

We would like to thank both reviewers for their positive feedback and valuable comments. We have revised the manuscript accordingly. Please find our responses to reviewer comments below in blue and the revised text in red. The reviewers' comments are in black. The revised manuscript with and without the changes highlighted is also provided. First, we summarise the main revisions to the manuscript. At the end of each reviewer's reply we provide a list with the references we added in the revised manuscript.

Based on the major comment from Rev. 1, we have revised the text on the model evaluation of Arctic SO_4^{2-} including sub-micron nss- SO_4^{2-} and discuss our findings within the context of other studies. We also replied to all Rev. 1's minor comments.

To address the comments and concerns made by Rev. 2, the paper has been re-organised and shortened. In particular, we revised the Title and re-wrote the Abstract, Introduction, the section on evaluation of the 100km results (old Section 4, now Section 5) and the Conclusions. The SSA emission scheme used in the base version of the model, together with the updates to this scheme, are now described in a separate Section 4.

We also would like to inform the reviewers that we found an error in the model simulations. Due to the complexities of the WRF-Chem model, unfortunately, the flag activating dinitrogen pentoxide (N_2O_5) hydrolysis was switched off. We have rerun all the simulations with N_2O_5 hydrolysis on, and the corrected results are included in the revised manuscript. Since N_2O_5 hydrolysis is an important source of nitric acid (HNO_3) at night-time, we assessed the impact on our results. Whilst changes in HNO_3 , and thus on NO_3^- aerosols, can be important over NO_x emission regions, we found a rather small influence on modelled NO_3^- in the Arctic (maximum changes of up to $0.1 \mu\text{m}^{-3}$ for super-micron and sub-micron NO_3^-). It appears that since N_2O_5 hydrolysis is faster where water vapour (humidity) is higher, the effects are larger over more southerly mid-latitude emission regions. Since humidity over the Arctic in winter is low, the results are affected to a lesser extent over this region. We include figures to illustrate this below. Figure 1 shows the absolute changes for sub-micron NO_3^- during February 2014 over the Arctic region. Note that the only difference between the two simulations for HEM_NEW is N_2O_5 hydrolysis. The rest of the set-up is the same as described in the submitted manuscript. Figure 2 illustrates the differences in modelled inorganic aerosols between model runs (HEM_NEW) with and without N_2O_5 hydrolysis compared to observations at Zeppelin. Only small differences are found at this and other Arctic sites.

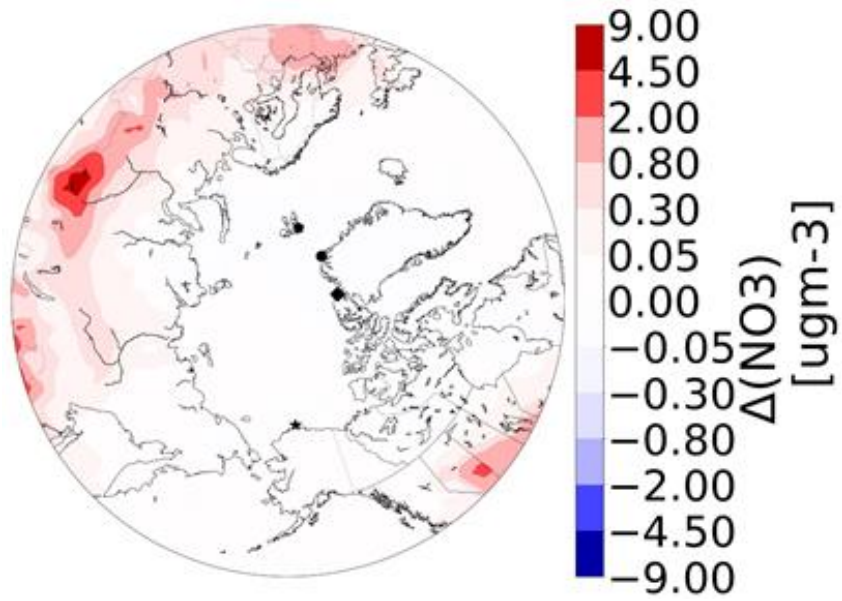


Figure 1: Average absolute differences in sub-micron NO_3^- concentrations (in μgm^{-3}) between HEM_NEW (with N_2O_5 hydrolysis) and HEM_NEW (without N_2O_5 hydrolysis) runs during February 2014 at the surface. The black star in northern Alaska shows where Utqiagvik is located. The black circle shows Alert, Canada, the black diamond shows Villum in Greenland, while the black pentagon shows Zeppelin, Svalbard. All the results are shown north of 50N.

