

## A point-to-point response to the referee's comments

### Reply to anonymous referee #2

On behalf of my co-authors, we would like to express our great appreciation for your constructive comments and feedback on our manuscript entitled “Increased Importance of Aerosol-Cloud Interaction for Surface PM<sub>2.5</sub> Pollution Relative to Aerosol-Radiation Interaction in China with Anthropogenic Emission Reduction” (egosphere-2023-502). We have studied your comments carefully and have revised our manuscript accordingly. Replies to comments are in blue, and the line numbers in the response refer to the clean revised manuscript.

### General comments:

The paper titled “Increased Importance of Aerosol-Cloud Interaction for Surface PM<sub>2.5</sub> Pollution Relative to Aerosol-Radiation Interaction in China With the Anthropogenic Emission Reduction” by Gao et al. investigates changes of the ARI- and ACI-induced PM<sub>2.5</sub> enhancements for 2013–2021 in China under the background of sharp anthropogenic emission reduction by using WRF-Chem model, and the causes for these changes was further explored from the perspectives of meteorological factors and PM<sub>2.5</sub> concentration distribution. With this, the authors claimed that the ACI-induced PM<sub>2.5</sub> enhancement inevitably becomes more important with the decrease of ambient PM<sub>2.5</sub>, that needs to be considered in the formulation of control policies. These results are interesting and useful, although some issues are needed to be clarified and properly improved.

**Response:** Thank you for your careful evaluation of this manuscript. All your comments and suggestions are important to us. They have direct significance to our paper writing and research work.

### Specific and technical comments:

1. Model configuration, I note that the regular chemistry modules like SAPRC-99 and MOSAIC were used in WRF-Chem, with no improvement or revision. Interestingly, the simulated PM<sub>2.5</sub> was correlated well with the observed values, especially during the heavy polluted period in January, 2013, during which period numerical models usually reported much lower values as suggested by previous studies. Thus, I hope the authors could give the comparison results between the simulated and observed particle chemical components in January 2013, as it had been provided in Fig. S4, which is very important to explore the role of ACI-induced secondary PM<sub>2.5</sub> component enhancement.

Response: Thank you for your comment. We did try to improve the PM<sub>2.5</sub> simulation before starting this work. Specifically, we noted the poor ability of nitrate simulation in the WRF-Chem model. Previous studies reported that the nitrate underestimation might be attributed to the HONO underestimation (Wang et al., 2015; Xue et al., 2020). The source of HONO is originally from some gas-phase chemical reactions in the WRF-Chem, but other HONO sources are lacking, such as hydrolysis of NO<sub>2</sub> on humid aerosol surfaces and heterogeneous conversion of NO<sub>2</sub> on ground surfaces. It also has been confirmed that these reactions could occur in the atmosphere (Li et al., 2018, Liu et al., 2019). We added these four heterogeneous HONO reactions to the WRF-Chem model (Table S1). The addition of these four reactions enhances atmospheric oxidant and promotes OH radical formation, thereby promoting the formation of gaseous nitric acid through strong OH radicals reacting with NO<sub>2</sub>. In addition, the reactions of hydrolysis of NO<sub>2</sub> on humid aerosol surfaces contributed extra nitric acid. More nitric acid is beneficial for nitrate formation through condensation and heterogeneous reactions. Other specific information can be found in Zhang et al. (2021). This improvement has been added in the revised Supplementary Information. Please see lines 27-43.

Lines 27-43: Before starting this work. We noted the poor ability of nitrate simulation in the WRF-Chem model. Previous studies reported that the nitrate underestimation might be attributed to the HONO underestimation (Wang et al., 2015; Xue et al., 2020). The source of HONO is originally from some gas-phase chemical reactions in the WRF-Chem, but other HONO sources are lacking, such as hydrolysis of NO<sub>2</sub> on humid aerosol surfaces and heterogeneous conversion of NO<sub>2</sub> on ground surfaces. It also has been confirmed that these reactions could occur in the atmosphere (Li et al., 2018, Liu et al., 2019). We added these four heterogeneous HONO reactions to the WRF-Chem model (Table S1). The addition of these four reactions enhances atmospheric oxidant and promotes OH radical formation, thereby promoting the formation of gaseous nitric acid through strong OH radicals reacting with NO<sub>2</sub>. In addition, the reactions of hydrolysis of NO<sub>2</sub> on humid aerosol surfaces contributed extra gaseous nitric acid. More gaseous nitric acid is beneficial for nitrate formation through condensation and heterogeneous reactions. Other specific information can be found in Zhang et al. (2021).

