogeneous reaction that severs as an import formation pathway of NO_3^{-} . The spatial distributions of AOD was generally well simulated except underestimation around the Himalaya mountains, Taklamakan Desert and Gobi Desert."

3Q6: Lines 191-193: should be an NMB of -300%?

Response: We removed section 3.2 from the manuscript (reasons are listed in our response to Q9). This sentence has been removed from the manuscript.

3Q7: Lines 196-197: Fig. S3 instead of Fig. S2?

Response: Here we want to refer to the location of HBT in figure S2 (now figure 1 in the revised manuscript).

3Q8: Lines 205-208: For northern and south-eastern EA these findings are not applicable. Could the authors explain why this behaviour is seen only for eastern EA, but not for the other analysed regions?

Response: On lines 205-208 (manuscript before revision), we described the model performance on simulating the trends of SO_4^{2-} and NO_3^{-} concentrations in eastern EA. Model underestimations in several months caused the correlations between model ensemble and observation to be low. This is also the reason for low R values in simulating SO_4^{2-} and NO_3^{-} in northern EA and southern EA.

This sentence has been removed from the manuscript.

3Q9: Lines 258-260: As Reviewer #1 already mentioned, using CO and BC concentration as proxies for emissions won't give a good indication of the emission's spatial distribution.

Response: After checking with the modelling groups, we notice that all models used the same anthropogenic and natural emissions. However, mismatch during the temporal and vertical treatment of emission files by different modelling groups have caused differences in the model inputs (Itahashi et al., 2019). In addition, different model set-up such as the heights of first vertical layer and spatial resolutions of model grids also affect the inter-model comparison on direct model outputs such as surface PM concentrations. Therefore, we decided to use indicators (such as SOR) instead of direct model outputs to facilitate comparison on model mechanisms. For this reason, we removed our discussion on the impacts of emissions and IC/BC on model performance (section 3.2 in the old manuscript) and focused on the comparison of the three processes: gas-aerosol portioning, dust mechanism and wet and dry deposition efficiency.

Reference: Itahashi, S., Ge, B., Sato, K., Fu, J. S., Wang, X., Yamaji, K., Nagashima, T., Li, J., Kajino, M., Liao, H., Zhang, M., Wang, Z., Li, M., Kurokawa, J., Carmichael, G. R., and Wang, Z.: MICS-Asia III: Overview of model inter-comparison and evaluation of acid deposition over Asia, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-624, in review, 2019.

3Q10: Lines 266-269: the models indeed do not provide the layer height in meters, but this can be easily calculated

Response: Thank you for the suggestion. As the reviewer mentioned in Q9, CO is not an appropriate indicator to reveal the vertical distributions of emissions, we removed the discussion and figure related to CO vertical distribution from the manuscript. However, discussion on the inter-model differences on the vertical layers can be found in our companion study (Li et al., 2019).

Reference: Li, J., Nagashima, T., Kong, L., Ge, B., Yamaji, K., Fu, J. S., Wang, X., Fan, Q., Itahashi, S., Lee, H.-J., Kim, C.-H., Lin, C.-Y., Zhang, M., Tao, Z., Kajino, M., Liao, H., Li, M., Woo, J.-H., Kurokawa, J., Wang, Z., Wu, Q., Akimoto, H., Carmichael, G. R., and Wang, Z.: Model evaluation and intercomparison of surface-level ozone and relevant species in East Asia in the context of MICS-Asia Phase III – Part 1: Overview, Atmos. Chem. Phys., 19, 12993– 13015, https://doi.org/10.5194/acp-19-12993-2019, 2019.

3Q11: Lines 273-276: these differences represent averaged values over the domain? It is difficult to connect these numbers with Fig S11. What do you mean by 2-6 ug m⁻³ for NO₃? Do these numbers represent the range of differences? From Fig, S11 the range seems to be between -4 and 6 ug m⁻³.

Response: The numbers represent the ranges of values over the continental regions. For NO_3^- , the range is -4 to 6 μ g m⁻³.

This sentence has been removed from the manuscript.

3Q12: Lines 277-279: have you compared these values with observations?

Response: Lines 277-279 explain the differences in PM components around the edge of the simulation domain between the M1 and M2 is caused by using different boundary condition as model input. M1 used downscale results from GEOS-Chem and thus considered the impacts of emission and pollutions from outside of the research domain. M2 used the default values in the CMAQ model as a boundary condition, which is much smaller than those from GEOS-Chem. This result is not comparable to observations.

This sentence has been removed from the manuscript.

3Q13: Lines 297-300: Why? A different aerosol treatment?

Response: Yes, there is an update in the formation pathway of SO_4^{2-} particles in CMAQv502. We added the following sentences in the manuscript for explanation:

Lines 229-234: "The X-CMAQ models (including WRF-CMAQ and RAMS-CMAQ) produce similar *SOR* patterns, except that the CMAQv5.0.2 models (M1 and M2) predict 10% higher

SOR in the HBT region than the CMAQv4.7.1 models (M4, M5, and M6). CMAQv502 updated the production of SO_4^{2-} in the aqueous reaction of the older version (Appel et al., 2013). The explicit treatment of Fe and Mn allows more consistent treatment of aqueous reaction from SO_2 to SO_4^{2-} ."

3Q14: Figs. 2 and S13 and subsequent discussion: Could the authors provide an explanation for the really low NO₃ concentration modelled with M8? Have they considered the M8 model when the ensemble mean was compared against observation? Using such outliers, the MMM mean can even be deteriorated compared to individual simulations.

Response: The reasons have been discussed on page 11920 of our companion paper (Chen et al., 2019). One possible reason is the low production of NH_3 by M8 (Fig S4 of Chen et al., 2019). Another reason is not including the N_2O_5 heterogeneous reaction in M8, which is an important pathway for the formation of NO_3^- (Chen et al., 2019).

Yes, M8 is included in the ensemble mean. We agree if the MMM is used to show the model performance, the NO_3^- of M8 should be taken out. However, this paper aims at identifying the differences among models. The MMM is used as a reference to evaluate the differences between models. Thus, we kept including M8 in the MMM.

3Q15: Fig. 2 c) M3 should be M4

Response: We have modified the figure.

3Q16: Lines 323-330: if the S emissions were insufficient, why the M13 and M14 models provide reasonable results?

Response: Our current analysis can only demonstrate that the SOR values of models are generally higher than observations. Since model inputs are not identical in different models due to the reason we mentioned in our answer to Q9, we can't confirm if the underestimation of SO₂

is caused by insufficient S emissions. For this reason, we removed the related discussion from the manuscript.

3Q17: Section 3.4: same as before, why not using the online calculation of natural emissions of dust particles within WRF-Chem?

Response: We have confirmed that the two WRF-Chem models do not include dust emissions during the simulation.

3Q18: Line 374: "perfectly" is a strong word

Response: We have changed the word to "reasonably".

3Q19: Lines 432-433: Table 4 and the associated discussion. Please be consistent when calculating the total wet and dry deposition of S/N and doing subsequent analysis.

Response: We have modified the manuscript and checked the calculation to be consistent and clear.