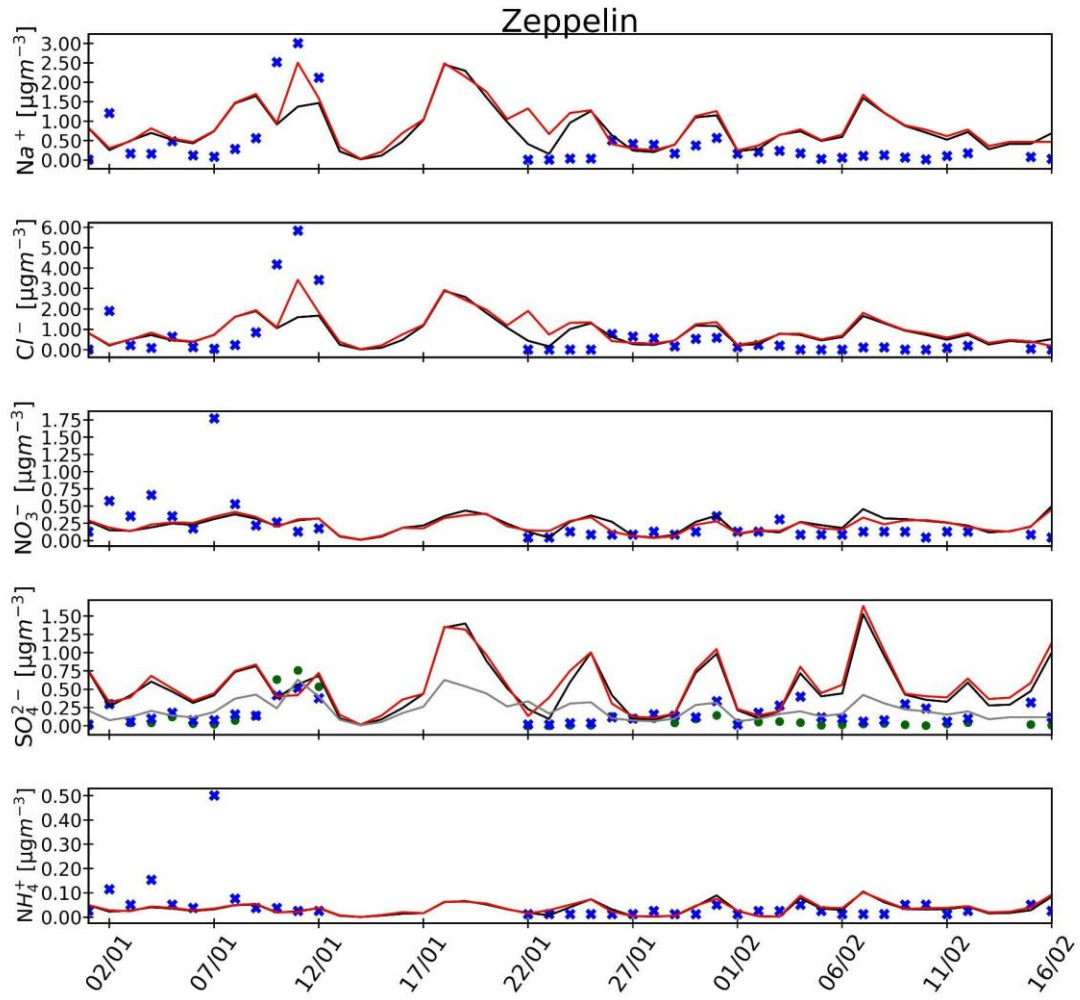


Figure 2: Evaluation of modelled aerosol composition against in-situ aerosol observations with $d_a < 10 \mu\text{m}$ at Zeppelin. The HEM_NEW simulation is shown with (red lines) and without (black lines) N_2O_5 hydrolysis. Dark green circles show observed ss-SO₄²⁻, while the grey line shows modelled ss-SO₄²⁻ from HEM_NEW including N_2O_5 hydrolysis.

Reviewer 1

This work applies the WRF-Chem model to the Arctic and compares with observations, from January and February, 2014, in an effort to evaluate and improve the model's capabilities to represent the atmospheric aerosol over the Arctic with a particular focus on the modelling the main chemical components of sea spray.

Comparisons of simulations from a "Control" version of the model and from an updated version ("HEM_NEW") are drawn with coarse particle observations of nitrate, sulphate, chloride, ammonium and sodium from Alert, Villum and Zeppelin. The domain, d1 shown in Figure 1, is simulated at a resolution of 100x100 km. Then, 'Control' and 'HEM_NEW' comparisons are made with fine particle observations from Simeonof and Gates of the Arctic, for the same suite of chemical components. In this case, the model domain (d2) is nested within the d1 domain, and the simulations are a resolution of 20x20 km. (Note: in Figure 1, Simeonof is shown outside of d2, but in Figure 3 it appears that the model resolution is the same for both Simeonof and Gates of the Arctic; please clarify). Subsequently, comparison of 'Control' and 'HEM_NEW' are drawn with observations of both super-micron and sub-micron particle composition measurements from the Utqiagvik Observatory within d2 at 20x20 km resolution. The differences between "'Control' and 'HEM_NEW' could be more clearly delineated, perhaps in a Table. I understand them to be the addition of marine organic aerosols, the addition of a sea-surface temperature component for sea spray emissions, application of satellite data to improve the whitecap fraction and the addition of a sea-salt sulphate component.

To clarify, the CONTROL and HEM_NEW simulations were run at 100 km. The results from these simulations are evaluated against observations over the wider Arctic and sub-Arctic, including stations in the south (Simeonof) and north (Gates of the Arctic) of Alaska. To clarify the domain maps have been updated in Figure 1.

Figures 4, 5 and 6 (now discussed in Section 5) show the comparison between CONTROL and HEM_NEW runs at 100km and the observations. In the case of Alert and Zeppelin, model results are compared to data corresponding to aerosols with diameters $< 10 \mu\text{m}$. At Villum the observations are total suspended particulates (TSP). Results are compared to fine-mode (diameters $< 2.5 \mu\text{m}$) aerosol data at Simeonof and Gates of the Arctic. There was a mistake in the text (line 168-submitted manuscript) about the size cut-offs of the measurements. The text has been corrected. We added the following text in Section 3 (lines 201-203):

"We note that, apart from the sub-micron observations at Utqiagvik and the fine mode observations obtained from the IMPROVE database, there are no other sub-micron or fine-mode observations in the Arctic, as also shown in the recent paper by Schmale et al. (2022)."

The model results at 20 km are discussed in Section 6 (old Section 5). The comparison with Gates of the Arctic data and 20km model results is provided in Appendix F. Simeonof data are not used to validate the model at 20 km since the site is outside the nested (d2) domain.

We now summarise the main updates to the model treatment of SSA emissions in the new Section 4 before discussing the results from 100km runs with (HEM_NEW) and without these updates (CONTROL). Details about SSA treatments and updates are now included in Table 2.

In section 5, Table 3 is a bit confusing, but it appears that ‘HEM_NEW’ run at 20x20 km resolution over Alaska becomes the “Alaska_Control” and “NEW_Alaska” is ‘Alaska_Control’ with updated dry deposition code, a local source of marine organics, a modified sub-micron particle dependence on wind-speed and increased resolution of seaice fractions.

We apologise for the confusion. HEM_NEW results at 100km are used as initial and boundary conditions for the runs on the d2 domain at 20km over northern Alaska. To clarify, Table 4 (Table 3 in the submitted manuscript) has been updated. In particular, we updated the title “WRF-Chem model simulations over northern Alaska” and the entries for ALASKA_CONTROL_FEB has been updated to (HEM_NEW run at 20 km for February 2014) and also, ALASKA_CONTROL_JAN to (HEM_NEW run at 20 km for January 2014).

Overall, the paper is an important document of justified changes to a model, made to improve its ability to represent the Arctic aerosol. The paper is long, a bit repetitious in a few spots, and reasonably well organized. It could benefit from a careful look at little details, including some of the figures. I have one major concern and a number of minor comments.

We thank the reviewer for these remarks. The paper has been shortened and the section on evaluation of the results at 100km has been re-written. The Abstract, Introduction and Conclusions have also been re-written to make the goal and outcomes of the study clearer.

Major comment:

1. Sea-spray aerosol (SSA) is a major component of this paper, and I agree that it is an important topic. However, Arctic Haze is in the title and nss-sulphate has been the major component of Arctic Haze. The deficiency in modelled submicron sulphate at Utqiagvik is substantial (Fig. 4b and 11), but little discussion is given to it. The sulphate time series is relatively flat in Figure 4b, which seems unrealistic. Sub-micron aerosol at Alert, Villum and Zeppelin is neither shown nor discussed, aside from the reference to OM at Alert on line 429. My concern here is that if the model does not simulate Arctic Haze well, that transport of sea spray from more distant sources may be a problem as well. If so, this could jeopardize your conclusion about open leads as the major source of SSA. On the other hand, sub-micron nss-SO₄²⁻ at Alert during January and February, 2014 was unusually low, and may be more consistent with your simulations. Do you have any references that indicate the model does well with Arctic Haze, or would you show comparisons of submicron sulphate from the model with observations at Alert, Villum and Zeppelin? This issue needs to be dealt with in the paper.