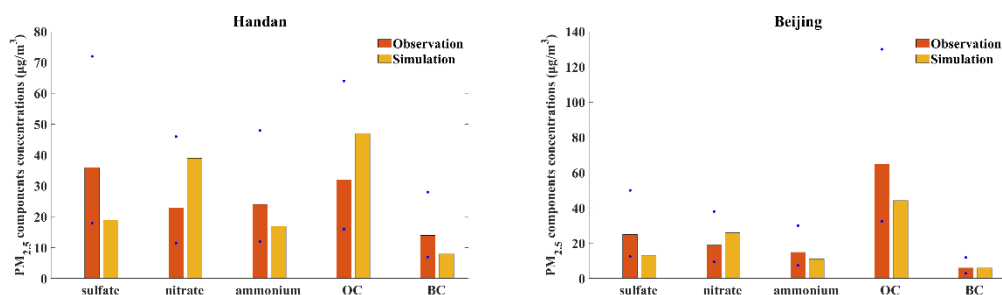
Table S1. Heterogeneous HONO reactions added in WRF-Chem (from Zhang et al., 2021).

number	Reaction	Reference
(1)	$\text{NO}_2 + \text{aerosol} = 0.5 \times \text{HONO} + 0.5 \times \text{HNO}_3$	Liu et al., (2019)
(2)	$\text{NO}_2 + \text{aerosol} = 0.5 \times \text{HONO} + 0.5 \times \text{HNO}_3$	Liu et al., (2019)
(3)	$\text{NO}_2 + \text{ground} = \text{HONO}$	Li et al., (2018), Liu et al., (2019)
(4)	$\text{NO}_2 + \text{ground} + h\nu = \text{HONO}$	Liu et al., (2019)

As reviewer mentioned, we reproduce the PM<sub>2.5</sub> pollution in January 2013 in China accurately. The PM<sub>2.5</sub> components data in January 2013 are very rare, and we sourced three sets of data, in Beijing, Handan, and Shanghai. As shown in Fig. S4, we find that the simulated PM<sub>2.5</sub> components in January 2013 are reproduced well generally, but they still have small discrepancies compared with the observations in three cities. Specifically, the simulated PM<sub>2.5</sub> components are larger than half of observational PM<sub>2.5</sub> components and less than the double observational PM<sub>2.5</sub> components (Fig. S4).

We also compared our simulated results with previous studies. We find that some previous studies have also successfully reproduced the PM<sub>2.5</sub> pollution in January 2013, but they also have certain discrepancies in the PM<sub>2.5</sub> components (Li et al., 2015; Mattias et al., 2017). Taking modeling in Beijing and Shanghai as two examples, Mattias et al. (2017) found that their model (COSMO-CLM-CMAQ) is generally able to reproduce the high PM<sub>2.5</sub> level measured in situ close to the ground in Beijing, but it overestimates the BC concentration. Li et al., (2015) reported that the simulated PM<sub>2.5</sub> concentration is slightly overestimated owing to the overestimation of fugitive dust. Therefore, the models all have difficulty in reproducing all the PM<sub>2.5</sub> chemical components very accurately. For 2013, our simulated PM<sub>2.5</sub> concentrations in January and July are good and simulated PM<sub>2.5</sub> components in January have small discrepancies with the observations (Fig. S4). The evaluation of the simulation in January 2013 has been added to the revised manuscript. Please see lines 285-291.

Lines 285-291: Given that the PM<sub>2.5</sub> components data in 2013 are very rare, we sourced three sets of data in January 2013, respectively in Beijing (Mattias et al., 2017), Handan (Zhang et al., 2015), and Shanghai (Li et al., 2015). The results show that the simulated PM<sub>2.5</sub> components are reproduced well generally. Specifically, the simulated PM<sub>2.5</sub> components are larger than half of observational PM<sub>2.5</sub> components and less than the double observational PM<sub>2.5</sub> components (Fig. S4).



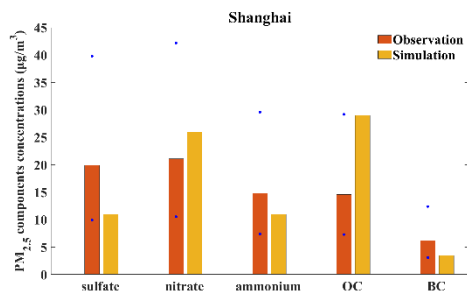


Fig. S4. Comparisons between PM<sub>2.5</sub> component observations and simulations in Handan, Beijing and Shanghai in January 2013. The blue points represent double or half of the PM<sub>2.5</sub> component observations. OA and BC represent organic aerosol and black carbon.

2. Line 274-276, it seems that not only sulfate, but the majority of BC and parts of OC are beyond the ratios of 2.0 in January 2021 (as showed in Fig. S4). I do not think such discrepancies is minor and will not cause obvious uncertainty in this research. The simulated BC alone would cause high uncertainty in the ARI-induced PM<sub>2.5</sub> enhancements. In addition, the higher values in the simulated OC is also weird. Please added more explanation here.

Response: Thank you for your comment. To test the impact of simulated BC overestimation in January 2021 on ARI-induced PM<sub>2.5</sub> enhancement, we utilize another set of particulate matter (PM) source profiles (Liu et al., 2018) and conduct the simulations for January 2021. The results indicate that the ratios of simulated BC concentration to observational BC concentration are within 2.0. The ARI-induced PM<sub>2.5</sub> enhancement is  $1.33 \mu\text{g m}^{-3}$ , which shows a negligible difference from the result ( $1.37 \mu\text{g m}^{-3}$ ) obtained using original PM source profiles (Fig. S6). However, the reduction in simulated BC concentration in January 2021 does not necessarily mean that this set of PM source profiles is better than the original PM source profiles, because this might be an accidental result caused by other uncertainties. For example, the current model underestimates the wet deposition of BC due to neglecting the increase in BC hygroscopicity brought about by BC aging. If this process is considered in the model, simulated BC concentrations might be better reproduced using original PM source profiles. Therefore, in this study, we still use the original results for our analysis.