3Q20: Lines 460-473: In the case of wet deposition of S and N, M11 shows a very different spatial pattern compared with the other models. What is the reason for this? It would be nice if the authors will try to analyse in detail the causes of these differences.

Response: We added the explanation in the manuscript as follows:

Lines 403-405: "Both λ_{wet} and V_d of M11 are much lower than the other models, especially over eastern EA. And this is a possible reason for the biased performance of M11 on wet deposition (Fig. 7)"

1 Why models perform differently on particulate matter over East

2 Asia? – A multi-model intercomparison study for MICS-Asia III

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Abstract. This study compares the performances of twelve regional chemical transport models 23 24 (CTM) from the third phase of Model Inter-Comparison Study for Asia (MICS-Asia III) on 25 simulating the particulate matter (PM) over East Asia (EA) in 2010. The participating models include WRF-CMAQ (v4.7.1 and v5.0.2), WRF-Chem (v3.6.1 and v3.7.1), GEOS-Chem, NHM-26 Chem, NAQPMS and NU-WRF. This study investigates three model processes as the possible 27 28 reasons for different model performances on PM: (1) Models perform very differently in the gas-29 particle conversion of sulphur (S) and oxidized nitrogen (N). The model differences in sulphur oxidation ratio (50%) is of the same magnitude as that in SO_4^{2-} concentrations. The gas-particle 30 conversion is one the main reasons for different model performances on fine mode PM. (2) Models 31 32 without dust emissions/modules can perform well on PM₁₀ at non-dust-affected sites, but largely 33 underestimate (upmost 50%) the PM₁₀ concentrations at dust sites. The implementation of dust emissions/modules in models has largely improved the model accuracies at dust sites (reduce 34 35 model bias to -20%). However, both the magnitudes and distributions of dust pollutions are not

fully captured. (3) The amounts of modelled depositions vary among models by 75%, 39%, 21%

and 38% for S wet, S dry, N wet and N dry depositions, respectively. Large inter-model differences
are found in the washout ratios of wet deposition (at most 170% in India) and dry deposition
velocities (general 0.3-2 cm s⁻¹ differences over inland regions). This study investigates the reasons
for different model performances on PM over EA and offers suggestions for future model
development.

42 **1 Introduction**

43 Atmospheric pollution due to particulate matter (PM) has raised world-wide attention for its relationship with environmental and public health issues (Fuzzi et al., 2015;Nel, 2005). Fine 44 particles $(PM_{2.5})$ are associated with cardiovascular and respiratory related cancer and premature 45 46 deaths (Hoek and Raaschou-Nielsen, 2014;Knol et al., 2009). Outdoor PM_{2.5} pollution is estimated 47 to cause 2.1-5.2 million premature deaths worldwide annually (Lelieveld et al., 2015;Rao et al., 2012; Silva et al., 2013). It accounts for eight percent of global mortality in 2015 and ranks fifth in 48 49 the global mortality risk (Cohen et al., 2017). East Asia (EA) has been suffering from severe PM pollutions due to anthropogenic emissions and natural dust emissions (Akimoto, 2003). China and 50 51 India are the top two countries suffering from outdoor air pollution, which altogether account for 52 20% of global mortalities caused by $PM_{2.5}$ exposure in 2010 (Lelieveld et al., 2015). The mixing of dust with anthropogenic pollutants can even enlarge the effects of pollution (Li et al., 2012). 53 However, the impact evaluation of PM pollution is of high uncertainty due to unclearness in the 54 toxicity of PM components (Lippmann, 2014) and difficulty in the measurement and prediction of 55 56 PM concentrations.

57 For a better understanding of PM pollution, modelling approach has been adopted to study 58 the spatial distributions of PM with the aid of measurements. Multi-model ensemble approach, 59 which interprets modelling results with combined information from several models, has been proven to increase the reliability of model accuracy (Tebaldi and Knutti, 2007). This method has 60 61 been widely used for studies in Europe (Bessagnet et al., 2016; Vivanco et al., 2017) and at global 62 scales (Lamarque et al., 2013;Galmarini et al., 2017) on air quality issues. The Model Inter-Comparison Study Asia Phase (MICS-Asia) aims at understanding the air quality issues over EA. 63 The first phase of MICS-Asia (MICS-Asia I) was carried out in the 1990s with eight regional 64 chemical transport models (CTMs). The study focused on air pollution issues related to sulphur 65 (S) (including SO₂, SO₄²⁻ and wet SO₄²⁻ deposition). The second phase of MICS-Asia (MICS-Asia 66

II) was launched in early 2000s with nine CTMs (Carmichael and Ueda, 2008). The study covered 67 the chemistry and transport of S, nitrogen (N), PM and acid deposition. Multi-model results on 68 SO₄²⁻, NO₃⁻ and NH₄⁺ (SNA) were evaluated with measurements from fourteen sites of Acid 69 Deposition Monitoring Network in East Asia (EANET) and the Fukue site in Japan. However, a 70 non-exhaustive evaluation on PM₁₀ concentrations in China with scarce datasets left an unclear 71 72 view of models' ability in this area, a region recognized as one of the most heavily polluted in EA. Meanwhile, model results were found with high inconsistencies on simulating both gas and aerosol 73 phases of S and N (Hayami et al., 2008). Further efforts are needed to investigate the reasons for 74 model differences to improve model accuracies. 75

76 This study compares the performances of twelve regional models participated in the third phase of MICS-Asia (MICS-Asia III) on simulating PM over EA. The comparison among models 77 78 aims at identifying the reasons for different model performances. The models involved in this study include Weather Research and Forecasting Model (WRF) coupled with Community 79 80 Multiscale Air Quality Modeling (CMAQ) (version 4.7.1 and v5.0.2), WRF model coupled with Chemistry (WRF-Chem) (v3.6.1 and v3.7.1), Goddard Earth Observing System coupled with 81 82 Chemistry (GEOS-Chem), Non-Hydrostatic Model coupled with Chemistry (NHM-Chem), Nested Air Quality Prediction Modeling System (NAQPMS) and NASA-Unified WRF (NU-83 84 WRF). The model performance on simulating PM has been reported in a companion paper (Chen et al., 2019). The main findings are described in sect. 3.1. Sections 3.2-3.4 examine the influences 85 of three model processes on model performances: (1) Formation of fine particles (PMF): model 86 differences in the gas-particle conversion. (2) Formation of coarse particles (PMC): model 87 improvements by implementing dust emissions/modules on simulating PM and the remaining 88 problems. (3) Removal processes of particles from the atmosphere: uncertainties lay on the 89 efficiencies of wet and dry depositions. Section 4 concludes the findings of this study and provides 90 91 suggestion for further study.

92 **2 Methodology**

93 2.1 Framework of MICS-Asia

MICS-Asia is a model intercomparison study with contributions from international modelling groups to simulate the air quality and deposition over EA. MICS-Asia I focused on air quality issues related to S. The multi-model performances on simulating SO₂ and SO₄²⁻ concentrations and SO4²⁻ wet deposition were evaluated with observation from eighteen stations (Carmichael et al.,
2002). A source-receptor relationship of S deposition was developed based on the sensitivity
simulations for seven prescribed receptor regions: Komae, Oki, Fukue, Yangyang, Beijing,
Nanjing and Taichung (Carmichael et al., 2002).