We thank the reviewer for their insights. The Title of the paper has been updated to “Modelling wintertime sea-spray aerosols under Arctic Haze conditions” to better reflect the fact that this study focuses primarily on SSA under wintertime Arctic Haze conditions.

We agree that nss-SO₄²⁻ is an important component of Arctic Haze. However, the main goal has been clarified and we focus on understanding and improving model simulations of wintertime Arctic SSA, including its effects on other inorganic aerosols. As such, there is now less emphasis on Arctic Haze in the Introduction. Since we introduce a sea-salt (ss) source of SO₄²⁻ in the model, we evaluate model performance in terms of total, nss-SO₄²⁻ and ss-SO₄²⁻ compared to available observations (in the new Section 5).

We also address the reviewer’s concern that difficulties in modelling nss-SO₄ may also point to issues in modelling fine-mode SSA transported from outside the Arctic. Whilst the model underestimates sub-micron nss-SO₄²⁻ (and SSA) at Utqiagvik, it overestimates at Zeppelin ($d_a < 10 \mu\text{m}$), and also, to a lesser extent, at Villum (TSP) while the model captures observations

at Alert ($d_a < 10 \mu\text{m}$). Therefore, the model's behaviour at Utqiagvik is not the same over the wider Arctic and provides some confidence in the model ability with regard to long-range transport. We also note NH_4^+ is well modelled in general. We emphasise that it is important to include ss-SO_4^{2-} in models since this can make an important contribution to observed total SO_4^{2-} (at some locations). Discrepancies in the model results may be due to different processes affecting observations at sites in different parts of the Arctic. Total SO_4^{2-} , ss-SO_4^{2-} , and therefore nss-SO_4^{2-} , are well captured at Simeonof to the south of Alaska, while the model underestimates sub-micron observations at Utqiagvik and fine mode observations at Gates of the Arctic during certain episodes with elevated concentrations. Missing regional anthropogenic emissions from the North Alaska Slope oilfields and reactions forming (fine-mode) SO_4^{2-} (in the dark or on SSA) could help to explain these discrepancies.

We further address the deficiencies in modelled SSA at Utqiagvik with the runs at 20km over northern Alaska for the periods corresponding to the Kirpes et al. (2019) field campaigns (now Section 6). The campaign observations showed that 75%, by number of fresh SSA were of local origin from open leads in the sea-ice near Utqiagvik. 73% of partially aged SSA, by number, in sub-micron contained secondary sulfate. Thus, deficiencies in modelled nss-SO_4^{2-} at this site do not necessarily jeopardise our conclusions about the source of super-micron and also, albeit to a lesser extent, sub-micron SSA being of local origin. This is also supported by our analysis of depletion factors in Section 6.4. Nevertheless, SSA emissions from open leads might be too low in the model, due to uncertainties about these emissions and also uncertainties the meteorological reanalysis products which may not properly resolve open leads in the sea-ice. Kirpes et al. (2019) stated that open leads might emit more SSA, including organics, per surface area than the open ocean and this is not yet included in the model, or models in general. It is also possible that other processes, such as wet or dry deposition over source regions or during transport might be overestimated in the model, affecting SSA and nss-SO_4^{2-} transported from lower latitudes. In the recent Arctic Council AMAP multi-model evaluation, Whaley et al. (2022) stated that vertical mixing, long-range transport, and deposition are highly uncertain processes in models, and also showed that models often underestimate SO_4^{2-} in Arctic winter.

There is now a section in Section 5 on evaluation of model sulphate aerosols over the Arctic (lines 392-419):

“Sulphate: Figures 4, 5 and 6 show observed ss-SO_4^{2-} and total- SO_4^{2-} together with results from CONTROL and HEM_NEW. With regard to total simulated SO_4^{2-} , the addition of ss-SO_4^{2-} improves the model results, for example, at Simeonof where observed fine mode ss-SO_4^{2-} makes a significant contribution (30-80%, up to $0.3 \mu\text{gm}^{-3}$) to total SO_4^{2-} . ss-SO_4^{2-} also contributes between 10-40% of total SO_4^{2-} at Alert and Villum and modelled ss-SO_4^{2-} agrees better with the observations. The remainder is nss-SO_4^{2-} , a dominant component of Arctic Haze resulting from long-range transport from sources in Russia and Europe at these sites (Leaith et al., 2018; Lange et al., 2018). Model results are at the lower end (up to $0.3 \mu\text{gm}^{-3}$) of reported sub-micron nss-SO_4^{2-} mass concentrations ($0.3\text{-}1.1 \mu\text{gm}^{-3}$) at Alert during winter 2014 (Leaith et al., 2018). On the other hand, HEM_NEW further overestimates total observed SO_4^{2-} at Zeppelin due to the inclusion of ss-SO_4^{2-} especially during certain episodes with elevated concentrations. We note that Zeppelin is a mountain site at 471m, and thus discrepancies with the observations may also be due to issues simulating the vertical distribution and transport of nss-SO_4^{2-} from Eurasian source regions (Hirdman et al., 2010). At Utqiagvik, on the northern coast of Alaska, most of total observed super-micron SO_4^{2-} is ss-SO_4^{2-} (up to $0.18 \mu\text{gm}^{-3}$, around 80%), and the inclusion of ss-SO_4^{2-} in HEM_NEW

improves agreement with the observations. With regard to total sub-micron SO_4^{2-} , high mass concentrations are observed at Utqiaġvik compared to other Arctic sites, consisting mostly of nss- SO_4^{2-} , peaking at $2.4 \mu\text{gm}^{-3}$, much higher than total super-micron SO_4^{2-} (peaking at $0.5 \mu\text{gm}^{-3}$), as also reported by Quinn et al. (2002). However, the model underestimates nss- SO_4^{2-} at this site. As noted by KRP18 and KRP19, this is likely to be due to the local influence from the North Slope of Alaska (NSA) oil fields to the east. In a companion paper, Ioannidis et al. (2022, in prep.) the influence of these regional emissions on BC at Barrow is investigated during winter 2014 finding that up to 30-50% of BC may originate from this source. Indeed, at GoA, 391km inland from the coast, and south of the NSA oil fields, the contribution of nss- SO_4^{2-} is more important and ss- SO_4^{2-} is negligible. The model captures total fine mode SO_4^{2-} (peaking up to $0.64 \mu\text{gm}^{-3}$) at this site and the addition of ss- SO_4^{2-} does not affect the results. As well as local sources, model difficulties simulating sub-micron nss- SO_4^{2-} at Utqiaġvik may be due to underestimation in transport of mid-latitude sources. SO_4^{2-} formation mechanisms under dark, cold winter conditions may also be lacking in the model. For example, high concentrations of hydroxymethane sulphonate (HMS) have been measured recently during winter in Fairbanks, Alaskan Interior (Campbell et al., 2022) contributing to secondary SO_4^{2-} formation during Arctic winter, although only a small contribution from HMS to SO_4^{2-} was found in observations at Oliktok Point, situated within NSA oil fields (Liu et al., 2021a). Oxidation of SO_2 by ozone in alkaline SSA could also contribute up to 9% to SO_4^{2-} formation (Alexander et al., 2005). However, the version of WRF-Chem used here does not include such reactions, in common with many chemistry-aerosol models run over the Arctic (Whaley et al., 2022).”

