

The authors appreciate the reviewers for reviewing our manuscript and providing constructive comments. As suggested, we carefully revised the manuscript thoroughly according to the valuable advices, as well as the typographical, grammatical, and bibliographical errors. Listed below are our point-by-point responses in blue to the review's comments (in italic).

**Anonymous Referee #1**

*This manuscript has presented the analyses of atmospheric nitrogen deposition to China as simulated by an ensemble of chemical transport models participating the MICS-Asia III model intercomparison. Available surface measurements of wet deposition fluxes are integrated to assess the model performances. This represents an important step towards a better understanding the model range and uncertainties in simulating nitrogen deposition. Different from previous studies on multi-model nitrogen deposition simulation, most models analyzed in this study used the same emissions and driving meteorology, allowing a closer attribution of the factors driving the model uncertainties. The results show that most models calculated consistent spatial and temporal variations of nitrogen deposition for both oxidized and reduced nitrogen, yet considerable differences exist among models.*

*I think the study is an important contribution to the MICS-Asia III special issue. The analyses are mostly fine, and it would be much scientifically stronger if having a deeper investigation on the drivers of model*

*differences. The following comments also need to be addressed. In addition to my specific comments as elaborated below, improvements on the language are necessary and need caution.*

**[Response]:** We would like to thank the reviewer for the valuable comments. Deeper discussion on the difference among the models as well as the difference between models and observations have been added in the revised manuscript. We also invite a native speaker to polish the language, which is mentioned in the acknowledgment. All the revision in the manuscript has been marked in blue. Following are the responses to the comments.

**Specific comments:**

*1. Page 5, Line 199-202: How about natural sources of nitrogen, e.g., nitrogen oxides from soil and lightning? Are they included in any of these models?*

**[Response]:** No, the nitrogen oxides emission from soil and lightning are not included in MICS-Asia III. The natural sources included in this project are biogenic emissions from MEGANv2.4 and biomass burning emissions from GFEDv3.

*2. Page 6, Line 220-223: Did any of the models also simulate dry deposition of other nitrogen species, e.g., PAN, isoprene nitrates? How*

*important are these nitrogen species contributing to dry deposition, and the uncertainty induced by excluding them in the analysis? Please clarify.*

**[Response]:** Unfortunately, the PAN as well as isoprene nitrates are not included in the simulated dry deposition in this study. PAN is an important photochemical product formed from the reactions between VOCs and NO<sub>x</sub> under sunlight. The loss of PAN was modulated mainly by dry deposition and horizontal transport (Yuan et al., 2018). However, their contributions to total N dry deposition are less important than the inorganic N, e.g., HNO<sub>3</sub> and NO<sub>x</sub>. A comparison between the Chemistry of Atmosphere-Forest Exchange (CAFE) Model and BEARPEX-2007 observations in California has been implemented and found that the HNO<sub>3</sub> dominate total dry deposition of oxidized N (~83%) in warm seasons, which indicated the other NO<sub>y</sub> (including NO<sub>x</sub>, PANs, etc) may take up less than 17%. (Wolfe et al., 2011). Besides, several studies also investigated the Organic N (ON) deposition accounted for about 20–30% of total N compounds in wet and dry deposition (Duce et al., 2008; Benitez et al., 2009). Thus, the uncertainties of excluding other nitrogen species, e.g., PAN, isoprene nitrates from the total dry deposition could be negligible compared with the uncertainties of the simulated HNO<sub>3</sub> and NH<sub>3</sub> dry deposition in this study. We have added this interpretation in our revised manuscript.

