Response to Comment 1 by Anonymous Reviewer 1

We thank you for providing helpful and constructive comments and suggestions. We have revised our manuscript accordingly. We hope that these revisions satisfactorily address all the points you have raised. Our point-by-point responses are provided below, and revisions are indicated in blue in the revised manuscript.

General Comments:

Authors compare nine meteorology-chemical transport model systems to estimate deposition amount of Sulfur, Nitrogen, and Ammonia. In general, this manuscript is well organized and delivers informative results and an interesting air quality issue over the Northeast Asia. For the model performance evaluations, authors compare the model outputs to the EANET observations. It is reasonable. However, those nine models may already present appreciable differences in airborne concentrations (authors address the companion papers in the text, but short discussion would be helpful for other readers), or may estimate different wet deposition amounts even under the same atmospheric concentrations due to difference in the implemented dry deposition mechanism in the models. Therefore, it would be better to explain more direct relationship between concentrations and dry/wet depositions for model inter-comparisons.

Authors evaluate the precipitation simulations, but wet deposition in CMAQ (6 out of 9 models) not only depends on the precipitation but the types (convective or nonconvective). Moreover, water mixing ratio also plays an important role in CMAQ depending on the meteorological conditions. Those analyses will be very helpful to the air quality research community.

Reply:

We agree that the simulated concentrations are needed to explain the differences in wet deposition identified in this study. However, because this study is part of the MICS-Asia project, we prefer not to explicitly show the simulated concentration fields within this manuscript to avoid redundancy with respect to our companion papers that were published in a special issue of MICS-Asia Phase III. Instead, to address your comment, we have added references to our companion papers (Chen et al., 2019; Tan et al., 2019). Additionally, we have evaluated wet deposition at the same observation sites for

consistency and prepared one additional figure (Figure S1 in the revised supporting material) and three additional tables (Tables S1, S2, and S3 in the revised supporting material). These points have now been addressed as independent paragraphs in Sections 3.2.1, 3.2.2, and 3.2.3.

In response to your comment about the precipitation types, please see our reply to your comment about Figure 2.

The revisions for further discussing atmospheric concentrations are as follows:

In Section 3.2.1:

"The model performances for atmospheric concentrations were presented in our companion paper (Figs. 3 and 5 and Table 2 of Chen et al., 2019). For consistency with that companion paper, we also performed the model evaluation at the same sites used for the analysis of atmospheric concentrations. The results are shown in Table S1 and the correspondence between the NMB of atmospheric concentration and wet deposition is shown in Figure S1 (a). The modeling performances were generally similar for the wet deposition of S using all data (Table 1) and using limited data (Table S1). Models M1, M2, M4, M5, M6, and M13 underestimated atmospheric concentrations of SO_4^{2-} over Asia, with an NMB of around –30 to –20% (Table S1) and accordingly these models also underestimated the wet deposition of SO_4^{2-}. Only models M12 and M14 overestimated atmospheric concentrations of SO_4^{2-} and model M14 was also distinguished by the overestimation of the atmospheric concentration of SO_4^{2-} over coastal regions, such as over Korea and Japan. Model M11 was the only model to overestimate the wet deposition of SO_4^{2-} and produced the largest underestimation of the atmospheric concentration of SO_4^{2-}, with an NMB of –34.5%. These overestimations (underestimations) of the atmospheric concentration of SO_4^{2-} are closely related to the underestimation (overestimation) of wet deposition of $SO4^{2-}$ found in models M11, M12, and M14. The close relationship between atmospheric concentration and wet deposition was also observed in a model inter-comparison study in Japan (Itahashi et al., 2018c). The atmospheric concentration of SO_4^{2-} was underestimated, especially in winter (Fig. 5 of Chen et al., 2019). Another companion paper (Tan et al., 2019) investigated the sulfur oxidation ratio, which represents the conversion rate from SO_2 to SO_4^{2-}. The observation-based ratio was 0.25. Models M1 and M13 both predicted a comparable