101 MICS-Asia II was initiated in 2003. Nine regional models simulated the air qualities for four months (March, July and December of 2001 and March of 2002) to study the chemistry and 102 transport of air pollutants and acid deposition (Carmichael and Ueda, 2008). All modelling groups 103 104 were enforced to use the same emission: the Transport and chemical Evolution over the Pacific (TRACE-P) emission of 2000, and common IC/BC to facilitate a comparison on the physical and 105 106 chemical mechanisms of models. The modelling species expanded to S, N, O₃, PM and acid deposition. Model evaluations and major findings can be found in literature (Carmichael et al., 107 108 2008;Fu et al., 2008;Han et al., 2008;Hayami et al., 2008).

MICS-Asia III is launched in 2010. The simulation time covers the whole year of 2010. All modelling groups are required to use the prescribed anthropogenic and natural inputs (Li et al., 2017). Three purposes are set for this project– topic I: evaluating the strengths and weaknesses of current multi-scale air quality models in simulating air qualities over EA and providing suggestion to reduce uncertainty for future simulations, topic II: developing a reliable anthropogenic emission inventory for EA, topic III: investigating the interaction of aerosol-weather-climate by using online coupled air quality models. This study focuses on topic I.

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117 **2.2 Model configurations**

The model set-up can be found in Table 1 of Chen et al. (2019). Fourteen modeling groups (M1-M14) participated, but M3 and M9 are not included in this study due to uncompleted model submission. M14 model has a smaller simulation domain than the others, therefore it is not included in the multi-model mean (MMM) results. The gas and aerosol modules and dust schemes employed by the participating models were introduced in detail in sector 2.1 of Chen et al., 2019. Following are the descriptions on the model set-up for wet and dry deposition.

Wet deposition removes gases and aerosols from the atmosphere by rain droplets, involvingboth in-cloud scavenging (rainout) and below-cloud scavenging (washout). The gases in the

atmosphere are dissolved in the raindrop and then removed from the atmosphere. For the non-126 reactive gases, the removal rate depends on the solubility of gases and is a function of the Henry's 127 Law. Particles participate in the cloud condensation nuclei in the presence of supersaturation water 128 vapor and then grow into cloud droplets. In this study, only M2, M4, M6, M11 and M12 have 129 submitted the main components of S and N depositions. All these models use the same wet 130 deposition scheme based on Henry's law. The efficiency of wet deposition is assessed by the so-131 called "washout ratio", calculated as the ratio of particle concentrations in deposition to particle 132 133 concentrations in surface air as shown in Eq. 1.

134
$$\lambda_{wet} = \frac{C_{depo}}{C_{surface_air}} \times 100\%$$
(1)

where λ_{wet} is the washout ratio for wet deposition, C_{depo} is the concentration of particles in deposition and $C_{surface_air}$ is the concentration of particles at near surface atmosphere.

Dry deposition is mainly driven by turbulent and molecular diffusion processes. All models except M12 use the same dry deposition scheme from Wesely (1989). The dry deposition flux is proportional to the concentration of pollutants at height. The dry deposition velocity is calculated with Eq. 2.

$$V_d = -F_c / C_a \tag{2}$$

142
$$V_d = \frac{1.0}{r_{surf} + r_a + r_{bc}}$$
 (3)

where F_c is the dry deposition flux, V_d is the deposition velocity and C_a is the concentration of species at height. The negative mark indicates the direction of the dry deposition velocity. The V_d is determined by the resistance of air layer (r). The total r is composed of three factors (Eq. 3): the aerodynamic resistance (r_a), boundary layer resistance (r_{bc}) and canopy resistance (r_{surf}).

147 M12 uses the general approach from Wesely (1989) and updates by Zhang et al. (2003). 148 Zhang et al. (2003) updates the value of non-stomatal resistance (R_{ns}), which is a component of 149 R_{surf} related to the soil uptake and cuticle uptake of dry deposition. Model evaluation shows the 150 updates can improve the model prediction on dry deposition velocities of SO₂ (Zhang et al., 2003).

152 **2.3 Observation data**

To make the discussion clear, we define the regions used in the following analysis here: northern 153 EA (Russia and Mongolia), central EA (China), western EA (Japan and Korea) and southern EA 154 (Cambodia, Lao PDR, Myanmar, Thailand, Vietnam, Indonesia, Malaysia and Philippines). 155 156 Following monitoring datasets are used in the analysis in sects. 3.2-3.4: Air Pollution Indices 157 (APIs) provides monthly average PM_{10} data from eighty-six sites (A1-A86 in Fig 1) (http://datacenter.mep.gov.cn/). This dataset has been widely used to study the PM pollution (Qu 158 et al., 2010; Chen et al., 2008; Deng et al., 2011) as well as model evaluation (Wang et al., 159 2012; Xing et al., 2015) in China. It is replaced by the Air Quality Index (AQI) after 2013. The 160 161 APIs data covers the eastern China well with intensively located sites, but sites in western China are very limited. EANET (E1-E54) provides monthly average concentrations of PM₁₀, SNA and S 162 163 and N depositions from fifty-four sites (http://www.eanet.asia/, last access: 28 May 2018). For PM_{10} , this dataset has very limited number of sites in China. The sites are generally located along 164 165 the east coast of China and couldn't well cover the areas with high PM_{10} pollution, such as the Hebei-Beijing-Tianjin (HBT) region (Fig. 1). And the data completeness in northern EA is not as 166 167 satisfied as the other regions. Only three sites located in Rishiri (E15), Ochiishi (E16) and Oki (E21) in Japan have PM_{2.5} observation during our study period. R1-R35 (green) are thirty-five 168 169 Reference (Ref) sites provided by the Institute of Atmospheric Physics Chinese Academy of 170 Science (IAP_CAS). The sites are intensively located in three regions: HBT region, Pearl River Delta (PRD) and Taiwan. 171

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173 **3 Result and discussion**

174 **3.1 Brief results of model performance evaluation**

175 All models have submitted the monthly average concentrations of PM_{10} , $PM_{2.5}$ and SNA at surface 176 layer except PM_{10} from M13 and NO_3^- and NH_4^+ from M10. Evaluation of model performance on 177 aerosols can be found in our companion paper (Chen et al., 2019). Following are the main findings: 178 PM_{10} concentrations were generally underestimated over the simulation domain. $PM_{2.5}$ 179 concentrations were also underestimated over Eastern EA, but were well simulated in Central EA. 180 Models failed to reproduce the high peaks of SO_4^{2-} concentration in Central EA, probably due to

missing SO_4^{2-} formation mechanisms (such as heterogeneous SO_4^{2-} chemistry), which has been 181 reported as an important formation pathway of SO₄²⁻ in China. NO₃⁻ concentrations were 182 183 overpredicted by most models over the simulation domain and were associated with the underestimation of SO₄²⁻. M7 and M8 models produced significantly lower NO₃⁻ concentrations 184 than observations and other models, due to model bias in simulating the NH₃ concentrations and 185 186 missing the N₂O₅ heterogeneous reaction that sever as an import formation pathway of NO₃⁻. The spatial distributions of AOD were generally well simulated except the underestimation around the 187 Himalaya mountains, Taklamakan Desert and Gobi Desert. 188