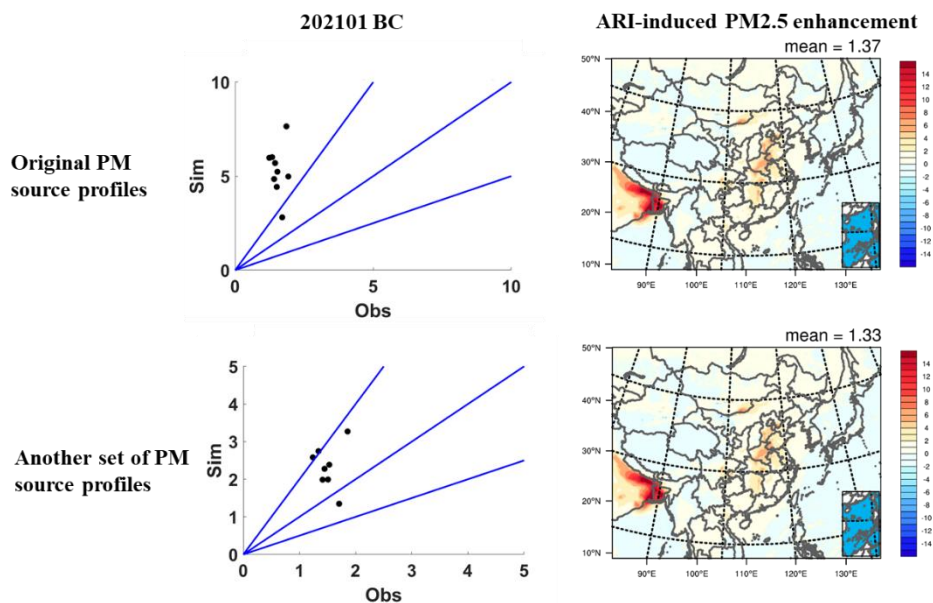


Fig. S6. The ratios of simulation to observation of BC and ARI-induced  $PM_{2.5}$  enhancement (unit:  $\mu g m^{-3}$ ) obtained from original PM source profiles and another set of PM source profiles in January 2021.

The reviewer also identified that the model underestimates the sulfate concentration and overestimates the part of OC concentration in January 2021. We think that neither of these discrepancies will cause significant uncertainties in ARI- and ACI-induced  $PM_{2.5}$  enhancement. Specifically, the majority of aerosol is scattering aerosol and the  $PM_{2.5}$  concentration in January 2021 is reproduced well. Therefore, we think that the impact of the sulfate underestimation on the ARI-induced  $PM_{2.5}$  enhancement would be largely offset by the overestimation of other scattering aerosol components, such as OC. In addition, the OC overestimation should not bring significant uncertainty to ACI-induced  $PM_{2.5}$  enhancement either, because of the relatively lower hygroscopicity of OC compared to secondary inorganic aerosol. The underestimation of sulfate simulation in January 2021 also minimally affects ACI-induced  $PM_{2.5}$  enhancement because the sulfate underestimation mainly occurs in the North China Plain, where cloud cover is low. In contrast, in southern cities such as Mianyang city in Sichuan province where there is plenty of cloud cover, the sulfate simulation was  $4.19 \mu g m^{-3}$  in January 2021, which is very close to the observed value of  $4.25 \mu g m^{-3}$  (Lin et al., 2022).

Corresponding discussions have been added in lines 292-332.

Lines 292-332: Fig. S5 shows the ratios of observation to simulation of ammonium, sulfate, BC and organic carbon (OC) in January and July 2021. The results exhibit that almost all the ratios of  $PM_{2.5}$  components are located between 0.5 and 2.0, while some ratios of sulfate in January, part of OC in January, and BC in January and July