With regard to the available data, observations at Alert and Zeppelin are aerosols with aerodynamic diameters below $10 \mu\text{m}$, while at Villum the observations are TSP. Based on recent assessments of Arctic aerosol observations, sub-micron or fine mode nss- SO_4^{2-} and OA data in the high Arctic are not available on a continuous basis (Schmale et al., 2022; Moschos et al., 2022) and not for winter 2014. We clarify this in the revised manuscript (lines 201-203) (see our reply above).

We also added a discussion in Section 5 comparing the model results with previous studies reporting data at various sites, notably, Leaitch et al. 2018 (Alert), Neilsen et al. 2019 (Villum), Moschos et al. 2022 (Utqiaġvik, Alert, Villum).

Minor comments:

We have taken into consideration all of the minor comments below. However, since the manuscript has been significantly revised, it is sometimes difficult to point to revisions of specific line numbers.

2. Line 4 – Concerning organics, you might consider here the paper by Mungall et al.: Microlayer source of oxygenated volatile organic compounds in the summertime marine Arctic boundary layer, P. Natl. Acad. Sci. USA, 114, 6203, <https://doi.org/10.1073/pnas.1620571114>, 2017.

Thank you for pointing out this study. Whilst this is indeed an interesting paper, we note that this is a summertime study highlighting the potential emission of OVOCs from open sea water, whereas our study focuses on wintertime when only open leads are present. Nevertheless, we recognise that such processes may also be contributing to wintertime production of organic aerosols, as noted by Moschos et al. (2022).

3. Line 16 – Maybe reduced biases, instead of improved biases.

The text is rephrased as suggested.

4. Line 31 – “mid-latitude”. Corrected

5. Line 55 – Also important to further knowledge about the vertical extent of SSA.

Thank you for this pertinent remark. We now refer to this in the Conclusions since vertical measurements of SSA are needed to improve understanding about their effects on clouds and radiative forcing.

“As well as ground-based measurements, vertical profiles of SSA components are also needed to better understand SSA sources and their impacts on clouds.” (lines 747-748).

6. Line 110 – Instead of “All the various processes”, maybe “The well-known processes” or “Basic processes”. We rephrased using the “well-known processes” suggestion.

7. Line 112-113 – What about dust? It can be an important factor in the Arctic.

We agree that it is indeed important to mention how dust is simulated in the model. It is now mentioned in Section 2.

“Dust emissions in the MOSAIC aerosol scheme are also calculated following Shaw et al. (2008).” (line 137)

8. Section 3.1 – With respect to Alert, I suspect the coarse-particle chemistry is based on the difference between high-volume essentially TSP) filters collected outside and submicron filters collected inside. The high uncertainties may be related to uncertainty in cut size of the submicron filters. Also, the high-volume filters are susceptible to blowing snow, which may (on occasion) bias concentrations a bit high.

Thank you for your insights about the data at Alert. We added the following in the text:

“These high uncertainties may be related to uncertainties in the size cut-off of sub-micron filters. Uncertainties in coarse particle observations are based on the difference between high-volume (TSP) filters collected outside and sub-micron filters collected inside.” (lines 172-175)

9. Line 224 – “long-raNge”. Corrected.

10. Lines 228-229 – The sea around Alert is also frozen in winter.

Thank you. This information is noted in the discussion in Section 5.

11. Figure 2 – I suggest only showing the weekly-averaged model points to enable the scale be expanded to show the proper comparisons better.

We updated all the figures showing time series. However, both weekly-averaged and daily model results are still shown to compare with the data and also the full time series from the model results (output every 6 hours) because we want to illustrate model variability during January and February 2014.

12. Figure 6 and like figures – Can you place the labels horizontally above or below the globes?

Yes, we updated the maps.

Figure 3 - What is the difference between the blue crosses and the grey dots in SO₄²⁻ in Figure?

The plots for SO₄²⁻ have been updated and we now show observed and modelled total SO₄²⁻ and ss- SO₄²⁻. The caption for Figure 4 has been revised (between lines 381 and 382) :

“Evaluation of modelled aerosol composition against in-situ fine mode aerosol observations (both sites) at (a) Simeonof, Aleutians Islands, Alaska and (b) GoA, north of Alaska, in local Alaskan time (AKST). Observations are shown as blue crosses, only when they are available. For observed SO₄²⁻: dark-green circles show ss-SO₄²⁻, while blue crosses are total SO₄²⁻.”

The black line shows model results from the CONTROL run; the red line shows results from the HEM_NEW run. Model daily or weekly averages are shown as black diamonds for CONTROL and red pentagons for HEM_NEW, while grey circles show ss-SO₄²⁻ for the HEM_NEW simulation. See the text for more details. Note the different scales.”

14. Lines 401-403 – This may be too simple a solution. The aqueous phase is unlikely at Alert in winter, unless in fine haze particles (possibly quite acidic). Transport to Alert through liquid clouds at more southern latitudes is possible, but then some sunlight might be a factor too.

We thank the reviewer for this comment. We updated the discussion about this. Please see the earlier reply.

15. Line 407 - It is not so easy to see this for NH₄⁺, since the concentrations of NH₄⁺ are quite small. What is the uncertainty in the NH₄⁺ measurements?

In Section 3 of the submitted manuscript, we reported the uncertainties for sub-micron observations of inorganic aerosols, including NH₄⁺ at Utqiagvik. We now added information about super-micron observation uncertainties at Utqiagvik. The uncertainties for sub-micron ammonium at Utqiagvik are 0.2 +/- 7.8% µgm⁻³. (lines 191-194)

16. Line 415 and Figure 4b - It is very difficult to tell from this figure that lower concentrations are better represented by HEM_NEW.

The plots have been updated.

17. Line 418 – Modelled sub-micron OA...

Corrected.

18. Line 429 – parenthesis after “2” not needed.

Corrected.

19. Line 455 – “difficulties in capturing sub-micron SSA and nss-SO₄ during wintertime...”

Corrected.

20. Line 462 – Demonstrations of the importance of wet and dry deposition goes back further than 2007. Maybe “the importance of the formulation of wet and dry removal...”?

The discussion about dry deposition was removed to reduce the length of the paper, and address the concerns of Rev. 2. We do still mention that uncertainties in deposition processes may explain some of the model discrepancies e.g. lines 368, 476, 716.