3. Page 7, Line 261: *It is not clear how the data are normalized as monthly wet deposition fluxes. Do you mean that the raw measurements are at different temporal resolutions (daily, weekly, etc.), and then are interpolated to monthly values? This shall be explained in the text.*

**[Response]:** Yes. Since the measured wet deposition data are collected from different sources, the temporal resolutions of the data are different from each other, i.e., daily in EANET and CREN, rainfall event collection in DEE and yearly in NNDMN, which has been mentioned in sec.2.3 L237-259 in the original manuscript. We have added the explanation in the revised manuscript, as ” *The temporal resolutions of the wet deposition data are different from each other, i.e., daily in EANET and CREN, rainfall event collection in DEE and yearly in NNDMN. For the convenience of comparison, all data from daily or rainfall event collecting samples at each type of measurement site has been summarized and interpolated as monthly wet deposition data to compare with the monthly simulation in MICS-Asia III in this study, except the yearly data provided by NNDMN*”.

4. Page 7, Line 279-281: *This sentence is not clear. Do you mean the correlation coefficients are lower than the value when only comparing with EANET data? Please clarify.*

**[Response]:** Yes. The EANET data used in this study is only 8 sites located in China (Supplementary material), which is different from the

whole EANET sites over East Asia listed in our companion paper (Itahashi et al., 2020). To avoid misunderstanding, the EANET sites used in this study are clarified as EANET sites in China in the revised Supplementary material. Meanwhile, the context in first paragraph of Section 3.1.1 has been revised as “*The NME was around 50% with the highest 82.2%, in M11, which is lower than that reported over the East Asia with the value of 70% based on EANET observation by Itahashi et al. (2020). However, the correlation coefficient R was around 0.2~0.3 (n=83) which is lower than the East Asia comparison based on the EANET data (0.3~0.4, n=54) (Itahashi et al., 2020)*”.

5. Page 7, Line 285-286: Need to add a sentence defining FAC2.

**[Response]:** It has been added in the revised manuscript as “*To judge the agreement between simulation and observation, the percentages within a factor of 2 (FAC2) has been calculated in this study.*”

6. Page 10, Line 417-423: The sentence here needs rewritten or removed.

*It is a long sentence, and the information is repetitive in the paragraph.*

**[Response]:** Agree. The sentence here has been removed in the revised manuscript.

7. Page 12, Line 478-480: Is Figure 8 for the surface layer or the atmospheric column? As dry deposition only applies to species at the surface layer, while wet deposition can extend to the whole tropospheric

*column, an explanation is needed here to justify why you use it for both dry and wet depositions.*

**[Response]:** Figure 8 is the CV of the related species' concentration in surface layer. It is indeed that the wet deposition can extend to the whole column through in-cloud and below-cloud scavenging process. However, the simulated data for most of the air concentrations in MICS-Asia III were at the surface layer except NO<sub>2</sub>, which also included the vertical column density (VCD) data. As for NO<sub>2</sub> VCD, the CV results show that the similar spatial distribution with that in surface layer (Figure S9 in revised supporting material). This indicated that the simulated concentration at surface layer could partly represent the differences of the whole column among the multi-models, especially in providing a broad overview in China. In this study, the CV of the related air mass concentrations at surface layer has been calculated and compared with that both in dry and wet deposition to explain the reasons for the differences among the simulated depositions in MICS-Asia III. The explanation has been added in the revised version. *“It should be noted that only concentrations of most of the related species at surface layer have been submitted in MICS-Asia III, except NO<sub>2</sub> vertical column density data (VCD). According to the comparison of CV between the NO<sub>2</sub> concentration at the surface layer and VCD data (Figure S5), it was shown that there is a similar spatial pattern over the whole of China. This*

*indicates that the simulated concentration on the surface layer can partly represent the difference of the whole column among the multi-models, especially in providing a broad overview in China. Thus, only concentration data at the surface layer has been used in this study.”*

*8. Page 12, Line 500-510: How about dry deposition velocities? Did all the models calculate the dry deposition fluxes as the products of surface concentration and dry deposition velocity? It is missing something that the discussion of dry deposition only examined concentrations and not include dry deposition velocities.*

**[Response]:** Thank you for suggestion. We have conducted the analysis of dry deposition velocities based on the ratio of the dry deposition fluxes and the surface concentration simulated by each model and prepared additional Figure S6~Figure S11 in the revised supporting material. These points have now been addressed in the last paragraph in Section 3.3.2.

