We acknowledge the referees for their insightful comments. We have made efforts to improve the manuscript accordingly. Please find our responses to the referees' comments in blue.

RC1 by Referee #3

Zhai et al. present a novel perspective on the impact of coarse particulate matter (PM) on the formation of fine PM nitrate in the North China Plain and the Seoul Metropolitan Area. The authors report that anthropogenic coarse PM, particularly fugitive dust from construction, which has not been considered in atmospheric chemical transport models, is capable of more efficiently absorbing gas-phase nitrate acid during the summer, resulting in a suppression of fine PM nitrate formation. Consequently, reducing emissions of coarse PM could lead to an unanticipated increase in fine PM nitrate levels, despite the decrease in NO_x and NH_3 emissions. The findings suggest the need for more extraordinary efforts to control NO_x and NH_3 emissions as fugitive dust pollution is controlled. The manuscript is well-organized, and the conclusions drawn are solid and well-supported by the data and simulation results. Therefore, I recommend the manuscript for acceptance with minimal revisions.

1. While the manuscript clearly outlines the relationships between coarse PM, fine PM, and gas-phase NO_x and NH_3, it would be helpful for readers if the authors could provide a diagram illustrating the tradeoffs associated with reducing emissions of coarse PM and NO_x/NH_3.

We added the following explanations for Figure 6 to illustrate the tradeoffs associated with reducing emissions of coarse PM and NO_x/NH_3.

Lines 44-46: "Model sensitivity simulations for 2015-2019 show that decreasing anthropogenic coarse PM directly increases $PM_{2.5}$ nitrate in summer, offsetting 80% the effect of nitrogen oxide and ammonia emission controls, …"

Lines 212-213: "The sum of changes driven by individual emission changes amounts to the total emission-driven net change."

Lines 225-226: "The decrease of coarse PM still quantitatively offsets the benefit from NO_x emission controls, which has been the main vehicle for controlling $PM_{2.5}$ nitrate."

Lines 233-234: "Over the NCP, the decrease of coarse PM offsets 80% of the benefits from NO_x and NH_3 emission controls."

Lines 255-256: "Decreasing coarse PM in the model in winter offsets the benefit of decreasing NO_x emissions, …"

Lines 258-259: "In summer, decreasing coarse PM in the NCP offsets 80% of the $PM_{2.5}$ nitrate benefit of decreasing NO_x and NH_3 emissions."

2. Line 160 suggests that the absorption of HNO_3 by coarse PM is three times more efficient than dry deposition, with alkalinity not being a limiting factor. However, it is not clear what the limiting factor is. Given that the available surface area typically restricts reactive uptake, the authors could provide clarification on how the surface area of coarse PM compares to that of fine PM, and which factor (e.g., surface area, chemical composition, relative humidity/temperature, etc.) limits the simulation.

We clarified lines 170-173: "We find that anthropogenic coarse PM takes up HNO_3 three times faster than dry deposition and that this uptake is limited by mass-transfer rather than alkalinity (only 60-70% of the coarse dust alkalinity in surface air is neutralized on average)."

RC2 by Referee #1

This paper presents an interesting analysis of the influence of coarse mode aerosol on the distribution of nitrate in the East Asian atmosphere. They use detailed aircraft and ground measurements during KORUS-AQ to show that a significant amount of nitrate appears to be associated with coarse mode particles and that including a more realistic representation of coarse mode particles reduces the GEOS-Chem model's high bias in fine mode nitrate. The analysis is also extended to simulations over the North China Plain. Overall the data and interpretation are clearly presented and I think the paper should be published following some clarifications, as requested below.

I found it difficult to follow the description of the approach for including coarse mode particles in the model on lines 142-147. Were the network observations applied as emissions or concentrations? How did the inclusion of the coarse mode in the lowest model level influence higher altitudes in the model domain? Was the linear interpolation in time, space?

We revised lines 145-152: "We compare the results from the standard model as described above to a simulation where we add anthropogenic coarse PM by using 24-hour average observed coarse PM concentrations from the air quality networks (Fig. 1) as boundary conditions at the lowest model level. For this purpose, we linearly interpolate the daily mean coarse PM data from the network to the GEOS-Chem model horizontal grid and apply them to the coarse dust GEOS-Chem model component with an effective diameter of 4.8 μm. This concentration boundary condition in the lowest model level serves as an implicit source and defines the vertical concentration profile. The resulting vertical profiles of coarse PM in GEOS-Chem over South Korea are consistent with KORUS-AQ aircraft observations (Fig. S3)."

We added a Figure S3.