21. Figure 7 caption – “sub-micron aerosol mass concentrations” instead of aerosol mass concentrations for sub-micron”.

Corrected.

22. Line 514 – not be expected

Corrected.

23. Line 517 – “in clean regions of the Arctic” is a little too broad. Wasn't it near Zeppelin? Also, clean regions of the Arctic is contradicted by Arctic Haze.

Indeed, we agree. To avoid any confusion, we removed the phrase “from the clean regions”. Now the relevant discussion is in lines 494-497.

24. Lines 621-622 – Maybe “Overall, the Alaska-New January simulations of sub-micron Na⁺ and Cl⁻ are an improvement over the CONTROL, but still ...”

Corrected.

25. Line 667 – “not a significant source of SSA”

Corrected.

26. Lines 726-728 - This is misleading, unless you can show that submicron aerosol at other sites compares well.

This text has been updated (lines 709-713):

“The addition of $ss\text{-SO}_4^{2-}$ agrees well with $ss\text{-SO}_4^{2-}$ derived from the observations at most Arctic sites and leads to improved modelled total SO_4^{2-} . However, at Zeppelin and Villum, which are dominated by $nss\text{-SO}_4^{2-}$, this additional source results in further overestimation. While super-micron SO_4^{2-} , primarily of sea-salt origin, is captured better at Utqiagvik on the northern Alaskan coast, sub-micron SO_4^{2-} is underestimated at this site, also at Gates of the Arctic further inland.”

27. Lines 729-730 - Not to underestimate the potential of aqueous-phase processes, I feel that the third reference to metal-catalysed aqueous-phase oxidation, without a comprehensive discussion, over-emphasizes the potential of this process.

The text about possible missing reactions has been updated with less emphasis on this possibility:

“Model discrepancies in OA and $nss\text{-SO}_4^{2-}$ may be due to missing local anthropogenic emissions, coupled with missing heterogeneous or dark reactions leading to secondary aerosol formation. In the case of OA, primary marine emissions may also be underestimated. It can be noted that such underestimations are a common feature in many other models (Whaley et al., 2022). Uncertainties in wet and dry deposition processes may also be responsible for deficiencies in modelled aerosols at quasi-hemispheric and regional scales (Whaley et al., 2022).” (lines 713-717)

28. Lines 766-769 – By radiative effects in the darkness of winter, I assume you are suggesting that SSA contributes to changes in longwave radiation or is somehow involved with ice crystals. A little more detail would help, including a reference or two.

Yes, we included a revised discussion:

“Ice fractures and the area of open ocean are likely to become more important with decreasing sea-ice cover in the Arctic as a result of climate warming. This may lead to more SSA which can act as CCN or INPs with implications for Arctic aerosol-cloud indirect effects, notably long-wave radiative forcing which dominates in winter (Eidhammer et al., 2010; Partanen et al., 2014). As well as ground-based measurements, vertical profiles of SSA components are also needed to better understand SSA sources and their impacts on clouds. Such studies will ultimately help to reduce uncertainties in estimates of aerosol-cloud indirect radiative effects and the magnitude of the associated radiative cooling (Liu et al., 2021b) or warming (Zhao and Garrett, 2015).” (lines 744-751)

Below we provide the new references we added in the revised manuscript:

1. Adachi, K., Tobo, Y., Koike, M., Freitas, G., Zieger, P., and Krejci, R.: Composition and mixing state of Arctic aerosol and cloud residual particles from long-term single-particle observations at Zeppelin Observatory, Svalbard, Atmospheric Chemistry and Physics, 22, 14 421– 14 439, 2022.
2. Berg, L. K., Shrivastava, M., Easter, R. C., Fast, J. D., Chapman, E. G., Liu, Y., and Ferrare, R.: A new WRF-Chem treatment for studying regional-scale impacts of cloud processes on aerosol and trace gases in parameterized cumuli, Geoscientific Model Development, 8, 409–429, 2015.
3. Browse, J., Carslaw, K., Mann, G., Birch, C., Arnold, S., and Leck, C.: The complex response of Arctic aerosol to sea-ice retreat, Atmos. Chem. Phys., 14, 7543–7557, 2014.
4. Burrows, S. M., Hoose, C., Pöschl, U., and Lawrence, M. G.: Ice nuclei in marine air: biogenic particles or dust?, Atmospheric Chemistry and Physics, 13, 245–267, 2013.
5. Chapman, E. G., Gustafson Jr, W., Easter, R. C., Barnard, J. C., Ghan, S. J., Pekour, M. S., and Fast, J. D.: Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating

- the radiative impact of elevated point sources, *Atmospheric Chemistry and Physics*, 9, 945–964, 2009.
6. Chen, Y., Cheng, Y., Ma, N., Wei, C., Ran, L., Wolke, R., Größ, J., Wang, Q., Pozzer, A., Denier van der Gon, H. A., et al.: Natural sea-salt emissions moderate the climate forcing of anthropogenic nitrate, *Atmospheric Chemistry and Physics*, 20, 771–786, 2020.
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 11. Gantt, B., Kelly, J., and Bash, J.: Updating sea spray aerosol emissions in the Community Multiscale Air Quality (CMAQ) model version 5.0. 2, *Geoscientific Model Development*, 8, 3733–3746, 2015.
 12. Gong, S.: A parameterization of sea-salt aerosol source function for sub-and super-micron particles, *Global biogeochemical cycles*, 17, 2003.
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Reviewer 2

The manuscript by Ioannidis et al. reports model simulations over the Arctic with the aim of improving SSA predictions. The study is a large effort trying to examine the impact of various recent parameterisations on the model output and agreement with the observations. Despite a significant effort by the authors it falls short of advancing the understanding of processes and their impact on model output. Just for a start, the paper title emphasizes Arctic haze while the main stated purpose is to advance SSA predictions. I am not suggesting that Arctic haze is not important, but the current version is a mix of everything: mostly sea spray, but frequently interspersed by acidic components and anthropogenic sources. That does not help to deliver focused conclusions, because there are already too many issues related to sea spray alone.

We thank the reviewer for their valuable comments and insights. We have made major revisions to the manuscript, in particular, to address the reviewer's concerns about the overall objectives and outcomes of the study. We first summarise the major revisions, followed by detailed replies to the reviewer's concerns.