*“For  $N_{ox}$  dry depositions, the anomalies of deposition and  $NO_x$  concentration in the air are shown in Figure S6 and Figure S7. It shows same variations among the models, i.e., both of higher deposition and concentration in M1, M5, M11, M13, and lower in M2, M4, M6, M12 and M14. This has also been proved in the distribution of CV values in  $NO_x$  (Figure 8a) and  $N_{ox}$  dry depositions (Figure 7a) during autumn and winter. As the most important precursor of  $N_{rd}$  dry deposition, gaseous*

*NH<sub>3</sub> also shows large CV values in central China during summer time (> 0.6). There were also significant high CV values in south of the Yangtze River during the autumn and winter period (0.7-0.8 in south of the Yangtze River vs 0.3-0.5 in north of the Yangtze River). A similar pattern but of uncertain significance was found in the simulated N<sub>rd</sub> dry deposition (0.3-0.4 vs 0.2-0.3 in Figure 7b). The anomalies of N<sub>rd</sub> dry deposition and the gaseous NH<sub>3</sub> in the air simulated by each model are shown in Figure S8 and Figure S9. Additionally, the dry deposition velocity (V<sub>d</sub>) of N<sub>rd</sub> - based on the ratio of the dry deposition fluxes and the surface concentration (same as Tan et al., 2019) - are also shown in Figure S10. The results show that the CMAQ models (M1~M6) predicted similar V<sub>d</sub> of N<sub>rd</sub>, and the N<sub>rd</sub> dry deposition was consistent with the gaseous NH<sub>3</sub> concentration in the air, i.e., overestimation in M1 and M2 but underestimation in M4 and M5. However, among the different models, high V<sub>d</sub> of N<sub>rd</sub> (low V<sub>d</sub> of N<sub>rd</sub>) was corresponds with the overestimation (underestimation) of dry deposition in M11 and M14 (M12 and M13). From the distribution of CV, similar patterns were also displayed both in V<sub>d</sub> (Figure S11) and dry deposition of N<sub>rd</sub>, with low CV value in NCP (0.1-0.4 for N<sub>rd</sub> dry deposition, 0.1-0.3 for V<sub>d</sub>) and high CV value in SE and SW (0.4-0.8 for N<sub>rd</sub> dry deposition, higher than 0.5 for V<sub>d</sub>).”*

*9. Page 13, Line 538-548: The discussion of different allocation is not clear and may not correct. Do you mean dry deposition or wet deposition*



*vs. gas column concentrations? From Figure 9, the spatial distributions of dry deposition of oxidized and reduced nitrogen are rather consistent with their column concentrations. Also the discussion of conservations is not clear. Higher emissions would have higher depositions as both oxidized and reduced nitrogen have short lifetimes, and the differences between emissions and depositions do not reflect their concentrations in the air.*

**[Response]:** We agree with that the differences between emissions and depositions do not reflect their concentrations in the air. The chemical transformation as well as the regional transport may also affect their atmospheric concentration in the air. The purpose of this discussion is to validate the reasonable distribution of the simulated depositions through the comparison between  $N_{ox}$  and  $N_{rd}$ , instead of the difference between deposition and gas column concentrations. To address the issues in your comment, the last paragraph of section 4.1 has been revised as:

*“Another interesting phenomenon is that the allocations of high values of depositions and VCD of  $N_{ox}$  are different from that of  $N_{rd}$ . As shown in the Figure 9, low depositions with high values of VCD for  $N_{ox}$  and high depositions with comparatively lower level of VCD for  $N_{rd}$  co-existed in East China. On a global scale, air pollutants must follow the conservation law - that is, the emissions can be divided into two parts, i.e., the depositions and their concentrations in the air. Here we apply this*

