Replies to the comments from the anonymous Referee #1

We greatly appreciate the Editor-in-Chief (Eleanor C. Browne) and Referee #1 for providing highly insightful and constructive comments, which have substantially improved the clarity of our manuscript. Please see below our point-to-point responses in blue (our comments) and red text (revisions) and refer to the revised manuscript.

Comment#1: Overall, the manuscript is significantly improved. My only major comment is that the conclusion section is hard to follow – a higher-level summary that succinctly makes the key points that are described in the main text would be more effective.

We are very thankful to the Referees for the time and effort that you have put into reviewing the previous version of the manuscript. We ensure that each comment has been addressed carefully and that the paper is revised accordingly.

Comment#2: I suggest:

Cutting the sentences from 478-483 (The model predicted average…to…not incorporated into the model"). Then cutting the end of the paragraph beginning at Line 490.

Response: Deleted in the revised manuscript.

At the beginning of the next paragraph remove "further" (line 494).

Response: Removed in the revised manuscript.

Then:

The revised model shows that by adding HCl emissions more NHx was partitioned to the condensed phase improving agreement with the observations. 3×Base HCl was able to represent well the diurnal variation of NH4+ and Cl- both in terms of amount and pattern with improved NMB for NH3. Additional sensitivities tests in changing NH3 emissions (reduction by a factor of 3) in the 3xBase HCl also improved the NH3, NH4, and NHx concentrations. [key last sentence beginning on 505]. These results high light the need to include correct industrial sources of HCl emissions along with appropriate emissions of NH3 to reduce biases in NHx.

Use current text:[Developing the appropriate NH3 emissions using country-specific emission inventories, which are currently under development as part of the Global Challenges Research Fund (GCRF), South Asian Nitrogen Hub (SANH). Also, there is potential to develop top-down constraints on NH3 emissions by taking inference from the satellite,

model, and ground-based observations.] Challenges remain in simulating NH3 as a contributor to particulate matter due to temporal factors in ammonia peaks including the role of fog and dew where more work is needed. This work also suggests model improvements to SO2 oxidation pathways could improve NHx partitioning.

Response: Conclusion section is corrected in the revised manuscript as follows:-

Conclusion:

"In this study, we have evaluated for the first time in South Asia the performance of a chemical transport model (WRF-Chem) in modeling NH_3, NH_4^+, and total NH_x, by comparing against the WiFEX measurements (MARGA). In daily means, we find NH_3 is significantly overestimated by the model, NH_4^+ was underestimated while simulated total NH_x agreed well with the measurement, indicating incorrect gas-to-particle partitioning along with missing chemical process may impact this mismatch in the model. The ability of the model to accurately describe the gas-to-particle partitioning of the MARGA was evaluated by the fraction of total NH_x (= NH_3 + NH_4^+) in the particulate phase (NH_4^+/NH_x). A strong relation of MARGA NH_4^+/NH_x was observed with dominant anion (Cl^-) (r = 0.79), whereas the standard model showed a strong correlation between NH_4^+/NH_x with dominant anion (SO_4^{2-}) (r = 0.77), pointing to the missing chloride (HCl/Cl^-) chemistry in the model.

We incorporated HCl/Cl^- emissions in the model and conducted three sensitivity experiments of varying HCl emissions, named as No HCl (0 mol km^{-2} h^{-1}), Base Case HCl (3× Sharma et al., 2019; 24.8 mol km^{-2} h^{-1}) and 3×Base HCl (74 mol km^{-2} h^{-1}) run. The revised model shows that by adding HCl emissions more NH_x was partitioned to the condensed phase improving agreement with the observations. 3×Base HCl was able to represent well the diurnal variation of NH_4^+ and Cl^- both in terms of amount and pattern with improved NMB for NH_3. Additional sensitivities tests in changing NH_3 emissions (reduction by a factor of 3) in the 3×Base HCl also improved NH_3, NH_4^+, and NH_x concentrations. We find excess NH_3 along with longer lifetime of NH_4^+ may act as a controlling driver for NH_x overestimation in the model. These results highlight the need to include correct industrial sources of HCl emissions along with appropriate emissions of NH_3 to reduce biases in NH_x. Developing the appropriate NH_3 emissions using country-specific emission inventories, which are currently under development as part of the Global Challenges Research Fund (GCRF), South Asian Nitrogen Hub (SANH). Also, there is potential to develop top-down constraints on NH_3 emissions by taking inference from the satellite, model, and ground-based observations. Challenges remain in simulating NH_3 as a contributor to particulate matter due to temporal factors in ammonia peaks including the role of fog and dew where more work is needed. This work also suggests model improvements to SO_2 oxidation pathways could improve NH_x partitioning.