The Title has been revised to better reflect the aims of our study which, as the reviewer points out, has an important focus on SSA:

“Modelling wintertime sea-spray aerosols under Arctic Haze conditions”

The Introduction has undergone major revision in order to clarify the motivation for this study, and to make our study objectives clearer and more focused. We emphasise that enhanced concentrations of aerosols during Arctic wintertime include both anthropogenic (Arctic Haze) and natural components, including sea-salt aerosols (SSA). An important motivation for our study is the findings of Kirpes et al. (2018, 2019) who observed, during wintertime campaigns at Utqiagvik (northern Alaska), evidence for local SSA sources, including marine organics. They also observed that partially aged SSA (a large number fraction of sub-micron aerosols) were always internally mixed with secondary sulphate or with both sulphate and nitrate aerosols, indicating that multiphase reactions are occurring during transport. Previous studies have also shown that SSA can influence anthropogenic nitrate and sulphate aerosols via heterogeneous uptake of acid gases on the surface of SSA and that this can also influence aerosol size distributions (Chen et al., 2016; Li et al., 2018; Wu et al., 2019). For example, heterogeneous uptake of nitric acid on SSA produces nitrate aerosols, in particular over anthropogenic NO_x emissions regions in mid-latitudes, which can then be transported to the Arctic and contribute to Arctic Haze during winter. It is therefore important to consider SSA together with other inorganic aerosols when investigating wintertime Arctic aerosols.

We also make it clearer that the main goal of the study is to investigate Arctic wintertime SSA and to evaluate and improve the WRF-Chem model. The SSA emission scheme in the base version of the model is based on Gong et al. (1997). Since this has been shown to have deficiencies, we update the scheme using more recent updates. We evaluate the performance of the updated model compared to the base version and compared to observations over the wider Arctic.

The description about the treatment of SSA emissions in the model has been improved. We created a new Section 4 describing the SSA emission scheme in the base version of the model

(used in the CONTROL run) and the updates to the treatment of SSA emission (run HEM_NEW).

Section 5 (previously Section 4), which evaluates results from the quasi-hemispheric 100km simulations against observations, has been shortened and revised. The detailed discussion about the observations (lines 223 to 266 in the submitted manuscript) has been removed. This section now focuses on the evaluation of modelled SSA, and related inorganic and organic aerosols, which in the case of SO_4^{2-} and OA include a sea-salt component. A table comparing biases with and without the updates to the SSA emission scheme is now included in the main text (Table 2). We highlight interactions between SSA and other inorganic aerosols, notably NO_3^- . Since it is interesting to examine the effects of the model updates on aerosol acidity, include a short discussion about NH_4^+ and neutralisation factors (details about the latter have been moved to Appendix D).

We removed the text about on dry deposition in the section about the sensitivity of model results to various processes which could be influencing SSA over northern Alaska (old Section 6). We also removed Figure 8 which showed super-micron SSA at 20km since only weekly observations are available, and the model performance is quite good at 100km.

The Abstract and Conclusions, as well as concluding remarks at the end of different sections, have been revised to make the results of our study clearer.

However, the most significant problem is related to the implementation of the study. What was the purpose of using an outdated sea spray source function – Gong97, which is not even based on observations, but is rather a mathematical extension of an even older, although pioneering at a time, study of Monahan, combined with the slightly more contemporary observational data of O'Dowd et al. 1997?

Being instrumentally limited, even O'Dowd et al. size distribution stopped at 0.1µm in diameter when there are plenty of recent papers providing evidence of large numbers of sea spray particles down to 10nm (L. Cravigan et al. 2015 JGR, A. Schwier et al. 2015 ACP, J. Ovadnevaite et al. 2014 ACP, W. Xu et al., 2022 Nature Geoscience just to name a few). Surprisingly, none of the above papers are mentioned despite a clear contextual value.

That is even more surprising given the choice of the state-of-the-art WRF-Chem model. If sea spray model output so outdated and diverging with observations, how can anyone trust inferences on marine organic matter, ssSO₄ and processes taking place in internally mixed particles?

A main aim of this study is to evaluate and improve the treatment of the SSA emission scheme in WRF-Chem run with the 8-bin MOSAIC aerosol scheme. It focuses on the Arctic during wintertime, a period of the year which has been much less well studied in general, and not at all in studies using WRF-Chem. Previous studies in our group focused on looking at aerosols, but not SSA, in spring and summer. Our results have also contributed to year-round model evaluations, such as the recent *Arctic Monitoring and Assessment Programme (AMAP) assessment* (e.g. see Whaley et al., 2022), but SSA was not a focus of these evaluations.

As noted above, the Gong97 (G97) SSA emission scheme, using O'Dowd et al. (1997) observations (down to 0.1µm), is coupled to the MOSAIC aerosol scheme (8-bins) is used in the base version of WRF-Chem used in this study. The new Section 4 now describes the original G97 scheme together with updates to the treatment of SSA emissions. Whilst, we agree that G97 is out-dated, a goal of our study is to improve it. The scheme in WRF-Chem

has since been coupled to the Fuentes parameterization for small particles, and including marine organics (Fuentes et al., 2010; Fuentes et al., 2011) (F10 from now on) following the work of Archer-Nicholls et al. (2014). Figure 14 in F10 shows the size distribution of different source functions including recent studies. The size-resolved source flux function from F10 extends from 3nm to 450 nm. This can be compared with, for example, Clarke et al. (2006) (surf zone), or the studies mentioned by the reviewer, which include particles down to 10 nm. The F10 source function is applied in WRF-Chem from the lowest model bin (1st MOSAIC bin, 39-78 nm). Within each MOSAIC bin, SSA emission fluxes are calculated for 1000 sub-bins. Note that the model does not represent aerosols smaller than 39 nm explicitly and nucleation is parameterised. This information is included in the revised manuscript (Section 4.2). In this case, results from O'Dowd et al. (1997) are not used. The F10 scheme coupled to G97 is an option in the model version we are running but it had not been activated in previous simulations of Arctic aerosols. Our study is the first to evaluate model simulations of Arctic wintertime SSA aerosols using the F10 option.

In addition, we also update the treatment of SSA emissions using more recent dependencies for wind speed and including a dependence on SSTs, as well as a source of sea-salt SO_4^{2-} . We evaluate the performance of the model run with G97 (CONTROL) and with the updates to SSA emissions (HEM_NEW) under wintertime conditions over the Arctic. It can be noted that there are only very limited field campaigns in this region at this time of year aiming to investigate SSA emission processes and providing data to improve and evaluate models. In Section 6, make use of these limited studies (notably Kirpes et al. (2018) and Russell et al. (2010)) to investigate model sensitivities over northern Alaska. Thus, our study is a first step towards improving SSA treatments under these conditions.

It can also be noted that many chemistry-aerosol-climate models are still using SSA emission schemes based on G97 or Gong (2003) parameterisations, (e.g. GEOS-Chem, HadGEM3, CNRM-ESM2-1), and including sub-micron SSA emissions. Barthel et al. (2019) showed that modelled SSA are affected more by dependencies on SSTs than the use of different source functions, based only on wind speed discrepancies. Compared to other models, e.g. GEOS-Chem (Jaeglé et al., 2011; Huang and Jaeglé, 2017), CMAQ (Kelly et al., 2010; Neumann et al., 2016), or LOTOS-EUROS (Barthel et al., 2019), there has been little focus on understanding and improving SSA source functions in WRF-Chem using the MOSAIC aerosol scheme. Our study aims to address this by implementing updated treatments for SSA emissions and including sources of marine organics, and sea-salt SO_4^{2-} . Nevertheless, we agree with Rev. 2 that it is important to move towards more realistic SSA source functions in models.