sulfur oxidation ratio of 0.26; however, models M2, M4, M5, and M6 underestimated the ratio, giving values of around 0.16–0.20, suggesting the insufficient oxidation from the precursor of SO_2 (Fig. 2 of Tan et al., 2019). The sulfur oxidation ratio was strongly underestimated by model M11, which gave a value of 0.12. This underestimation can be corrected by refining the treatment of catalysis using O_2, introducing the aqueous-phase production of SO_4^{2-} using NO_2, or newly established gas-phase oxidation by the stabilized Criegee intermediate (Itahashi et al., 2018b, 2018c, 2019). Moreover, another study pointed out that heterogeneous chemistry is a possible explanation for the missing production of SO_4^{2-} in models (Zheng et al., 2015; Shao et al., 2019). The modeled sulfur oxidation ratios of model M12 and M14 were 0.33 and 0.57, respectively; that is, the ratios were overestimated. This overestimation is one reason for the overestimation of the atmospheric concentration of SO_4^{2-} by models M12 and M14. In summary, for models M11, M12, and M14, the model performance for the wet deposition of S is characterized by a close relationship with either the overestimation or underestimation of atmospheric concentration, and models M1, M2, M4, M5, M6, and M13 underestimated both atmospheric concentration and wet deposition of S. ”

In Section 3.2.2:

“The model performances for the atmospheric concentration of NO_3^- in our companion paper also showed large differences between models (Figs. 3 and 5 and Table 2 of Chen et al., 2019). The model evaluation for the analysis of atmospheric concentrations for N was conducted at the same sites as those for S (Table S1), as shown in Table S2. The correspondence between the NMB of atmospheric concentration and the NMB of wet deposition is shown in Figure S1 (b). Models M2, M4, M5, and M6 showed underestimation, whereas models M1, M11, M12, M13, and M14 showed overestimation. Models M1, M2, and M5 showed better performance in terms of NMB (NMBs of between –10% and 10%) (Table S2). If both H_2SO_4 and HNO_3 are present, H_2SO_4 preferentially reacts with NH_3, and therefore NH_4NO_3 is produced only if excess NH_3 is present. The underestimation of the atmospheric concentration of SO_4^{2-} can lead to the overestimation of the atmospheric concentration of NO_3^-. This can explain the performance of models M1, M11 and M13 but not that of models M12 and M14 because they overestimated the atmospheric concentrations of both SO_4^{2-} and NO_3^-. Another companion paper revealed

that models M12 and M14 also used a higher nitrogen oxidation ratio (i.e., the ratio of oxidation from NO_2 to NO_3^-) than that of other models and observation, in addition to using a higher sulfur oxidation ratio (Fig. 2 of Tan et al., 2019). The higher oxidation capacity in models M12 and M14 is connected to the overestimation of the atmospheric concentration of both SO_4^{2-} and NO_3^-. On the other hand, models M2, M4, M5, and M6 underestimated the atmospheric concentration of both SO_4^{2-} and NO_3^-. These four models of M2, M4, M5, and M6 also had lowernitrogen oxidation ratios of between 0.08 and 0.14 than the observed value of 0.18 (Fig. 2 of Tan et al., 2019). In summary, for the wet deposition of N, all models except M5 and M11 underestimated this parameter; however, the relationship between the wet deposition of N and the atmospheric concentration of was not obvious different from this relationship for S. Because the correlation coefficient for the model performance of the wet deposition of N is lower than that for S, future studies should focus on N and related species in greater detail. Our future companion paper will attempt a detailed analysis of N using an intensive observation network over China."