This study compares the model performances with global-scale model study. The Task 189 190 Force on Hemispheric Transport of Air Pollution (TF HTAP) is an inter-comparison study of global and regional models to assess the impact of hemispheric transport of air pollutants on 191 192 regional atmosphere. The second phase of HTAP (HTAP-II) involved more than twenty global models to simulate the air quality in 2010 (Galmarini et al., 2017). Most models utilize coarse-193 194 resolution grids at about 2°-3°. The HTAP-II and MICS-Asia III share some common points like using the same emission inventory in East Asia (Li et al., 2017) and using the same observation 195 dataset to evaluate PM₁₀ (more than 100 EANET and API sites) and PM_{2.5} (two EANET sites) 196 (Dong et al., 2018). The mean bias (MB) of PM_{10} over EA is -30.7 µg m⁻³ and -18.6 µg m⁻³ for 197 HTAP-II and this study, respectively (values for sites used by both studies). And the MB of PM_{2.5} 198 is -1.6 µg m⁻³ and -4.3 µg m⁻³ for HTAP-II and this study, respectively. Both studies find 199 underestimation of PM₁₀ concentrations, while PM_{2.5} concentrations are well produced. Models of 200 MICS-Asia III perform slightly better than those of HTAP-II with lower model bias in PM_{10} , 201 202 probably taking the advantage of finer resolutions of model grids.

The so-called "diagnostic evaluation" approach is adopted to check the model bias oriented 203 204 by individual process (Dennis et al., 2010). Although all modelling group are required to use the 205 prescribed emission inventory, but mismatch was found during the temporal and vertical treatments of emission files by different modelling group and has caused differences in the model 206 207 inputs (Itahashi et al., 2019). To avoid the possible impacts on inter-model comparison, we compare the indicators (i.e. sulphur oxidation ratio (SOR)) instead of direct model outputs (i.e. 208 SO₄²⁻ concentrations) to focus on the differences caused by model mechanisms. The following 209 three processes are examined: 210

(1) Formation of PMF: sect. 3.2 investigates the differences in the gas-particle conversion of Sand N among different models.

- (2) Formation of PMC: sect. 3.3 assesses the model abilities in reproducing the spatial and
 temporal distributions of PM in regions affected by dust storm. A comparison is conducted
 between models with and without dust emissions/modules.
- (3) Removal of particles from the atmosphere: sect. 3.4 compares the model performances in
 simulating the amounts of deposition and the efficiencies of wet and dry depositions.
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219 **3.2 Gas-particle conversion**

220 The following two indicators are calculated to illustrate the gas-particle conversions of S and N.

221
$$SOR = \frac{n - SO_4^{2-}}{n - SO_4^{2-} + n - SO_2}$$
(4)

$$C(NO_2) = \frac{n - NO_3^{-1}}{n - NO_3^{-1} + n - NO_2}$$
(5)

where $n-SO_4^{2-}$, $n-SO_2$, $n-NO_3^{-}$ and $n-NO_2$ are the mole concentrations of SO_4^{2-} particle, SO_2 gas, NO₃⁻ particle and NO₂ gas. The $C(NO_2)$ indicator only has NO₃⁻ and NO₂ in the denominator due to the limitation of observation data. But it still can portrait the conversion of N between gas phase and particle phase.

227 Figures 2 and 3 show the distributions of SOR and $C(NO_2)$ values of models. The SOR 228 values are lowest around the HBT region in north-eastern China (10-40%) and highest in southwestern China (60-80%) (Fig. 2). The X-CMAQ models (including WRF-CMAQ and RAMS-229 230 CMAQ) produce similar SOR patterns, except that the CMAQv5.0.2 models (M1 and M2) predict 10% higher SOR in the HBT region than the CMAQv4.7.1 models (M4, M5 and M6). CMAQv502 231 updated the production of SO_4^{2-} in the aqueous reaction of the older version (Appel et al., 2013; 232 Fountoukis and Nenes, 2007). The explicit treatment of Fe and Mn allows more consistent 233 treatment of aqueous reaction from SO₂ to SO₄²⁻. For the X-Chem models (including WRF-Chem, 234 GEOS-Chem and NHM-Chem), the two WRF-Chem models (M7 and M8) produce similar 235 magnitudes and distributions of SOR in all regions, except the south-western China (around Tibet 236 in Fig. 1) and the open oceans, while the NHM-Chem (M12) and GEOS-Chem (M13) models 237 produce slightly higher SOR values over the whole simulation domain. The differences between 238

the X-CMAQ and the X-Chem models are significant over the inland regions of northern and eastern China, Japan and southern EA. The X-CMAQ models generally predict 5-20% higher *SOR* values than the X-Chem models. Similarly, the X-CMAQ models generally give 20% higher $C(NO_2)$ values than the WRF-Chem models, especially in eastern EA (Fig. 3). The $C(NO_2)$ of M8 is extremely low due to unreasonably low NO₃⁻ concentrations.

244 Figure 4 shows the gas-particle conversions of S and N by models and observation at the EANET sites. The red bars represent concentrations of gases and the black bars represent 245 concentrations of aerosols. The values with blue color above the bars are observed and modelled 246 SOR and $C(NO_2)$ values. Results for individual sites are available in supplementary Fig. S1. 247 According to Fig. 4(a), the total amount of S (SO₂ gas+SO₄²⁻ particle) is about 0.15 μ mole(S) m⁻³. 248 Most models have biases on this value, especially the moderate underestimation by M7, M8 and 249 250 M13. On the other hand, the SOR value (0.25) is well simulated by M1 (0.26), M2 (0.20), M10 (0.29) and M13 (0.26). Other models generally under-predict the SOR value except M12 (0.33) 251 252 and M14 (0.57). The WRF-CMAQv5.0.2 models (M1 and M2) produce higher SOR than WRF-253 CMAQv4.7.1 models (M4, M5 and M6), probably attributed to the updates in the formation pathway of SO_4^{2-} . 254

Figure 4(b-e) show the results in different regions. In northern EA, the total amount of S is 255 underestimated by all models except M13 and M14. However, the SOR value (0.12) is well 256 257 reproduced by most models (0.08-0.20) except M12 (0.25) and M10 (0.32). There is only one site 258 available for central EA. Most models (except M12 and M13) have largely underestimated the 259 SOR value, while M14 has largely overestimated it. For eastern EA, the total amount of S is well captured by all models except M11, M12 and M14. The SOR value (0.55) is generally 260 underestimated by all models except M10 (0.55) and M14 (0.71). For southern EA, the total 261 262 amount of S is generally overestimated by all models except M13, while the SOR value is 263 underestimated by all models except M13 and M14. Overall, the models have both positive and negative biases in simulating the total amounts of S, but generally underestimated the SOR values 264 265 in all regions. Furthermore, the modelled SOR values vary largely among models (ranging from 0.12 to 0.57), resulting in a large inter-model difference (1sd% = 50%). This variation is of the 266 same magnitude as the variation of SO_4^{2-} concentration (1sd% = 50% in supplementary Fig. S2). 267

The results suggest that differences in gas-particle conversion among models could account largely for the models' inconsistency in simulating the SO_4^{2-} concentrations.