Comment#3: Minor comments:

Line 56: remove analyses

Response: Removed in the revised manuscript.

Line 75: remove “to its alkaline nature” – this would be an issue globally; could replace with diverse (sources)

Response: Removed in the revised manuscript.

Line 143: add “and” between anion and cation instead of the comma

Response: Added “and” in the revised manuscript.

Line 149: change to past tense; may have stuck on…

Response: Modified in the revised manuscript.

Line 247: remove the before NH3

Response: Removed in the revised manuscript.

Line 420: decreases

Response: Added in the revised manuscript.

Response to the Editor's comments

Comment#1: Comments to the author:

Dear Authors:

Thank you for your careful consideration of the referee comments and your extensive revisions in response to those comments. I think that the paper is improved - the main conclusions are easier to follow and the reasoning is presented in a reasonable layout. Given the magnitude of the changes, I sent this to one of the referees again and they agree with my overall conclusions. I am happy to accept this for publication following the attention to a few suggestions and technical corrections.

Dear Eleanor C. Browne,

Thank you very much for the supportive feedback and consideration of our paper for final publication in ACP, subject to minor comments. Our point-to-point replies to the suggestions and technical corrections are listed below:

Comment#2: Suggestions:

1) Please see the referee report for suggestions on modifying the conclusions to shorten the conclusions section and increase the clarity of the major findings of the work.

Response: Corrected in the revised manuscript and refer to author response to RC-*Comment#2*.

2) I found the results of the reducing NH3 model intriguing. It would be interesting to know if the diel profile of the NH3 in the model also more closely matched the observations as opposed to just the total amount. Given how the diel profile carries some information about processes controlling NH3 and NH4+ concentrations (which the manuscript nicely discusses earlier), I think this information would be of interest to the reader and is important information for future studies to consider. I think this could be accomplished in a few sentences - there is no need to add an extensive discussion.

Response: Added in the revised manuscript.

"In order to better understand the relationship between NH_3, NH_4^+ and NH_x concentrations in the diurnal profile of model, one sensitivity study is conducted in the best case HCl experiment to simulate the response of NH_x concentrations by changing NH_3 emissions. In these simulations, only NH_3 emissions were reduced further by a factor of 3 (-3×NH_3_EMI) in the 3×Base HCl experiment, while all other processes and chemical schemes were unchanged. Figure S6 in the Supplement shows the diel profile of model/obs ratio for NH_3

(Fig. S6a), NH_4^+ (Fig. S6b), and total NH_x (Fig. S6c) concentration simulated with the 3×Base HCl and -3×NH_3_EMI scenario. Reducing NH_3 emissions in the model (-3×NH_3_EMI) significantly improves model-measurement agreement for NH_3 (mean model/obs = 1.9), NH_4^+ (mean model/obs = 0.9), and total NH_x concentration (mean model/obs = 1.2) compared to the 3×Base HCl run, further suggesting that the longer lifetime of NH_4^+ may be the controlling driver for the total NH_x concentration in the model."