are beyond this range. But these discrepancies will not cause obvious uncertainties in this research. Specifically, considering BC low hygroscopicity, BC overestimations in January and July 2021 probably bring low uncertainties in ACI-induced  $PM_{2.5}$  enhancement. To test the impact of simulated BC overestimation in January 2021 on ARI-induced  $PM_{2.5}$  enhancement, we utilize another set of particulate matter (PM) source profiles (Liu et al., 2018) and conduct the simulations for January 2021. The results indicate that the ratios of simulated BC concentration to observational BC concentration are within 2.0. The ARI-induced  $PM_{2.5}$  enhancement is  $1.33 \mu\text{g m}^{-3}$ , which shows a negligible difference from the result ( $1.37 \mu\text{g m}^{-3}$ ) obtained using original PM source profiles (Fig. S6). In view of the results in January 2021, the BC overestimation in July 2021 also probably brings low uncertainties in ARI-induced  $PM_{2.5}$  enhancement. However, the reduction in simulated BC concentration in January 2021 does not necessarily mean that this set of PM source profiles is better than the original PM source profiles, because this might be an accidental result caused by other uncertainties. For example, the current model underestimates the wet deposition of BC due to neglecting the increase in BC hygroscopicity brought about by BC aging. If this process is considered in the model, simulated BC concentrations might be better reproduced using original PM source profiles. Therefore, in this study, we still use the original results for our analysis. The model also underestimates the sulfate concentration and overestimates the part of OC concentration in January 2021. We think that neither of these discrepancies will cause significant uncertainties in ARI- and ACI-induced  $PM_{2.5}$  enhancement. Specifically, the majority of aerosol is scattering aerosol and the  $PM_{2.5}$  concentration in January 2021 is reproduced well. Therefore, we think that the impact of the sulfate underestimation on the ARI-induced  $PM_{2.5}$  enhancement would be largely offset by the overestimation of other scattering aerosol components, such as OC. In addition, the OC overestimation should not bring significant uncertainty to ACI-induced  $PM_{2.5}$  enhancement either, because of the relatively lower hygroscopicity of OC compared to secondary inorganic aerosol. The underestimation of sulfate simulation in January 2021 also minimally affects ACI-induced  $PM_{2.5}$  enhancement because the sulfate underestimation mainly occurs in the North China Plain, where cloud cover is low. In contrast, in southern cities such as Mianyang city in Sichuan province where there is plenty of cloud cover, the sulfate simulation was  $4.19 \mu\text{g m}^{-3}$  in January 2021, which is very close to the observed value of  $4.25 \mu\text{g m}^{-3}$  (Lin et al., 2022).

3. Line 432-435, it is unclear about the high ambient  $PM_{2.5}$  concentration conditions, is it about the January 2021 or July 2021? If it is about the latter, please provide more discussion about the relationship of PM and CCN to show the CCN is enough.

Response: Thank you for your comment. The description of the high ambient  $PM_{2.5}$  concentration condition originally represents the regional and temporal average  $PM_{2.5}$  concentration in eastern China in January and July, which is simulated using background meteorology in 2021 and emissions in 2013.

The analysis of the relationship between  $PM_{2.5}$  and CCN might not accurately indicate whether there is an adequate amount of CCN because CCN is a diagnostic variable in the model and varies continuously with the change in  $PM_{2.5}$  concentration without being constrained by cloud water. Here, to better elucidate the relationship between the change in  $PM_{2.5}$  concentration and the change in ACI-induced SWDOWN reduction, we find it more straightforward to discuss the impact of  $PM_{2.5}$  reduction on the change in LWP and cloud effective radius ( $Re$ ) induced by ACI. In our simulations, the influence of ACI-induced  $Re$  change is relatively smaller than that of ACI-induced LWP change with a large decrease in  $PM_{2.5}$  concentration (Fig. S7). Therefore, we are only concerned with change in ACI-induced LWP with a reduction in  $PM_{2.5}$ . As shown in Fig. S8a, when the ambient  $PM_{2.5}$  concentration exceeded  $15 \mu\text{g m}^{-3}$ , the decrease in ACI-induced LWP increase is relatively low with a  $PM_{2.5}$  reduction from  $120$  to  $15 \mu\text{g m}^{-3}$ , indicating that aerosols are not a key limiting factor to cloud formation in this range. In contrast, when the ambient  $PM_{2.5}$  concentration decreases from  $15$  to  $0 \mu\text{g m}^{-3}$ , the decrease in ACI-induced LWP increase is much faster. Previous studies have also demonstrated that the decrease in ACI-induced LWP increase is relatively fast or slow with the ambient  $PM_{2.5}$  reduction in the  $PM_{2.5}$ -clean or polluted condition, respectively (Myhre et al., 2007; Savane et al., 2015).

The regional and temporal average  $PM_{2.5}$  concentration in eastern China in January and July simulated using background meteorology in 2021 and emissions in 2013 is  $63$  and  $25 \mu\text{g m}^{-3}$ , which is much higher than  $15 \mu\text{g m}^{-3}$ . Therefore, the decrease in ACI-induced SWDOWN reduction in both months is weak.

We have revised the original descriptions. Please see lines 489-493 in the revised manuscript.

Lines 489-493: The reason is that the decrease in ambient  $PM_{2.5}$  concentration directly weakens the ARI-induced SWDOWN reduction, but it has only a minor impact on the ACI-induced SWDOWN reduction because the change in LWP and cloud effective radius ( $Re$ ) induced by ACI is not sensitive to  $PM_{2.5}$  reduction in the  $PM_{2.5}$ -polluted regime (Fig. S8a).

