Response to the comment from Anonymous referee #1

[1] Reviewer response: Thank you for addressing this. First, do you mean "chloride depletion of SSA through…" as opposed to 'chloride deposition of SSA through…"

I see that your model results show the production of Cl radicals from ClNO and $ClNO_2$ to be more important than production from reactions involving H_2SO_4, HNO_3, and SSA. If I understand correctly, reactions forming ClNO and $ClNO_2$ would result in a replacement of particulate Cl^- with NO_3^- in sea salt particles. Thus if pathways involving ClNO and $ClNO_2$ are the dominant way in which reactive Cl in SSA is being released to the atmosphere, would you expect the composition of depleted SSA to contain mostly NO_3^- with relatively small amounts of SO_4^{2-}? I ask because studies looking at the composition of aged SSA such as Braun et al. (2017; https://doi.org/10.1021/acs.est.7b02039) and AzadiAghdam et al. (2019; https://doi.org/10.1016/j.atmosenv.2019.116922) show that a considerable amount of chloride depletion in submicron SSA can be attributed to SO_4^{2-} in certain conditions. Braun et al. is considering a different region, while AzadiAghdam et al. features data from the Philippines. I have seen other studies as well attributing non-negligible amounts of chloride depletion to reactions between H_2SO_4 and SSA when analyzing the composition of aged sea salt particles. Your study is looking at it from a different perspective (where you are NOT studying chloride depletion from the perspective of the chemical composition of the sea salt particles themselves), so I wonder how to resolve the differences between the perspectives when it comes to the role played by HNO_3 and H_2SO_4. I certainly am not casting doubt on your model results, and I see they show changes in SO_2 are small and that HCl is not a dominant contributor to Cl radicals.

I think researchers studying chloride depletion from the perspective of SSA chemical composition will find your results interesting regarding the chemical pathways leading to the presence of HNO_3 in aged sea salt particles. I think currently many researchers see HNO_3 in aged SSA and report that the HNO_3 came to exist in the SSA through the pathway $HNO_3 + NaCl \rightarrow HCl + NaNO_3$, whereas you show that other pathways involving N_2O_5 and NO_2 may lead more dominantly to the presence of $NaNO_3$ in SSA (if I understand your work correctly, please correct me if I'm misunderstanding). For example, here is a quote from Su et al. (2022; https://doi.org/10.1016/j.atmosenv.2022.119365): "The acid displacement reaction is regarded as a major chemical pathway in chloride depletion, releasing HCl:HA (g or aq) + NaCl (aq or s) → NaA (aq or s) + HCl (g or aq)where HA (g or aq) represents acidic species (e.g., sulfuric acid,nitric acid, organic acid) in gaseous (g) or aqueous (aq) phases. NaCl (aq or s) is the major component of SSA in aqueous or solid (s) phases. NaA is the sodium salt from the interaction between HA and NaCl. It should be noted that the acid displacement reaction involving organic acid in SSA is reversible."

I apologize for the long-winded comment. I just find it interesting that from the modeling perspective Cl radical formation from HCl is small compared to that from ClNO and $ClNO_2$, whereas in sea salt chemistry papers, the main reaction presented in describing the process of Cl-depletion is HA + NaCl → NA + HCl.

This paper showcases how it is important to consider (1) modeling studies looking at the most important pathways regarding the production of Cl radicals and (2) studies examining chloride depletion from the perspective of SSA chemical composition as the two perspectives may be inclined to emphasize different reaction pathways in explaining the presence of HNO_3 in aged SSA. No action needed for this comment, just wanted to put these thoughts out there and see if you had any thoughts on this. I am more familiar with the particle chemistry side, so it would be interesting to hear your perspective from the modeling side.

Response: Firstly, thanks for pointing out this typo. We have replaced "chloride deposition" with "chloride depletion" in the manuscript.