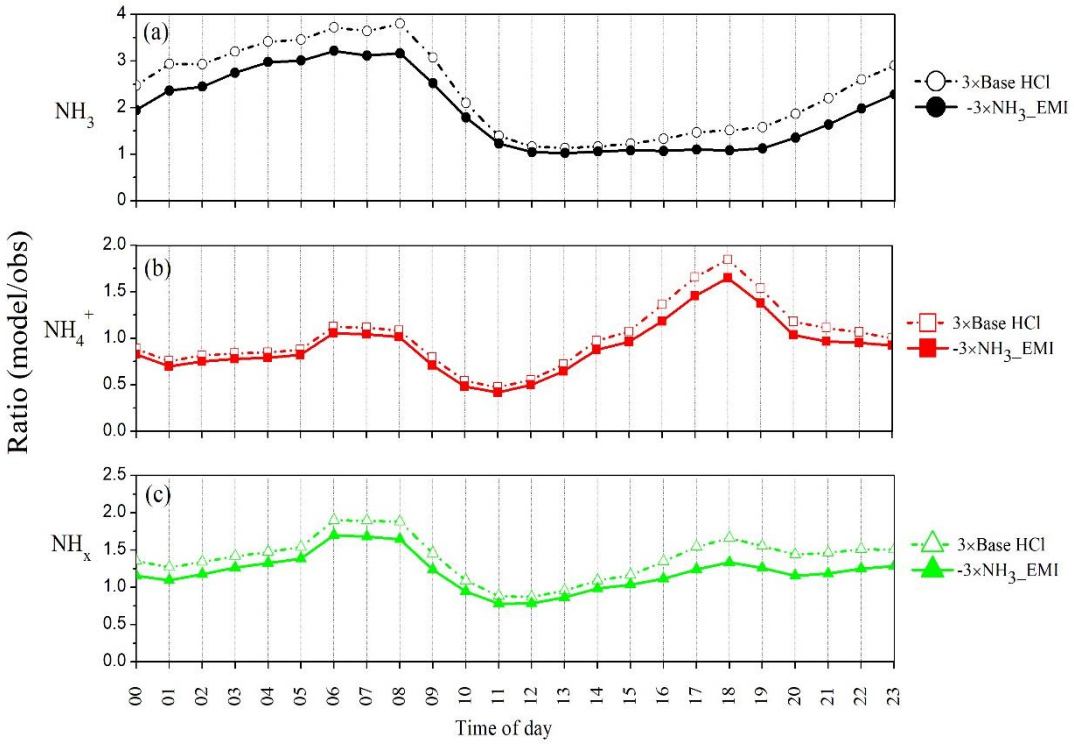

Figure S6. Comparison of diel profile of model/obs ratio for the mean (a) NH_3 concentration (b) NH_4^+ concentration, and (c) total NH_x concentration in 3×Base HCl and -3×NH_3_EMI scenario.

Comment#3: Technical

See technical corrections in Referee's report as well

Response: Yes, corrected in the revised manuscript.

Line 141: Suggest changing "all the precautions" to "several precautions" or "followed best practices"

Response: Yes, corrected in the revised manuscript.

"we have followed best practices during the study"

Line 190: Is organic carbon or total organic mass being discussed? It is unclear from the phrasing.

Response: Since organic carbon or total organic mass is not discussed, the statement is corrected as follows:

"This study used the Model for Ozone And Related chemical Tracers (MOZART-4) gas-phase chemical mechanism coupled with the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) aerosol scheme, that simulates SO_4^{2-}, NH_4^+, NO_3^-, methanesulfonate, Na^+, Ca^{2+}, Cl^-, carbonate, black carbon ~~(BC)~~, and primary organic mass ~~(OC)~~."

Line 264: "ratio between observed and simulated" --> "ratio between simulated and observed" so that the wording matches the equation

Response: Thanks for the suggestion; corrected in the revised manuscript.

"To assess the validity of the model, the ratio between simulated and observed (model/obs) was tested."

Line 296: "aqueous phase oxidation of" --> "aqueous phase oxidation by"

Response: Modified in the revised manuscript.

Line 297: In the supporting information version I have, Figure S3 shows ozone and not anything directly to do with SO2/SO4. The reference to the figure is thus unclear.

Response: We agree with your comment; hence for clarity referencing the figure is not required, and hence we deleted it in the revised manuscript.

"In a normally NH_3-rich atmosphere, gas-phase oxidation of SO_2 is much slower than the aqueous phase oxidation of O_3, and due to nearby sources, much of the sulfur is present as SO_2 (Li et al., 2007) ~~(Fig. S3 in the Supplement)~~."

Lines 320-321: Comparing the wording and the figure are confusing because the wording addresses particle phase Cl- increasing at high RH whereas the figure shows HCl/Cl- (the inverse of the wording) and thus it decreases. I suggest harmonizing the wording and the figure.

Response: Corrected in the revised manuscript.

"We plotted the fraction of the ratio of HCl to Cl^- (HCl/Cl^-) as a function of NH_4^+ concentration and RH in Fig. 4. The decrease in the fraction of HCl/Cl^- is associated with an increase in NH_4^+ concentration at high RH between 70-100 %."