The authors should clarify the definition of PM_{1-4} nitrate from the DC-8 observations. The AMS only measures submicron semi-volatile nitrate whereas the SAGA measures all PM_4 nitrate. It appears, though is not stated, that the difference between the SAGA and AMS nitrate measurements is used to define PM_{1-4} nitrate. What if there is refractory PM_1 nitrate? Figure S2 does not rule out this possibility because the AMS would likely not be sensitive to nitrate associated with refractory minerals in dust. What if there is semi-volatile supermicron nitrate? Would this impact the measurement model comparisons (it's unclear from line 151 if the model nitrate is PM_1 or any ammonium nitrate)? These details are unlikely to impact the qualitative outcomes of the analysis, but it would be useful to have more precision in the definitions.

We added lines 117-118: "The AMS only detects non-refractory nitrate, taken here to be ammonium nitrate (Fig. S2)."

Lines 157-158: "PM_{1-4} nitrate is derived as the difference between SAGA PM_4 nitrate and AMS PM_1 nitrate."

Lines 159-162: "In this way, any dust-associated refractory PM_1 nitrate is included in the PM_{1-4} profiles, for both observations and the GEOS-Chem model. Such classification does not allow for supermicron ammonium nitrate, but KORUS-AQ observations found ammonium nitrate to be mainly submicron (Kim et al., 2018)."

We revised the caption of Figure 4 in lines 543-547: "(a) Median diurnal variation (error bars are 25th and 75th percentiles) of non-refractory PM_1 nitrate (taken to be ammonium nitrate) at the Korea Institute of Science and Technology (KIST) site. (b)-(e) Median vertical profiles of non-refractory PM_1 nitrate, PM_{1-4} nitrate, coarse PM ($PM_{2.5-10}$), and HNO_3 concentrations for the ensemble of flights over the SMA. Horizontal bars for the observations indicate 25^{th}-75^{th} percentiles."

Related to the points above, the authors note that the impact of including coarse mode aerosol that can uptake HNO_3 is less impactful in the NCP, but in this example, they compare $PM_{2.5}$ nitrate rather than PM_1 nitrate. Very little is said about the observations from China, but are these measurements limited to semi-volatile nitrate, or would they also include dust nitrate in the fine mode?

We added lines 181-182: "$PM_{2.5}$ nitrate observations in NCP are mostly filter-collected bulk $PM_{2.5}$ nitrate, which could be biased low in summer due to volatilization (Chow et al., 2005)."

At line 188, "modeled $PM_{2.5}$ nitrate" is changed to "modeled ammonium nitrate".

Lines 189-191: "The comparison with $PM_{2.5}$ nitrate observations here indicates that fine dust associated nitrate should be considered when comparing modeled particle nitrate to bulk $PM_{2.5}$ nitrate data."

Reference:

Chow, J. C., Watson, J. G., Lowenthal, D. H., and Magliano, K. L.: Loss of $PM_{2.5}$ nitrate from filter samples in central California, J. Air Waste Manag. Assoc., 55, 1158-1168, 2005.

In the analysis of the overall sensitivity of fine mode nitrate to recent emissions trends, the authors explain the more modest sensitivity of nitrate to dust by the abundance of NH_3 and kinetic arguments about mass transfer (lines 207-212). I find this surprising as I would assume that in the model the ammonium nitrate remains semi-volatile, whereas the coarse nitrate formation is irreversible. In that case, it doesn't matter how quickly the ammonium nitrate forms, given sufficient time the presence of a reactive coarse mode would still siphon off fine mode nitrate, unless there are other competing sinks?

We added lines 129-130: "Formation of semi-volatile ammonium nitrate aerosol is governed by ISORROPIA version 2.2 thermodynamics (Fountoukis and Nenes, 2007)."

Lines 221-225: "Decreasing coarse PM has relatively little direct effect on $PM_{2.5}$ nitrate in winter in the NCP because abundant atmospheric NH_3 combined with low temperatures drives HNO_3 near-quantitively to ammonium-nitrate particles, and subsequent mass transfer of HNO_3 from ammonium nitrate to coarse PM is very slow because of the weak HNO_3 partial pressure (Wexler and Seinfeld, 1992)."

Lines 226-230: "Consideration of coarse PM in the model further increases the sensitivity of $PM_{2.5}$ nitrate to NH_3 and SO_2 emissions respectively by 30% and 46%. This is because coarse PM provides an additional sink for the small fraction of HNO_3 that remains in the gas phase, which increases the sensitivity of the atmospheric lifetime of total nitrate (ammonium nitrate + HNO_3) to changes in NH_3 or SO_2 emissions (Zhai et al., 2021a)."

Reference

Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+–Ca^{2+}–Mg^{2+}–NH_4^+–Na^+–SO_4^{2-}–NO_3^-–Cl^-–H_2O aerosols, Atmos. Chem. Phys., 7, 4639-4659, https://doi.org/10.5194/acp-7-4639-2007, 2007.

Wexler, A. S. and Seinfeld, J. H.: Analysis of aerosol ammonium nitrate: Departures from equilibrium during SCAQS, Atmos. Environ. Part A. General Topics, 26, 579-591, https://doi.org/10.1016/0960-1686(92)90171-G, 1992.