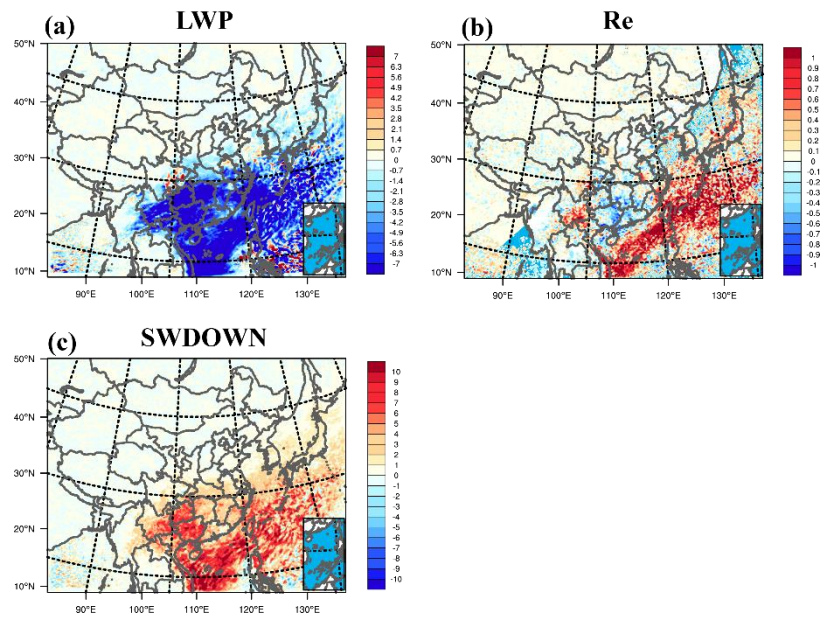


Fig. S7. Change in aerosol-induced LWP (unit:  $\text{g m}^{-2}$ ), Re (unit:  $\mu\text{m}$ ) and ACI-induced SWDOWN reduction (unit:  $\text{W m}^{-2}$ ) in January owing to the emission reduction from 2013 to 2021.



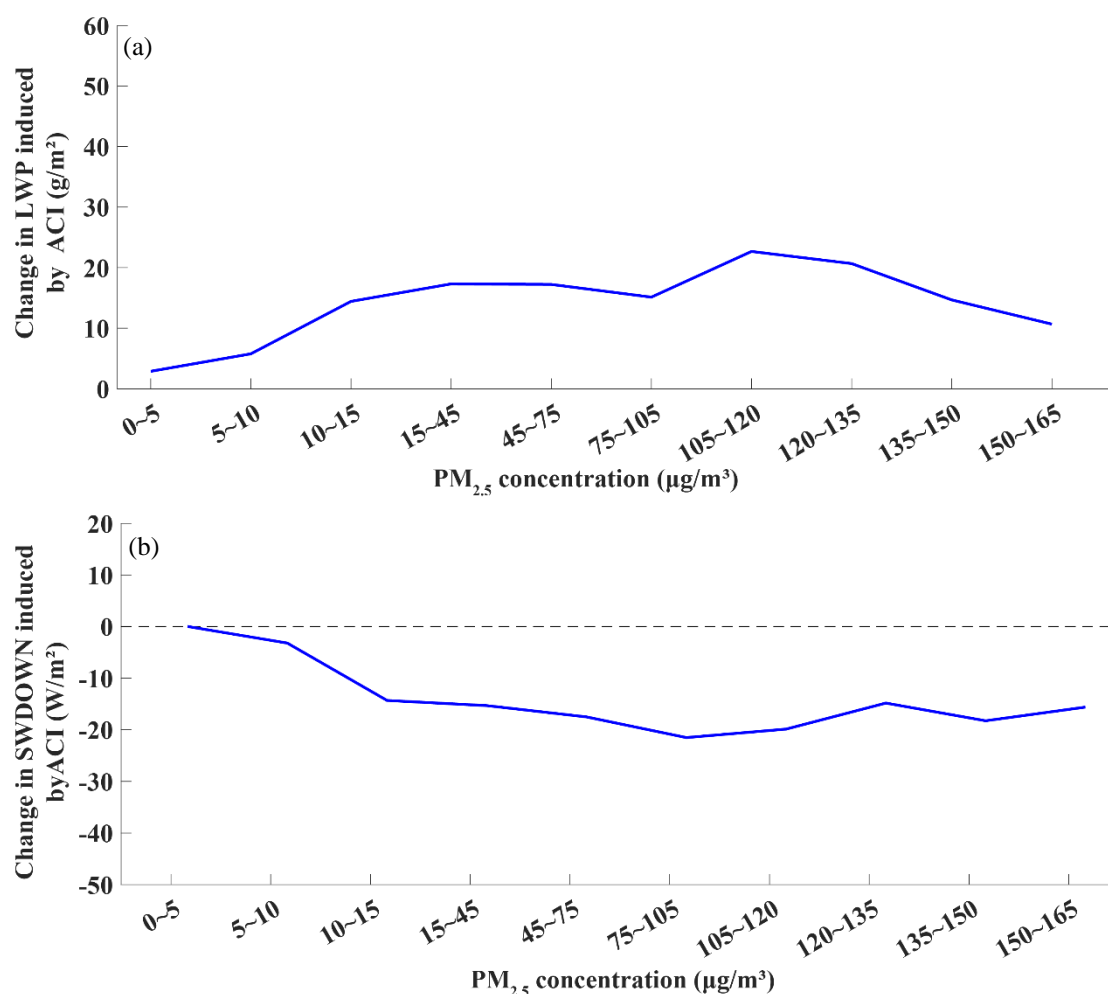


Fig. S8. Change in LWP and SWDOWN induced by ACI at different PM<sub>2.5</sub> levels.