Secondly, we sincerely appreciate this insightful comment, which illuminates a fascinating methodological duality in interpreting sea salt chloride depletion mechanisms. Below, we integrate your observations with our model results to reconcile these viewpoints:

(1) The relative importance of sulfate vs. nitrate in chloride depletion can be regionally heterogeneous. In high-SO_2 regions, sulfate accumulation via $H_2SO_4 + 2NaCl \rightarrow Na_2SO_4 + 2HCl$ may dominate chloride loss. In high-NO_x regions, nitrate pathways (N_2O_5 hydrolysis: $N_2O_5 + NaCl \rightarrow NaNO_3 + ClNO_2$; or $HNO_3 + NaCl \rightarrow NaNO_3 + HCl$) prevail, explaining the prominence of particulate NO_3^- over SO_4^{2-}. This spatial variability reconciles the seemingly contradictory literature – both pathways operate, but their relative weights depend on local precursor emissions.

(2) The small contribution of HCl to Cl radicals in our model may not imply low HCl production from acid displacement reactions (HA + NaCl → NaA + HCl). Rather, it reflects that the $HCl + OH \rightarrow Cl + H_2O$ reaction is slow, especially compared to rapid $ClNO/ClNO_2$ photolysis. Thus, even

sufficient HCl production may not yield significant Cl radicals.

While quantifying sulfate/nitrate partitioning in depleted SSA was not this study's primary focus, we agree this represents a compelling research frontier. We will conduct targeted sensitivity experiments in future work (e.g., zero-out H_2SO_4/HNO_3 emissions) to isolate their individual impacts on chloride depletion products.

Revision in the manuscript:

(1) Line 144: “The chlorine depletion of SSA through its equilibrium reactions with H_2SO_4 and HNO_3 were considered in the model”

[2] Reviewer response: Thank you for responding to this comment and for the Molnar and Meszaros citation. I looked at this paper, and it does not specifically mention that fine SSA particles have a higher extinction effect compared to coarse SSA particles. The work you are referencing sampled particles from 10 m on the Great Hungarian Plain, which appears to be inland and far from marine sources. To me it seems unlikely they sampled considerable mass concentrations of sea salt particles, and when searching for the words ‘sea salt’ and ‘marine’ in the paper, nothing comes up. Thus, I recommend you revise your citation of Molnar and Meszaros (2001) when stating fine SSA have a higher extinction effect than coarse SSA. Perhaps you can find another work explicitly mentioning SSA in this context. Alternatively, Molnar and Meszaros (2001) do support your statement that fine particles are more dominant in governing extinction, so perhaps you can write this without explicitly mentioning SSA in the statement.

Response: Thanks for your valuable comment. We agreed that Molnár and Mészáros (2001) did support the statement that fine particles are more dominant in governing extinction. To avoid misleading, in the revised manuscript, we stated this without explicitly mentioning SSA.

Revision in the manuscript:

(1) Line 307: “Fine particles have a higher extinction effect than coarse ones (Molnár and Mészáros, 2001), and they can be transported to higher levels”

Additional reviewer comments:

[3] On line 153 you now state ‘This parameterization was identical across different aerosol modes (Aitken, accumulation, and coarse).’ and on line 192, you now mention the composition of dry SSA in different modes remains consistent. Would you mind adding the exact size range of SSA considered in your model (e.g., XX – XX nm)?

Response: Thanks for your valuable comment. According to the codes in the CMAQ model, the exact size range of SSA is from ~0.02 to 20 μm. We have added this information in the manuscript.

Revision in the manuscript:

(1) Line 180: “The estimated diameters of SSA range from ~0.02 μm to 20 μm in the model.”

[4] There are several instances of issues with subject-verb agreement still in the manuscript. Hopefully additional editing stages will handle these issues.

Response: We sincerely appreciate the reviewer's careful attention to grammatical details. All instances of subject-verb agreement have been thoroughly reviewed and corrected in the revised manuscript.