We thank both anonymous reviewers for their careful read of the revised manuscript. Our responses are below in blue with revisions in bold.

Reviewer #1

(1) Missing heterogenous chemistry of SO2 is listed as the main reason for model underestimate of sulfate. But the model result of adding SO2 heterogenous chemistry is only briefly mentioned and the figure is in the supplementary section S17. I suggest incorporating it in the main part.

Thank you for this suggestion. We agree this would be helpful. We have moved Figure S17 to the main text as Figure 13. We added the following to line 522 to further describe the impact of this figure in the main text.

"The derived values for γ described above may need to be revised in future work to consider the impacts of errors in the nitrate simulation (discussed below) as well as errors in SO_2 emissions."

(2) In the abstract, the shallow nighttime PBL height, or rapid collapse of mixing layer, is listed as one of main reasons for model bias in nighttime chemistry. But based on Figure 9, the run with rising PBL (to the observation level) shows no improvement in model performance, i.e. the red line and blue line are almost the same for most species. Please clarify.

Restate abstract

As the model is run with assimilated meteorology, artificially raising the PBL is not coupled to a physical cause (i.e. increased heat fluxes), hence the effect of changing the height is limited. We explain this with respect to ozone on line 448 and add the word "significant" to add clarity:

"We illustrate in Section S6 that reducing the collapse of the evening MLH without a **significant** change to the drivers of mixing (i.e., heat fluxes, friction velocity) also has negligible impact on decreasing model ozone titration (Fig. S12b)."

On line 481, we discuss the need to address the drivers of mixing (i.e., not just the outcome of an increased PBL height) and add a reference to Table 4 to increase clarity:

"We drive additional nocturnal mixing **(Table 4, increased nighttime mixing)** by increasing the sensible heat flux at night from slightly negative (-4 W m^{-2}) to weakly positive (+10 W m^{-2}), representative of anthropogenic heat fluxes in this region (Hong and Hong, 2016; Varquez et al., 2021)."

Line 482 explains the result of this sensitivity test:

"This sensitivity test (Table 4) largely resolves the incorrect model ozone titration and the severe model overestimate of nighttime NO_2 on 5/23-5/24 and on 5/24-5/25…"

(3) Figure 6, where is 5x dry deposition run results in (b), (c), and (d)? No impact on other species? NO2 deposition is under-estimated? Increasing or decreasing dry deposition look more like tuning model toward observations.

The model runs are plotted underneath the base model run, thus they are difficult to see. We refer the reviewer to this statement in our caption of Figure 6: Model sensitivity simulations that are not significantly different than the base model run are plotted underneath the base model line.

We have tuned HNO3 dry deposition towards observations but are clear that our analysis shows that this tuning is suggestive of the need for stronger loss, and here we implement that using dry deposition. See line 567:

"The model overestimate in nitric acid was not due to overestimated production, insufficient loss to wet deposition, or uptake to dust or seasalt. Increasing the loss of nitric acid, implemented here as an increase in the nitric acid dry deposition velocity by a factor of five, was required to reconcile the model with observations."

- One more suggestion: the "No local emissions" run can be a very useful model simulation to quantify the relative contribution from the local emissions vs long range transport. It is not showed in most analyses (figures), right?

We agree that this sensitivity test is essential to show the relative contribution of local emissions vs. long-range transport. This sensitivity test is used for this purpose in Fig. 13 to calculate the foreign contribution to $PM_{2.5}$. This may not have been clear, so we added the following reference to Table 4 to line 528:

We simulate $PM_{2.5}$ with heterogeneous conversion of SO_2 as described above, and then remove South Korean emissions **(Table 4),** in order to investigate changes to the fraction of transported pollution.

We also realized that a sensitivity test was missing from Table 4 where we remove local emissions with the addition of heterogeneous SO_2 chemistry. This has been added.

Table 4. Description of model experiments

Name	**Resolution**	**Simulation Length**	**Description of changes**
Base model	0.25° × 0.3125° over East Asia. Boundary conditions (BCs) from a global 2° × 2.5° simulation[1.]	1 month initialization + KORUS-AQ period (May 1-June 9).	N/A
No nighttime production	0.25° × 0.3125° over East Asia.	KORUS-AQ period	Remove reactions R2-R5.
Old wet scavenging scheme	0.25° × 0.3125° over East Asia.	KORUS-AQ period	Remove recently implemented wet scavenging scheme (Luo et al., 2019).
5x dry deposition	0.25° × 0.3125° over East Asia.	KORUS-AQ period	Increase the deposition velocity of HNO_3 by a factor of 5.
No local emissions	0.25° × 0.3125° over East Asia.	KORUS-AQ period	Turn off anthropogenic emissions over South Korea.
Raise nighttime PBL	0.25° × 0.3125° over East Asia.	KORUS-AQ period	Increase the nighttime MLH to 500m.
Increased nighttime mixing	0.25° × 0.3125° over East Asia.	May 23 to May 31	Increase the nighttime MLH to 300m and set nighttime sensible heat flux to 10 W m^{-2}.
Het SO_2	0.25° × 0.3125° over East Asia.	KORUS-AQ period	Uptake of SO_2 on aerosol with $\gamma_{RH_{100\%}} = 3 \times 10^{-4}$ and $\gamma_{RH_{50\%}} = 3 \times 10^{-5}$.
Het SO_2 with no local emissions	**0.25° × 0.3125° over East Asia.**	**KORUS-AQ period**	**Uptake of SO_2 on aerosol with $\gamma_{RH_{100\%}} = 3 \times 10^{-4}$ and $\gamma_{RH_{50\%}} = 3 \times 10^{-5}$ and turn off anthropogenic emissions over South Korea.**

[1]Boundary conditions from the base simulation are applied to all sensitivity simulations.

- The line number in the draft is only partially labelled and mismatched to the number mentioned in the response to reviewer. We have to use search function to locate the content mentioned in 'ResponsetoReviewers'.

We apologize for this confusion, we assume this was some error in the upload.

Reviewer #2

After reading the response letter and the revised manuscript, I think the authors have adequately addressed both reviewers' comments, and suggest publish on ACP.

Minor typo: Table 4, Base model resolution, '2 x 2.25' should be '2 x 2.5'

This has been fixed.