4. Line 477-484, the explanation about the ARI- and ACI-induced PM<sub>2.5</sub> enhancements from the perspective of PM<sub>2.5</sub> concentration is weak, and no more new information was deduced here as illustrated in section 3.4.1. I would suggest to add more discussion on the meteorological conditions and PM<sub>2.5</sub> chemical compositions under different PM pollution levels, to explain the changes of ARI- and ACI-induced PM<sub>2.5</sub> enhancements.

Response: Thank you for your comment. We try to analyze the change in ARI- and ACI-induced PM<sub>2.5</sub> enhancement by discussing the change in ARI- and ACI-induced meteorological conditions and PM<sub>2.5</sub> chemical components. The changes in ARI- and ACI-induced PM<sub>2.5</sub> components between the different pollution levels could not be directly compared. As an example to demonstrate this process, if the PM<sub>2.5</sub> concentration in Shanghai and Lanzhou is 80 and 100 µg m<sup>-3</sup>, when we discuss the changes in ARI- and ACI-induced PM<sub>2.5</sub> components with PM<sub>2.5</sub> concentration decreasing from 100 to 80 µg m<sup>-3</sup>, we would probably find ARI- and ACI-induced OIN enhancement mainly decrease owing to large and small contribution of dust to

PM<sub>2.5</sub> concentration in Lanzhou and Shanghai. Therefore, we do not think that the ARI- and ACI-induced PM<sub>2.5</sub> components between the different PM<sub>2.5</sub> pollution levels are comparable.

We focus on the relationship between the change in ARI and ACI-induced meteorological conditions and change in ARI- and ACI-induced PM<sub>2.5</sub> enhancement. We find that, in the heavily PM<sub>2.5</sub>-polluted regime (135–180  $\mu\text{g m}^{-3}$ ), the decrease in SWDOWN induced by ARI is much larger than that induced by ACI (Fig. S9a). Then, the decrease in PBLH and T2 and the increase in RH induced by ARI are also larger than those induced by ACI (Fig. S9b–d). Thus, the enhanced PM<sub>2.5</sub> induced by the ARI is much larger than that by the ACI (Fig. 5a). However, when the PM<sub>2.5</sub> concentration decrease to the range of 15–45  $\mu\text{g m}^{-3}$ , the decrease in SWDOWN, PBLH, and T2 and the increase in RH induced by ACI significantly exceed those induced by ARI. Thus, the ACI-induced PM<sub>2.5</sub> enhancement significantly exceeds the ARI-induced PM<sub>2.5</sub> enhancement and becomes more important. This indicates the fast decrease in the ARI-induced PM<sub>2.5</sub> enhancement and the increasing contribution of the ACI-induced PM<sub>2.5</sub> enhancement with the decrease in the PM<sub>2.5</sub> concentration. In summary, the percentage decrease in the PM<sub>2.5</sub> enhancement induced by ACI is weaker than that induced by ARI with the decrease of PM<sub>2.5</sub> concentration because of the lower percentage decrease in the ACI-induced SWDOWN, which causes the lower percentage decrease in the ACI-induced PBLH and T2 reduction and the RH increase. Furthermore, as shown in Fig. S8a, the low percentage decrease in the ACI-induced SWDOWN reduction is due to a low decrease in the ACI-induced LWP in the PM<sub>2.5</sub>-polluted regime.

Our findings thus indicate that it is not an accident that the increasing importance of ACI-induced PM<sub>2.5</sub> enhancement occurs in China with emission reduction from 2013 to 2021, and it is a matter of course with ambient PM<sub>2.5</sub> decrease. The change in ARI- and ACI-induced meteorological factors is a major reason for the change in ARI- and ACI-induced PM<sub>2.5</sub> enhancement.

We have revised the original description. Please see lines 541-560 in the revised manuscript.

Lines 541-560: As shown in Fig. 5a, in the heavily PM<sub>2.5</sub>-polluted regime (135–180  $\mu\text{g m}^{-3}$ ), the decrease in SWDOWN induced by ARI is much larger than that induced by ACI (Fig. S9a). Then, the decrease in PBLH and T2 and the increase in RH induced by ARI are also larger than those induced by ACI (Fig. S9b–d). Thus, the enhanced PM<sub>2.5</sub> induced by the ARI is much larger than that by the ACI (Fig. 5a). However, when the PM<sub>2.5</sub> concentration decrease to the range of 15–45  $\mu\text{g m}^{-3}$ , the decrease in SWDOWN, PBLH, and T2 and the increase in RH induced by ACI significantly exceed those induced by ARI. Thus, the ACI-induced PM<sub>2.5</sub> enhancement significantly exceeds the ARI-induced PM<sub>2.5</sub> enhancement and becomes more important. This indicates the fast decrease in the ARI-induced PM<sub>2.5</sub> enhancement and the increasing contribution of the ACI-induced PM<sub>2.5</sub> enhancement