Figure 4(f-h) compares the gas-particle conversion of N with the $C(NO_2)$ indicator. Only 270 one site in China and one site in Japan have both NO₂ and NO₃⁻ observations. At the Hongwen 271 272 sites in China, all models except M5 underestimate the sum of NO₂ and NO₃, but the modelled 273 $C(NO_2)$ values are close to the observation (0.18) except M5 (0.07), M8 (0.00) and M12 (0.40). Similar to the results of S conversion, the newer version of WRF-CMAQ model generally produces 274 higher $C(NO_2)$ than the older version, but the differences between the two are smaller. At the 275 Banryu site in Japan, the sum of NO_2 and NO_3^- is well simulated by all models except M8. The 276 277 $C(NO_2)$ (0.19) value is also well simulated by all models except M8 (0.00), M12 (0.53) and M14 (0.77). Overall, the model accuracy on $C(NO_2)$ is slightly higher than that on SOR according to the 278 279 comparison with observed values. Models also have higher consistencies on $C(NO_2)$ than SOR. However, further validation is required due to the limited number of observations for the 280 281 conversion of N.

282

3.3 Implementation of dust emissions/modules in models

284 The PMC concentrations at surface layer are calculated by subtracting PM_{2.5} from PM₁₀. Figure 5 shows the spatial distribution of annual average PMC of models. Most models show very low (< 285 2µg m⁻³) concentrations of PMC around the Takalmakan Desert and the Gobi Desert in northern 286 China except M10, M11 and M14. These three models use dust emissions/modules in simulations 287 (Chen et al., 2019). M12 also includes dust emissions, but its PM_{10} concentrations over northern 288 China are much lower than the three models. The predicted PMC concentrations of the three 289 models differ largely. The domain-average concentrations of PMC are 21, 7 and 12 μ g m⁻³ for 290 M10, M11 and M14, respectively. The distributions of PMC also differ largely over north-west 291 292 China, where the impacts of dust are most significant. The differences among the models mainly 293 comes from the different parameterizations such as source functions, dust-lifting mechanisms and 294 size distributions of particles (Chen et al., 2019). Different PMC concentrations are also found 295 over oceans, mainly attributed to the sea-salt emissions in this study. The sea-salt emissions are parameterized in the models with various formula (Chen et al., 2019). In this study, the WRF-296 297 Chem models (M7 and M8) do not account for sea-salt emissions, thus their PMC concentrations

over the oceans and seas are not defined. The two WRF-CMAQ models use the in-line sea-salt emission module of Gong (2003) and updated by Kelly et al. (2010). They predict consistent distributions of PMC over oceans. M10 and M11 use the same module as the CMAQ models (Gong, 2003), but produce higher PMC on oceans. M12 adopts the method of breaking wave over seashore by Clarke et al. (2006) and produces the highest PMC over oceans among all models.

303 The implementation of dust emission is expected to improve the model performances, but how significant could the improvement be? And can models predict the PM concentrations 304 reasonably at regions affected by dust with current dust emissions/modules? To answer these 305 questions, all sites are grouped to dust and non-dust sites according to their locations. The sites 306 307 located in regions that have been reported to receive severe impacts and rapid deposition of dust are marked as dust sites (Shao and Dong, 2006) (grey-color shaded areas in Fig. 1). Figure 6(a-b) 308 309 and Table 1 compare the model performances at the dust and non-dust sites. For the non-dust sites (Fig. 6(b)), most models have well captured the magnitudes of PM₁₀ at the "API non-coastal, non-310 311 dust" sites (MB = -8% and NMB = -8%). The sites marked as "API coastal" sites, which are located close to the coastal regions, are all slightly underestimated by about 25 μ g m⁻³ (30%). Similarly, 312 the PRD and Taiwan sites, which are also located near the coastal regions, are all underestimated 313 by about 20 μ g m⁻³ (37%). Bias in sea-salt emissions is the possible reason. Sea-salt emission is 314 315 reported to contribute to 20-40% of SNA and PM₁₀ over coastal regions (Liu et al., 2015). 316 Including the sea-salt emission in model simulation can improve the model accuracy with 8-20% increase in PM₁₀, SNA, Na⁺ and Cl⁻ (Kelly et al., 2010;Im, 2013). The influence of sea-salt 317 emission is not the focus of this study, but further study is strongly recommended. 318

For the dust sites (Fig. 6(a)), most models have generally underestimated the PM₁₀ 319 concentrations by 10-40 μ g m⁻³ (15-50%). And the three models with dust module perform better 320 321 than the others at the dust sites, especially site A2, A30, A68, A69, R5 and R18. However, they miss the high PM₁₀ concentrations at sites like R1-R3 and R11, and overestimate the PM₁₀ 322 concentrations at sites such as A60 and A80. This indicates that the dust emissions/modules 323 324 involved in this study can't fully capture the magnitudes and distributions of dust pollutions over EA. In addition, the modelled PMC differ a lot with different dust emissions/modules (Fig. 5). 325 326 M10 model produces very high PMC over the whole eastern China, while M11 model only predicts high PMC around the HBT region. Overall, the model performance on PM over dust regions can 327

be improved largely by including dust emissions/modules. However, the concentrations and
 distributions are not yet well captured and large inconsistencies are found among different dust
 emissions/modules.

Figure 6(c-d) compares the modelled monthly trends of PM₁₀ with observations at the dust 331 332 and non-dust sites and Fig. 6(e) shows the correlations (R) values between models and observation. 333 For the non-dust sites (Fig. 6(d)), the trends are well caught by most models. The R values are close to 0.70 for all models except M7 (0.62), M8 (0.58) and M14 (0.63). The WRF-Chem models 334 (M7 and M8) simulate too low PM₁₀ concentrations in winter. M14 model overestimates the PM₁₀ 335 concentrations during March to May. Most models have much lower R values at the dust sites than 336 337 the non-dust sites (Fig. 6(e)), due to underestimation of the PM₁₀ concentrations during winter. For instance, R values of M10 drop from 0.7 at the non-dust sites to 0.11 at the dust sites. Spring 338 339 (March, April and May) has the largest model biases at the dust sites, which is coincident with the dust storm season in Asia (Arimoto et al., 2006). M10 and M14 models perform well in most 340 341 months at both the dust and non-dust sites, taking the advantage of their dust emissions/modules. But their R values at the dust sites are very low. Future study is strongly suggested on a better 342 343 understanding of the seasonal variations of dust pollutions.