In Section 3.2.3:

"Our companion paper reported model performances for the atmospheric concentration of NH_4^+ (Figs. 3 and 5 and Table 2 of Chen et al., 2019). The model evaluation for the analysis of atmospheric concentrations for A was conducted at the same sites as those for S and N (Tables S1 and S2), as shown in Table S3, and the correspondence between the NMB of atmospheric concentration and that of wet deposition is shown in Figure S1 (c). Generally, the behavior of NH_4^+ is associated with the atmospheric concentrations of SO_4^{2-} and NO_3^- as counterions. The studied models generally underestimated the atmospheric concentration of S and overestimated the atmospheric concentration of N; consequently, all models except M4 overestimated the atmospheric concentration of A. The reason for the different behavior of model M4 is that this model underestimated atmospheric concentrations of NO_3^-. In general, the models overestimated the atmospheric concentration of A and underestimated the wet deposition of A (Fig. S1 (c)); this indicates a close relationship between atmospheric concentration and wet deposition processes. "

Finally, we also added the following to the concluding section:

"Comparisons of atmospheric concentrations revealed that model performances are either characterized by a close relationship between the overestimation (underestimation) of the wet deposition of S and the underestimation (overestimation) of the atmospheric concentration of S or characterized by the underestimation of both the atmospheric concentration and wet deposition of S species. The general features for underestimation of wet deposition of A and overestimation of atmospheric concentration of A were clarified. The relationships between atmospheric concentration and wet deposition of N are complicated and further research focusing on nitrogen species especially targeting the nitrogen cycle is required."

Scientific Comments:

Table 1: It would be better to include the WRF configurations. Physical options in WRF may affect the wet depositions of those target species.

Reply:

We are grateful for this helpful comment. Because almost all the models considered in this study are driven by the WRF model, we have revised the main manuscript to explicitly mention the WRF configurations. An explanation about the WRF configurations has been added to Section 2.1, as follows:

"The WRF is configured as follows: longwave radiation is computed with the rapid radiative transfer model (Mlawer et al., 1997), shortwave radiation with the Goddard scheme (Chou et al., 1994; Matsui et al., 2018), microphysics with Lin's scheme (Chen et al., 2002), cumulus physics with the Grell 3D ensemble scheme (Grell, 1993; Grell and Devenyi, 2002), the planetary boundary layer with the Yonsei University scheme (YSU) (Hong et al., 2006), the surface layer with the revised Fifth-Generation Penn State/NCAR Mesoscale Model (MM5) (Jimenez et al., 2012), and land surface with the unified Noah model (Tewari et al., 2002). The WRF also includes the urban canopy model (Chen et al., 2011)."

Figure 2: Wet scavenging is affected by not only the precipitation amounts but also precipitation

intensity (and the lasting time), and types (at least in CMAQ parameterization). For example, convective precipitation in MCIP outputs for CMAQ may increase the total rain amounts in summer but may have less influence on fine particle removal, compared to non-convective rain during spring and fall. In case of wet removals of gases species, total surface areas of rain droplets as function of droplet size and ambient bulk concentrations would be important. Size distributions and concentrations of ambient aerosol would be critical for wet scavenging.

Reply:

We agree that precipitation type on the analysis of wet deposition is important. In the framework of the current MICS-Asia Phase III activity, all the submitted results for wet deposition were the sums of wet depositions caused by convective and non-convective precipitation and it is difficult to distinguish between the two types.

We will consider this point in the strategy for wet deposition in the next phase of MICS-Asia, which we are now planning. Additionally, we have revised Section 5 to mention this explicitly as follows:

"Moreover, precipitation type (convective or non-convective) should be analyzed and the impacts of differences in the characteristics of fine and coarse particles on wet deposition should be investigated."

Figure 3: Wet depositions in current three dimensional grid models (CMAQ and CAMx) deal with both in-cloud and below-cloud scavenging together. It would be okay to explain total wet depositions, but may mislead to the comparisons to the EANET measurements.

Reply:

EANET ground-based observations use wet-only samplers and measure wet deposition (volume-weighted mean concentrations and precipitation); therefore, these EANET measurement data and model output data as wet deposition could be comparable.

Figures 3-8: In terms of wet deposition comparisons, Figures and their explanations are well represented in the text. Considering that wet scavenging amounts are determined by airborne concentrations and removal mechanism, it is expected to relate modeled concentrations and wet deposition amounts, including the removal module used in the models.

Reply:

Please see our reply to your 'General Comments'. In the revised manuscript, we have added a detailed discussion of the ambient concentration by appropriately referring to our companion papers, not simply citing them.