with the decrease in the  $PM_{2.5}$  concentration. In summary, the percentage decrease in the  $PM_{2.5}$  enhancement induced by ACI is weaker than that induced by ARI with the decrease of  $PM_{2.5}$  concentration because of the lower percentage decrease in the ACI-induced SWDOWN, which causes the lower percentage decrease in the ACI-induced PBLH and T2 reduction and the RH increase. Furthermore, as shown in Fig. S8a, the low percentage decrease in the ACI-induced SWDOWN reduction is due to a low decrease in the ACI-induced LWP in the  $PM_{2.5}$ -polluted regime.

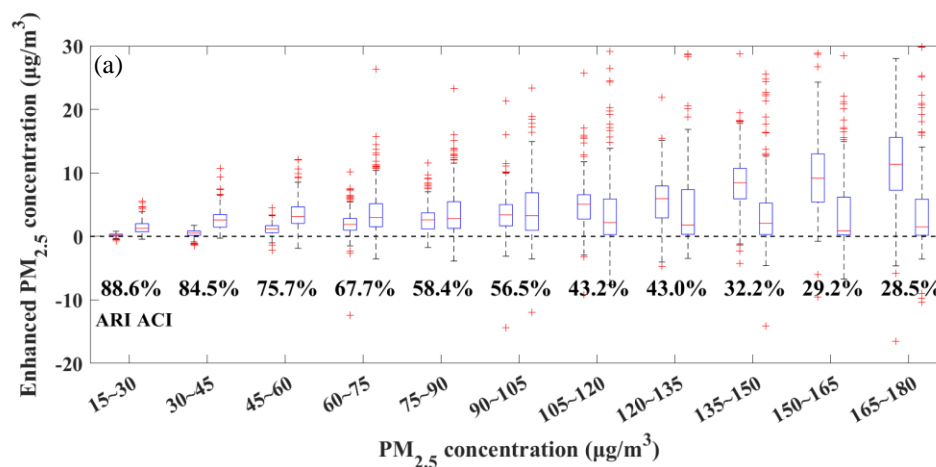
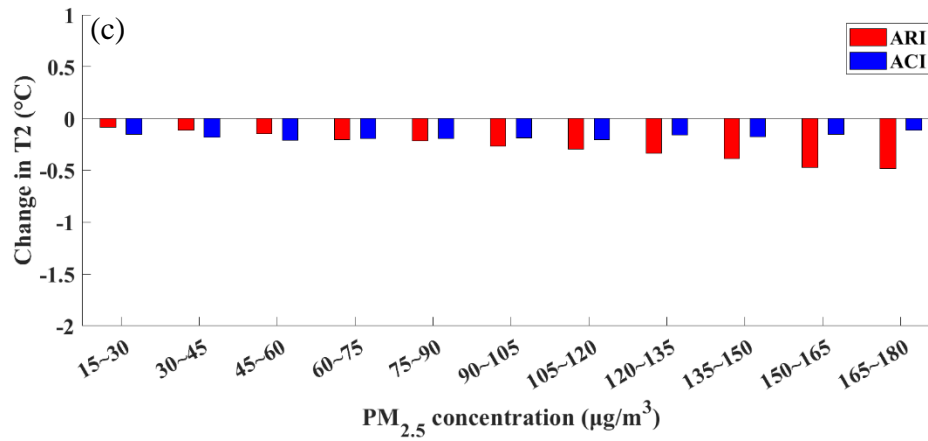
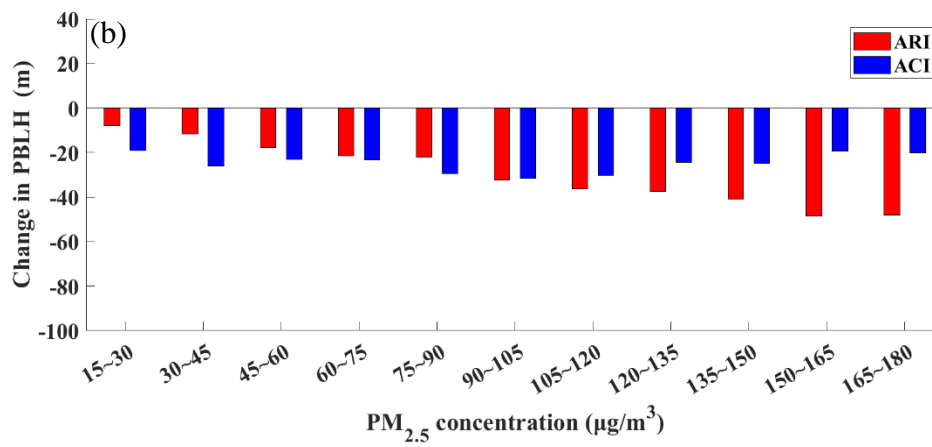
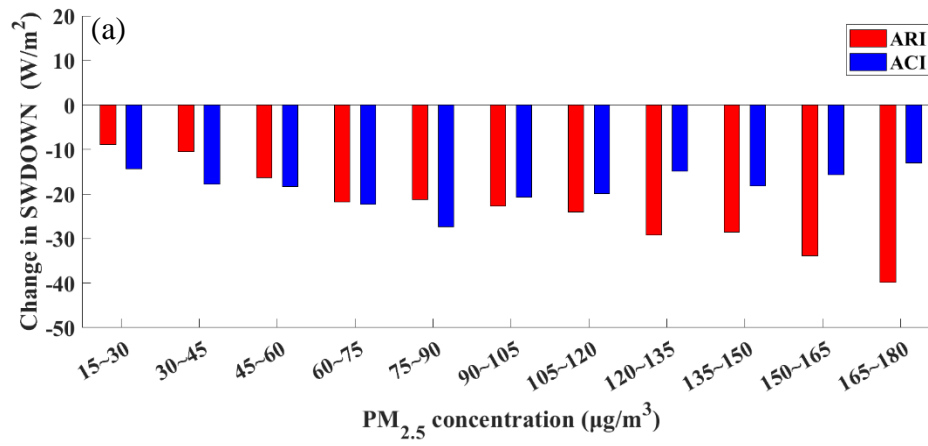