344 **3.4 Wet and dry depositions**

Figure 7 and Table 2 show the model performance on wet deposition. For wet SO_4^{2-} deposition, 345 346 despite that the two sites with highest deposition (E2 and E3) in China are underestimated, the other sites are generally well simulated by MMM with a low MB of -8%. The individual model 347 bias varies from -22% to 41%. The CMAQ models (M2, M4 and M6) all underestimate the wet 348 SO₄²⁻ deposition. There are large differences between CMAQv4.7.1 and CMAQv5.0.2 in JP, 349 where the CMAQv4.7.1 models (M4 and M6) slightly overestimate the wet SO_4^{2-} deposition at 350 E19 and E23, while the CMAQv5.0.2 model (M2) slightly underestimates the value at these sites. 351 The M11 model produces considerably higher wet deposition of SO_4^{2-} and NO_3^{-} than the other 352 353 models in East EA. The possible reasons are discussed later. The MMM underestimates the NO3⁻ wet deposition by 29%, due to large under-prediction in southern EA. The southern EA has several 354 sites with very high deposition, such as E29 site in MY and E35 and E36 sites in PH, but all models 355 356 fail to catch those high peaks. The individual model bias varies from -59% to 30% among models. M2, M4, M6 and M12 perform similarly with high underestimation ranging from 39% to 59%. 357

The M11 is the only model that succeed to capture the high wet NO_3^- deposition at E2 and E3 in 358 CH, but it overestimates most sites in CH, JP and KR. In case of wet NH₄⁺ deposition, the MMM 359 360 generally underestimates the amount at all sites with a bias of -40%, especially at E2-E4 in CH, 361 E45 in TH and E35 and E36 in PH. The individual model bias varies from -10% to -37%. The M2, M4 and M6 models perform similarly, while M11 and M12 models predict higher depositions at 362 all sites. Overall, large inter-model disagreements are found in eastern EA for wet deposition of 363 SO_4^{2-} and NO_3^{-} and in southern EA for the wet NH_4^+ deposition. The observation of dry deposition 364 is composed by observed concentration of air pollutants and simulated deposition velocity. Since 365 the EANET network only provides the former one, complete evaluation of the dry deposition is 366 367 not available in this study (complete dry deposition with velocity is available after 2013).

Table 3 lists the domain-total annual-accumulated amounts of S and N depositions by 368 models. The total wet S deposition (D_{Swet}) includes wet depositions of SO₂, H₂SO₄ and SO₄²⁻. The 369 total dry S deposition (D_{Sdry}) includes dry deposition of SO₂, H₂SO₄ and SO₄²⁻. The total wet N 370 deposition (D_{Nwet}) includes wet depositions of NO₃⁻, NH₄⁺, HNO₃, NH₃. The total dry N 371 depositions (D_{Nwet}) includes dry deposition of NO, NO₂, NO₃⁻, NH₄⁺, HNO₃ and NH₃. D_{Swet} values 372 range from 10.5 to 31.3 Tg(S) yr⁻¹ among models (1sd%=75%). The estimation by M11 model is 373 two folds higher than the other four models. The inter-model difference is significant even among 374 375 the same type of models with different versions. The CMAQv4.7.1 models (M4 and M6) produce 12.5 Tg(S) yr⁻¹ (M4) and 13.8 Tg(S) yr⁻¹ (M6) of D_{Swet}, while the prediction by CMAQv5.0.2 376 model (M2) is 25% lower. Despite the large discrepancies in the total amount, all five models 377 agree that over 95% of D_{Swet} is wet SO_4^{2-} deposition. The total amounts of D_{Sdrv} range from 4.3 to 378 10.6 Tg(S) yr⁻¹ among models (1sd% =39%). M11 predicts higher D_{Sdry} than other models and the 379 CMAQv5.0.2 model (M2) predicts 45% lower D_{Sdry} than the two CMAQv4.7.1 models (M4 and 380 M6). Similar to D_{Swet}, all models have high agreements on the proportions of the components. 381 D_{Nwet} range from 12.2 to 20.0 Tg(N) yr⁻¹ among models (1sd%=21%). The CMAQ models (M2, 382 M4 and M6) simulate close results (12-15 Tg(N) yr⁻¹), while M11 (20.0 Tg(N) yr⁻¹) and M12 (16.5 383 Tg(N) yr⁻¹) simulate slightly higher amounts. As for the proportion of components, M2, M4, M6 384 and M12 models predict high proportions of wet NO_3^- and wet NH_4^+ depositions (particle phase), 385 while M11 model produces higher percentages of wet HNO3 and wet NH3 depositions (gas phase). 386 D_{Ndry} range from 3.9 to 14.1 Tg(N) yr⁻¹ (1sd%=38%). M12 gives a considerably lower amount 387 than the other models. Models are quite consistent on the proportions of components. 388

Figure 8(a-e) show λ_{wet} of S deposition (λ_{Swet}) by models. The CMAQ models (M2, M4 and 389 M6) have similar patterns in λs_{wet} over the inland regions, while M12 model predicts 30-90% lower 390 391 ratios in India. M11 model generally predicts about 20-70% lower λs_{wet} than the other four models except India, where the difference could reach upmost 170%. For λ_{wet} of N deposition (λ_{Nwet}) (Fig. 392 8(f-j)), the CMAQv4.7.1 models (M4 and M6) and M12 perform similarly, but the CMAQv5.0.2 393 394 model (M2) predicts 30% lower λ_{Nwet} in India, Japan and Korea. M11 generally predicts lower ratios in India (60% lower), Indonesia and Philippines (120% lower) than the CMAQ models. 395 Figure 9 shows the spatial distributions of V_d . For V_d of S deposition (V_{Sd}) (Fig. 9(a-e)), the CMAQ 396 models (M2, M4 and M6) simulate very similar spatial distributions. M11 and M12 models predict 397 0.5 cm s^{-1} lower V_{Sd} than the CMAQ models over the whole inland regions, especially in east China 398 and India peninsular. For V_d of N deposition (V_{Nd}) (Fig. 9(f-j)), the CMAQ models (M2, M4 and 399 M6) predict very similar distributions. M11 and M12 predict about 0.3 cm s⁻¹ and 1-2 cm s⁻¹ lower 400 V_{Nd} than the CMAQ models over the inland regions. Both λ_{wet} and V_d of M11 are much lower than 401 the other models, especially over eastern EA. And this is a possible reason for the biased 402 performance of M11 on wet deposition (Fig. 7). Overall, large inter-model differences are found 403 404 in predicting both the amounts of depositions and the efficiencies of depositions.

405

406 4 Conclusion

The topic I of the MICS-Asia III aims at (i) evaluating the strengths and weaknesses of current multiscale air quality models in simulating concentration and deposition fields over East Asia and (ii) providing suggestions for future model developments. This study compares the performances of twelve regional models for the prediction of PM concentrations over EA. The participating models includes WRF-CMAQ (v4.7.1 and v5.0.2), WRF-Chem (v3.6.1 and v3.7.1), GEOS-Chem, NHM-Chem, NAQPMS and NU-WRF. Three processes/mechanisms are investigated to identify the causes of inter-model differences:

(1) For the formations of PMF, *SOR* and *C(NO₂)* values are used to demonstrate the inter-model
differences in gas-particle conversions. The *SOR* values are generally underestimated by most
models at the EANET sites. A generally trend is found that the WRF-CMAQv5.0.2 models
produce the highest *SOR* values among all models, followed by the WRF-CMAQv4.7.1 models
(10% lower in HBT region), the WRF-Chem models and other models (5-20% lower over

419 inland regions). The inter-model variation on *SOR* (1sd% =50%) is of the same magnitude as 420 that on SO_4^{2-} concentration. Similar results are found in *C*(*NO*₂), but models have higher 421 agreements on *C*(*NO*₂) than *SOR*. The different treatments of gas-particle conversions account 422 largely for the different model performances on PMF.