*concept to the entire region of China. We assume that the amount of  $N_{ox}$  and  $N_{rd}$  transported out of the research areas is equivalent under the same atmospheric advection. The emissions of  $N_{ox}$  and  $N_{rd}$  in China are also comparable ( $8.3 \text{ kg N}\cdot\text{ha}^{-1}$  and  $8.7 \text{ kg N}\cdot\text{ha}^{-1}$  for  $\text{NO}_x$  and  $\text{NH}_3$ , respectively). At the same time, the simulated low deposition of  $N_{ox}$  and observed high VCD match exactly with the high deposition in  $N_{rd}$  and observed low VCD in central and eastern China. Although there is no directly observed distribution map to verify the total deposition in our models, the close correlation between the observed VCD and deposition can verify the rationality of the simulated total deposition distribution.”*

*10. Page 14, Line 591-592: “higher contribution of  $\text{Nox}$  to TIN in SE indicated more oxidant ratio of the precursors than NC”. It is not clear what “more oxidant ratio of the precursors” means. Please clarify.*

**[Response]:** The “more oxidant ratio of the precursors” means higher nitrogen oxidant ratio (i.e., the ratio of oxidation from  $\text{NO}_2$  to  $\text{NO}_3^-$ ). According to your comment, the sentence has been revised as “Considering the lower ratio of  $\text{NO}_x/\text{NH}_3$  emission in SE (21.4/21.6, 0.99) than NC (30.4/24.4, 1.25), higher contribution of  $N_{ox}$  to TIN in SE indicated a higher nitrogen oxidant ratio (i.e., the ratio of oxidation from  $\text{NO}_2$  to  $\text{NO}_3^-$ ) than NC. Our companion paper (Tan et al., 2019) also revealed the higher nitrogen oxidation ratio in SE as 0.4-0.6, compared with that in NC as 0.2-0.4.”

**Technical comments:**

1. Page 5, Line 194: “US 25 National Aeronautics and Space Administration”, should delete “25” here?

**[Response]:** It has been deleted in the revised manuscript.

2. Page 8, Line 322: “almost double higher than”, should be “almost double” or “almost a factor of 2 higher than”

**[Response]:** It has been replaced by “almost double” in the revised manuscript.

3. Page 12, Line 488: What do you mean by “the correlated consistence”?

**[Response]:** The correlated consistence here means the similar variation of CV in the simulated particulate  $\text{NO}_3^-$  concentration in the air mass and  $\text{N}_{\text{ox}}$  wet deposition were shown in Figure 7 (c) and 8 (a). To avoid misunderstanding, the “correlated consistence” has been replaced as “consistent distribution of CV” in the revised manuscript.

4. Page 12, Line 496: “the magnitude difference”, do you mean “large differences”?

**[Response]:** Yes, the difference can reach at magnitude level. It has been replaced by *“large difference even at magnitude level”* in the revised manuscript.

5. Page 12, Line 510: *“this need to be”* should be *“this needs to be”*

**[Response]:** It has been changed in the revised manuscript.

6. Page 13, Line 547: What do you mean by *“this conservation data”*

**[Response]:** The *“conservation data”* means that the simulated low deposition in  $N_{ox}$  and observed high VCD match exactly with the high deposition in  $N_{rd}$  and observed low VCD in central and eastern China. This phenomenon was perfectly constrained by the comparable emissions between  $N_{ox}$  and  $N_{rd}$  in China. In avoid to misunderstanding, these expressions have been deleted. Detailed information could be found in the response to specific comment 9.

7. Page 13, Line 555-558: Use *“major contributions”* to denote *“18”*

**[Response]:** The *“major contributions”* have been revised as *“important contributions”* in the revised manuscript.

8. Page 14, Line 572: Change *“increasing trend”* to *“increasing order”*

**[Response]:** It has been changed accordingly.

9. Page 14, Line 579: Change *“correspondingly”* to *“corresponding”*

**[Response]:** It has been changed accordingly.

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