Fig. 5. (a) The enhanced  $PM_{2.5}$  concentrations induced by ARI and ACI at different ambient  $PM_{2.5}$  levels. These data are from the simulations for January and July in the experiments of 21M13E and 21M21E. The percentage represents the ratio of the ACI-induced  $PM_{2.5}$  enhancement to the sum of ARI- and ACI-induced  $PM_{2.5}$  enhancements.



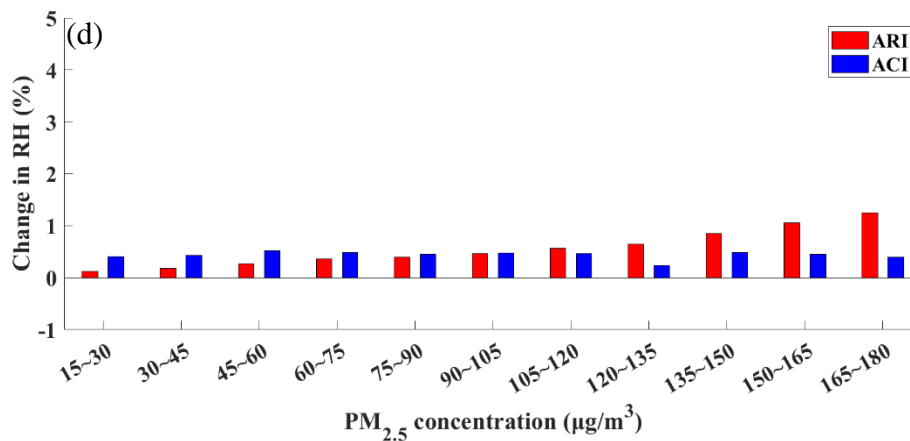


Fig. S9. Change in SWDOWN (a), PBLH (b), T2 (c), and RH (d) induced by ARI and ACI at different ambient PM<sub>2.5</sub> levels. These data are from the simulations for January and July in the experiments of 21M13E and 21M21E.

Minor revisions:

1. Line 109-114, about the conclusions cited from Moch et al. (2022) and Zhang et al. (2022), more details like which regions and which periods should be provided.

Response: Thank you for your comment. Detailed information has been added to the conclusions. Please see lines 111-117 in the revised manuscript.

Moch et al. (2022) found that the decrease in mean PM<sub>2.5</sub> concentration from the winter months of 2012–2013 to the winter months of 2016–2017 in China weakened the cloud–snowfall–albedo feedback induced by the aerosol semi-direct effect. For air quality, Zhang et al. (2022) found that the decrease in black carbon from 2013 to 2017 in China reduced the enhanced PM<sub>2.5</sub> concentration induced by the ARI by 1.8 µg m<sup>-3</sup> in January and 0.3 µg m<sup>-3</sup> in July.

2. Line 176-177, about the influence of prognostic aerosol in the model, more explanation is better to provided.

Response: Thank you for your comment. The prognostic aerosol can only be activated as CCN. It does not directly contribute to ice nucleation, which is only influenced by air temperature and supersaturation (Kanji et al., 2017). Furthermore, CCN would influence grid-scale clouds. However, limited by the horizontal resolution of 27 km × 27 km, cumulus clouds could not be resolved in this grid. Therefore, prognostic

aerosol does not influence ice nucleation and cumulus clouds in the model. This explanation has been added to the revised manuscript. Please see lines 186-191

3. Line 388-389, please define the inorganic aerosol (OIN) here, as sulfate and nitrate is also recognized as inorganic aerosol.

Response: Thank you for your comment. Other organic aerosol (OIN) refers to inorganic compositions other than sulfate, nitrate, ammonium, and BC. These compositions include sea salt and mineral elements. This has been corrected in the revised manuscript. Please see lines 444-446.

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