Overall, the updated treatment of SSA emissions in WRF-Chem leads to significantly improved results, in particular for coarse mode and super-micron SSA, including organics and ss- SO_4^{2-} over the wider Arctic and sub-Arctic. The results for the 100 km simulations are now discussed in Section 5 (old Section 4) by aerosol component. We show that by improving coarse/ super-micron SSA, fine and coarse mode NO_3^- are also improved in the model, supporting previous modelling studies (Chen et al., 2016, 2019). We also examine the sensitivity of modelled SSA, and other aerosol components, to wintertime Arctic conditions over northern Alaska, based on findings from Arctic field campaigns. We investigate the effect of including a local source of marine organics, use of a higher wind speed dependence, and sea-ice fractions. These issues have not been addressed in previous studies to our knowledge. We also demonstrate that blowing snow or frost flowers are unlikely to be the source. However, there are still issues to be resolved with regard to simulating episodes of elevated sub-micron SSA at Utqiagvik, and we make some suggestions for further

improvements in the Conclusions. As the reviewer mentions, no model is perfect, and this also applies to WRF-Chem. We discuss possible further areas for improvement in the Conclusions.

We include a link for the MOSAIC module where the SSA emissions are calculated in the “Code availability” part.

We thank the reviewer for pointing out the other studies and agree that these are important, especially with regard to putting our study into a wider context. We were aware of these studies but had not originally included them since our study focuses on wintertime Arctic aerosols. We now cite these papers in the revised manuscript.

I wonder why the authors did not make an attempt to compare with size distribution data. That is a critical aspect, because focusing on mass balance does not advance process understanding and produces little value when it comes to studying aerosol-cloud interactions and predicting CCN. Mass balance is dominated by supermicron fraction in sea spray while the number is dominated by the submicron fraction.

The goal of our study is first to understand and improve simulations of wintertime Arctic aerosols, in particular SSA. We agree that a motivation for this work is to improve knowledge about aerosols which can act as CCN or INP, and that it is useful to compare with size distribution data, especially in the sub-micron range, even if this is more challenging for 3D chemistry-aerosol models. During January-February 2014, sub-micron size distributions are available at Utqiagvik, but not at the other sites discussed in the paper, as discussed by Freud et al. (2017). In addition to comparisons with sub- and super-micron mass concentration data at Utqiagvik, we added a new Figure (11) and discussion comparing results from the 20km simulations to sub-micron size distribution data at Utqiagvik (Section 6.3). See lines 601-612.

“Observations of particle number concentration are also used to validate the regional model results at Utqiagvik (see Fig. 11). High number concentrations are observed during both periods, up to 10^3 particles per cm^3 , especially for particle sizes less than 20 nm. Freud et al. (2017) reported similar wintertime magnitudes in the accumulation mode (diameter range 100-150 nm) at Utqiagvik, averaging between 1×10^2 and 2×10^2 particles per cm^3 , whereas the magnitude is smaller for particles with diameters less than 50nm. They also noted that particle number concentrations are higher at Utqiagvik and Tiksi (in northern Russia) compared to other Arctic sites (Alert, Villum and Zeppelin). The model tends to underestimate observed number concentrations, especially in the 4th (312.5 to 625.0 nm) and 5th (625.0 to 1250 nm) MOSAIC bins, even if the model compares better in January when measured number concentrations are lower. This is consistent with the evaluation of submicron SSA and other aerosol components, particularly for episodes when observed aerosols were enhanced (Fig. 10). Note that the model results cannot be compared to measurements smaller than 39 nm because MOSAIC does not represent these aerosols explicitly and nucleation is parameterised. Inclusion of a source function to account for ultrafine SSA emissions, for example, from breaking waves at the surf zone, may lead to improved model results (Clarke et al., 2006).”

The paper is very long and well organized, but suffers from lack of focus.

We thank the reviewer for their remarks. As mentioned above we have revised the paper to sharpen the focus of the paper and to make the objectives clearer in the Introduction. The manuscript has also been shortened.

For example, take dry deposition section. There is no context of reason given why the impact of dry deposition is explored given the fact that the model generally underestimates SSA and larger dry deposition is only making matters worse.

The aim of this sensitivity analysis was to investigate uncertainties in processes affecting modelled SSA and other inorganic aerosols over northern Alaska. Since the Arctic wintertime boundary layer is very shallow with strong surface inversions, it is possible that modelled SSA would be sensitive to dry deposition. The results showed that modelled super-micron SSA is sensitive to dry removal treatments. However, we removed this sub-section on dry deposition from the paper since, as pointed out by the reviewer, the influence of dry removal on sub-micron aerosols, including SSA, was found to be very small.

The other example is sulphate. Although there is a clear distinction between ssSO₄ and nssSO₄, the model comparison only deals with total SO₄ making all findings or inferences vague as those species come from entirely different sources.

Section 5 (old Section 4) has been re-written and the results are discussed by aerosol component. There is now a discussion about model results versus observations of total, sea-salt and non-sea salt components of SO₄²⁻ for sites over the wider Arctic. Figures 4, 5, and 6 have been updated to show observed and modelled (HEM_NEW) total SO₄²⁻ and ss-SO₄²⁻. However, there was a discussion about ss-SO₄²⁻ and nss-SO₄²⁻ in the submitted manuscript (e.g. lines 399-400 - discussion about the results at the Simeonof site).

The authors devote a large section on neutralization factor, but use total sulphate in calculations despite the fact that only nssSO₄ is making aerosol acidic. By contrast, ssSO₄ is balanced by sea salt cations.

We thank the reviewer for pointing out this oversight on our part. We corrected the discussion about neutralisation factors which has now been moved to Appendix D. We refer to this discussion in the main text (lines 430-438) noting that the SSA updates lead, in general, to slightly less acidic aerosols in the model. This is in better agreement with observation-derived neutrality factors at some sites.

Conclusions are a compilation of mostly speculative statements. Some of them are pure speculations as to suggesting what was not included in the model and what impact it would have had if it was (anyone's guess really), some are speculative in a sense that no quantitative support is given. If something improved the simulations, then by how much? Was better agreement marginal or significant? Same criticism applies to Abstract where there is a single number to illustrate the results.

The Abstract and Conclusions have been rewritten to address the Reviewer's concerns. We include information about which processes, out of the ones considered in this study, are affecting the simulation of wintertime SSA in the model more than others over the wider Arctic and over northern Alaska.

Although the value of the paper would not dramatically improve, a reorganisation of the paper by dropping anthropogenic components and by providing better context, reasoning and the outcome of specific parameterisations would make it more readable and useful.