423 (2) For the formations of PMC, the models without dust emissions/modules generate very low 424 ($<2\mu g m^{-3}$) PMC concentrations. They can well capture the PM₁₀ concentrations at non-dust-425 affected sites but underestimate the PM₁₀ concentrations at sites affected by dust storms by 426 upmost 50%. This underestimation is largely improved by implementing dust 427 emissions/modules (bias reduced to around -20%). However, both the magnitudes and 428 distributions of dust pollutions are not fully captured. In addition, models employing different 429 dust emissions/modules show large disagreements on the distributions of PMC.

(3) For the removal of PM from the atmosphere, the amounts of atmospheric deposition vary largely among models (1sd%) by 75%, 39%, 21% and 38% for D_{Swet} , D_{Sdry} , D_{Nwet} and D_{Ndry} , respectively. The λ_{wet} and V_d indicators are used to exclude the influences brought by model inputs. For λ_{wet} , models agree more on the D_{Swet} than D_{Nwet} . The largest model inconsistencies are found in India (upmost 170%), Indonesia and Philippines (upmost 120%). For V_d , models differ more on D_{Ndry} than D_{Sdry} , which is opposite to λ_{wet} . The inter-model differences are widely found over the inland regions for D_{Sdry} (about 0.5 cm s⁻¹) and D_{Ndry} (0.3-2 cm s⁻¹).

The main contributions of this study are: (1) comparing the conversions of S and N between gas 437 and particle phases among different models as well as with observations. The comparison with 438 observation makes it possible to both quantify the inter-model differences and tell which module 439 440 might be more reasonable; (2) Several new updates on dust modules have been published in recent literature, but there is limited study on the inter-comparison. This study provides an opportunity 441 to bring together the new updates on dust modules/emission and review their performance in EA; 442 (3) providing a comprehensive view on the total budget of S and N aerosols, by including the 443 analysis on the removal processes. It turns out that this process brings significant uncertainties to 444 445 inter-model differences. It should be noted that other factors such as vertical diffusion can also contribute to model differences. Meanwhile, this study focuses on comparing the model abilities 446 in simulating PM in 2010. The chemical regimes may have changed drastically due the rapid 447 changes of emissions and implementation of control policies in Asia. Studies on more recent years 448 449 and heavily polluted episodes are under preparation.

450

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455	

- 456 Data Availability. The observation data are introduced with details in supplementary sect. S2.1
- 457 with web links of public available datasets. The model data are available upon request.
- 458
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- 465

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- **Figures and tables** 635
- 636

Figure 1 637



638 639 Figure 1 The geographical locations of observation networks of API (red color, A1-A86), EANET (blue color, E1-640 E54, only sites with available observation during simulation time are shown) and Ref (green color, R1-R35) sites.

641 Grey shaded regions have been reported to be affected by dust storms.



643 90°E 105°E 120°E 135°E 90°E 90°E 90°E 105°E 105

648 Figure 3



649 650 Figure 3 Same as Fig.2 but for $C(NO_2)$ (unit: %). $C(NO_2)$ is calculated by NO₃^{-/}(NO₂+NO₃⁻)×100%. The $C(NO_2)$ of

651 M8 is extremely low due to unreasonable low NO₃⁻ concentration, which is considered as outliner in this study. Values

are calculated by annual average data.





Figure 4 Gas-particle conversions of S and N of observation and models at EANET sites. The unit is μ mole (S or N) m⁻³. The red bars and black bars represent the concentrations of gases and aerosols. The blue-color values above the bars are observed/modelled *SOR* and *C*(*NO*₂). Values are calculated with annual average concentrations. The concentrations of gases and aerosols are all transferred to μ mole (S or N) m⁻³ before calculation. The blue-color numbers on top-right (e.g. E22) are site numbers. The locations of the sites are illustrated in Fig. 1. Results for individual sites are shown in supplementary Fig. S1.



662 90°E 105°E 120°E 135°E 90°E 105°E 120°E 135°E
663 Figure 5 Annual average PMC concentrations at surface layer of individual models (μg m⁻³). The value is calculated by subtracting PM_{2.5} from PM₁₀. The values in left-bottom are domain average (Avg) and maximum (Max) values.



668Figure 6 Multi-model performances on (a-b) annual average PM_{10} concentrations at the dust sites and non-dust sites669and (c-d) monthly average PM_{10} concentrations at the dust sites and non-dust sites. X axis for (a-b) indicates site670numbers. The locations of the sites are illustrated in figure 1. The yellow bars are observations, the blue lines are the671MMM and different markers represent individual model results. (e) R values of models with observations at the dust672and non-dust sites.

Figure 7



Figure 7 Modelled annual-accumulated wet deposition of SO₄²⁻, NO₃⁻ and NH₄⁺ compared with observation from
 EANET network. The units are mg(S or N) m⁻² yr⁻¹. Abbreviation for regions: RU-Russia, MN-Mongolia, CH China, JP-Japan, KR-Korea, KH-Cambodia, MN-Myanmar, TH-Thailand, VN-Vietnam, ID-Indonesia, MY-





68390°E105°E120°E135°E90°E105°E120°E135°E0684Figure 8 Washout ratios (λ_{wet}) of (a-e) S deposition and (f-j) N deposition of models. Values are calculated with
annual accumulated depositions. The unit is %.0



Figure 9 Dry deposition velocities (V_d) of (a-e) S deposition and (f-j) N deposition of models. Values are calculated with annual accumulated depositions. The unit is cm s⁻¹.

690 with

Table 1

693Table 1 Multi-model performance on annual average concentrations of PM_{10} at the dust and non-694dust sites (unit: $\mu g m^{-3}$)

				aust	sites (unit: µg	(m °)					
Dust site	M1	M2	M4	M5	M6	M7	M8	M10	M11	M12	M14	MMM
Mean Obs						12	20.7					
Mean MMM	77.2	82.2	81.6	51.7	65.6	47.5	44.3	102.5	73.5	77.3	92.1	69.2
S	0.4	0.4	0.4	0.3	0.3	0.2	0.2	0.1	0.2	0.2	0.3	0.3
MB	-43.5	-38.5	-39.2	-69.0	-55.1	-73.2	-76.4	-18.2	-47.2	-43.4	-28.6	-51.5
R	0.4	0.4	0.4	0.4	0.4	0.2	0.2	0.1	0.2	0.2	0.2	0.3
F	66.7	69.2	69.2	38.5	56.4	35.9	33.3	84.6	59.0	66.7	66.7	66.7
<i>NMB</i> (%)	-36.1	-31.9	-32.4	-57.2	-45.7	-60.6	-63.3	-15.1	-39.1	-36.0	-23.7	-42.6
<i>NME</i> (%)	38.3	35.4	36.4	57.2	46.2	60.6	63.3	32.8	42.3	40.5	36.1	42.7
<i>MFB</i> (%)	-49.4	-44.6	-44.6	-83.4	-64.1	-92.9	-98.8	-19.3	-51.8	-46.8	-31.7	-56.9
MFE (%)	51.8	48.3	48.7	83.4	64.7	92.9	98.8	36.1	55.3	51.7	44.5	56.9
Number of Sites							39					

				Tał	ole 1 C	ontinu	ed					
Non-dust site	M1	M2	M4	M5	M6	M7	M8	M10	M11	M12	M14	MMM
Mean Obs						77	7.2					
Mean MMM	58.2	58.5	66.5	45.2	55.2	44.8	39.0	90.0	64.4	66.3	89.5	57.8
S	1.0	1.1	1.2	0.8	1.0	0.7	0.6	1.0	1.0	0.9	1.1	0.9
MB	-19.0	-18.7	-10.8	-32.1	-22.1	-32.5	-38.3	12.7	-12.9	-10.9	12.2	-19.4
R	0.7	0.8	0.7	0.8	0.8	0.6	0.6	0.7	0.7	0.7	0.6	0.8
F	82.5	81.0	84.1	66.7	82.5	52.4	46.0	85.7	90.5	93.7	84.1	82.5
<i>NMB</i> (%)	-24.6	-24.2	-14.0	-41.5	-28.6	-42.0	-49.5	16.5	-16.6	-14.1	15.8	-25.1
<i>NME</i> (%)	30.7	30.7	27.3	41.5	31.4	43.9	50.7	25.7	26.3	26.1	30.8	28.0
MFB (%)	-36.8	-37.5	-25.1	-59.2	-41.8	-62.0	-75.0	13.1	-24.9	-20.3	8.3	-34.6
<i>MFE</i> (%)	42.0	42.8	35.3	59.2	44.4	64.0	76.1	23.4	33.5	31.3	29.1	37.5
Number of Sites						6	i3					

Table 2

Table 2 Multi-model performances on wet deposition (unit: mg(S or N) m⁻² y⁻¹)

_		V	Vet SO ₄ ²⁻	Deposition	n	Wet NO ₃ ⁻ Deposition						
	M2	M4	M6	M11	M12	MMM	M2	M4	M6	M11	M12	MMM
Mean Obs	931.3	931.3	931.3	931.3	931.3	931.3	460.9	460.9	460.9	460.9	460.9	460.9
Mean MMM	633.7	724.2	775	1313.2	826.2	854.5	187.5	266.7	279.5	597.8	308.3	328
S	0.3	0.3	0.3	0.3	0.2	0.3	0.1	0.1	0.1	0.2	0.1	0.1
MB	-297.7	-207.1	-156.3	381.9	-105.1	-76.9	-273.4	-194.2	-181.4	137	-152.6	-132.9
R	0.5	0.4	0.4	0.2	0.3	0.4	0.1	0.2	0.2	0.2	0.1	0.2
F	61.2	61.2	61.2	24.5	40.8	51	38.8	49	46.9	44.9	38.8	46.9
NMB	-32	-22.2	-16.8	41	-11.3	-8.3	-59.3	-42.1	-39.4	29.7	-33.1	-28.8
NME	49.3	50.2	51.5	117.3	62.8	53.6	66.2	60.9	60.6	78.4	68.8	58.2
MFB	-37.4	-23.4	-15.8	4.6	-11.4	-4.6	-75.8	-49.8	-42.1	25.8	-40.9	-27.6
MFE	57.8	55.9	53.7	93.8	66.7	57.6	84.9	71.2	69.3	61	74.6	62.3
Number of Sites	49	49	49	49	49	49	49	49	49	49	49	49

$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$									
	Wet NH ₄ ⁺ Deposition								
	M2	M4	M6	M11	M12	MMM			
Mean Obs	558.4	558.4	558.4	558.4	558.4	558.4			
Mean MMM	459.9	349.4	497.4	505	478	337.6			
S	0.3	0.1	0.3	0.3	0.2	0.2			
MB	-98.5	-208.9	-61	-53.4	-80.4	-220.7			
R	0.3	0.2	0.3	0.4	0.4	0.3			
F	40.8	44.9	44.9	51	46.9	38.8			
NMB	-17.6	-37.4	-10.9	-9.6	-14.4	-39.5			
NME	64.8	65.5	64.9	58.2	57	63.6			
MFB	-21.2	-42.4	-14.4	-18	-12.6	-41.9			
MFE	70.7	77.9	69.1	65.9	62.9	76.1			
Number of sites	49	49	49	49	49	49			

Table 3

Table 3 Domain-total annual-accumulated S and N depositions of models (Tg(S or N) yr⁻¹). Empty values mean no model submissions or the values are 0.

	I J										
Modal	Wet S deposition				Dry S deposition						
Model	SO_2	H_2SO_4	SO4 ²⁻	Total Wet S	SO ₂	H_2SO_4	SO4 ²⁻	Total Dry S			
M1	0.06	-	-	-	-	-	-	-			
M2	0.04	-	10.4	10.5	3.4	0.01	0.9	4.3			
M4	0.06	-	12.5	12.5	6.6	0.01	1.1	7.6			
M5	-	-	-	-	-	-	-	-			
M6	0.05	-	13.7	13.8	6.3	0.01	1.4	7.7			
M7	-	-	-	-	-	-	-	-			
M8	-	-	-	-	-	-	-	-			
M10	-	-	-	-	-	-	-	-			
M11	1.1	0.3	29.9	31.3	6.9	2.2	1.5	10.6			
M12	-	-	16.3	16.3	3.7	-	0.4	4.2			
M13	6.0	-	-	-	-	-	-	-			
M14	0.02	-	6.2	_	5.4	-	3.2	-			

Table 3 Continued

	Wet N deposition						Dry N deposition							
Model	NO ₃ -	$\mathrm{NH_4^+}$	HNO ₃	NH ₃	Total Wet N	NO	NO_2	NO ₃ -	$\mathrm{NH_{4}^{+}}$	HNO ₃	NH ₃	Total Dry N		
M1	-	-	-	-	-	-	-	-	-	4.3	6.9	-		
M2	4.0	8.3	-	-	12.2	0.03	0.4	0.6	0.6	2.0	7.5	11.0		
M4	5.4	7.4	-	-	12.8	0.03	0.3	0.7	0.5	2.8	4.7	9.0		
M5	-	-	-	-	-	-	0.5	-	-	-	-	-		
M6	5.6	9.1	-	-	14.6	0.02	0.3	0.8	0.7	2.9	6.5	11.1		
M7	-	-	-	-	-	-	-	-	-	-	-	-		
M8	-	-	-	-	-	-	-	-	-	-	-	-		
M10	-	-	-	-	-	-	-	-	-	-	-	-		
M11	1.5	2.8	8.1	7.6	20.0	-	-	1.3	2.4	3.3	7.1	14.1		
M12	5.4	11.0	-	-	16.5	0.04	0.4	0.4	0.3	0.5	2.2	3.9		
M13	-	-	4.1	-	-	-	-	-	-	4.5	4.6	-		
M14	-	-	-	-	-	-	-	-	-	-	-	-		



Supplementary Material

2 Figures

Figure S1





Figure S1 Gas-particle conversion of S and N from observation and individual models at EANET sites. The unit is μmole (S or N) m⁻³. Values are calculated by annual average data.

Figure S2 21



22 23 24 Figure S2 The inter-model variations of PM and components among models. The 1sd is the 1 standard deviation

among models (μ g m⁻³). The 1sd% is calculated by dividing 1sd by MMM (%).