Please see our earlier replies addressing the reviewer's concerns about the context and objectives of this study. We also tried to improve the manuscript with regard to assessment of specific model improvements in simulated wintertime aerosols, in particular SSA, over the wider Arctic (Section 5) and related to the sensitivity tests over northern Alaska (Section 6), as also mentioned earlier. As noted above, the Conclusions and Abstract have been rewritten. We would like to point out that our study is one of the first studies to examine these issues in the Arctic wintertime (using WRF-Chem). As such, we feel that it does make a valuable contribution to advancing our understanding about wintertime SSA under Arctic Haze conditions.

Other comments as they appeared

Line 2. Models will always have difficulties reproducing observations, because they are just model approximations. Model agreement does not need to be perfect if it captures key processes consistently.

Thank you for this insight. We agree.

Line 8. The statement that the model overestimates sea salt is contradicted by all further results where model consistently underestimates sea salt.

Thank you for pointing this out. This statement has been corrected:

“Models tend to underestimate sub-micron, and overestimate super-micron, SSA in the Arctic during winter, including the base version of the Weather Research Forecast, coupled with chemistry (WRF-Chem), model used here, which includes a widely used SSA source function based on Gong et al. (1997).” (lines 5-8)

Line 10. Sea salt sulphate is part of sea salt, why is it included second time? Please clarify if not.

The original version of WRF-Chem does not include sea-salt SO_4^{2-} . This was pointed out on line 87 in the submitted manuscript. This source was included following the analysis by Kelly et al. (2010) (CMAQ model) (see lines 309-315 in the revised manuscript).

Line 21. Breaking waves (open ocean or open leads) are the main source of SSA, how can it be missing?

We meant that there is no specific treatment of SSA emissions from breaking waves in the surf zone at the coast. SSA emissions are modelled based on the same source function in all oceanic grid cells. However, we removed this text from the Abstract and now mention this point in the main text (e.g. lines 581-583).

Line 55. How come open ocean be a new SSA source if it was always the main one? The statement is either completely wrong or should be rephrased/clarified.

We rephrased the text to clarify this point (lines 54-57):

“Arctic warming is leading to a decrease in summer sea-ice and thinner sea-ice during wintertime (Stroeve et al., 2012). Increases in the area of the open ocean or more open leads in sea-ice may increase winter SSA over Arctic coastal regions, potentially influencing radiative forcing (Ma et al., 2008; Eidhammer et al., 2010; Partanen et al., 2014; Schmale et al., 2022).”

Line 129. Sea spray is not just Na and Cl. Sea spray is sea salt (including sea salt sulphate) and primary organic matter. Those early SSA functions were derived from physical particle measurements, not chemical measurements. This aspect is crucial for comparing model and

observations which often report just Na and Cl and rarely sea salt sulphate, other major ions and almost never OM.

We agree with the reviewer's comment. Models still often lack sources of marine SSA organics and ss-SO_4^{2-} . We therefore improved the model by including these sources. The original SSA emission scheme in MOSAIC, based on G97, treats sea spray emissions only as Na and Cl.

How Gong et al. source function was translated into Na and Cl emissions? Was it necessary at all, because it is rather straightforward to split sea salt into major components, like Na, Cl, ssSO_4 , etc. What was the rationale to improve model simulations by using three decades old source function alongside with newly discovered SSA sources from open leads and frost flowers?

Please see the earlier reply about implementation of the G97 scheme in the model. A goal of our study is to improve this scheme. When using only G97, SSA emissions are assumed to be pure NaCl. The Fuentes option includes a source function for small particles and marine organics as a fraction of total sea-salt mass emissions. Based on the same logic, a source of ss-SO_4^{2-} was also added as part of total sea-salt mass emissions. This is explained in the new Section 4 (see lines 309-315).

Line 187. It is crucial to acknowledge and discuss sampling losses in sea salt observations, because models do not take into consideration of sampling inlets and other sampling artefacts. Sampling losses are mostly related to super-micron range, but depend on specific inlets or samplings ducts. Therefore, given the fact, that the model is generally underestimating observations, sampling losses would make underestimation even more dramatic.

We agree that it is important to take observational uncertainties into account. We added a discussion in Section 3 about sampling losses at least for Utqiagvik where studies have been published. Text below only for Utqiagvik: (lines 194-197)

"Measurement uncertainties are also due to sampling losses in the inlets. At Utqiagvik, for particles with a diameter $10\ \mu\text{m}$ the losses in the inlet system from all loss mechanisms are estimated to be less than 10%, and losses of particles between the diameters of 0.01 and $1\ \mu\text{m}$ are below 5% (Sheridan et al. 2001)."

Figure 2. HEM_NEW or CONTROL simulations were not introduced up to this point.

The names of the simulations at 100km are now introduced in Section 2.

Line 217. How could Fuentes parameterisation for sub100nm be fully utilized if Gong97 or its extension based on O'Dowd97 stopped at 100nm?

When Fuentes 2010 is applied, O'Dowd97 is not used. Please see our earlier reply. This is clarified in the new Section 4.

Line 224. 20% of ss-SO_4 is not a small fraction.

Indeed, however this text has been re-written in the revised manuscript.

Line 233. I am not sure that anthropogenic Na was properly estimated given Cl depletion in anthropogenic air masses, which tends to be translated into excess Na and thus anthropogenic source. The authors do not discuss Cl/Na ratio which would be informative and also considering derivation of ssSO_4 as $0.25 \times \text{Na}$.

Sources of anthropogenic Na^+ are not included in the model (or other models, as far as we know). All Na^+ in the model is natural so the estimation of ss-SO_4^{2-} is not affected.

There is a discussion about $\text{Cl}^-:\text{Na}^+$ molar ratios in Section 6.5 (old 5.6 in the submitted manuscript) about the potential contribution of blowing snow and frost flowers to sub-micron SSA over northern Alaska. We calculated molar ratios of $\text{Cl}^-:\text{Na}^+$ using observations and model results from the regional simulations. We agree that further analysis of this ratio could be informative and may have implications for the determination of ss-SO_4^{2-} . In general, the observations at Utqiagvik do not show indications of Cl^- depletion in super and sub-micron

SSA indicating a sea-water origin. The model results do indicate that some ageing (Cl^- depletion) has occurred. However, this process affects Cl^- rather than Na^+ , making it less likely to affect the estimation of ss-SO_4^{2-} .

Line 259. What was the degree of neutralization considering the fact that NH_3 neutralizes the stronger sulphuric acid and only then the nitric acid?

We updated the text about the discussion about neutralisation and aerosol acidity. The details are now provided in APPENDIX D with brief mention in Section 5 (lines 430-438):

“To investigate aerosol acidity, the mean neutralized factor (f) is calculated as the ratio of NH_4^+ to the sum of ($2\text{nss-SO}_4^{2-} + \text{NO}_3^-$), in molar concentrations, following Fisher et al. (2011), for sites in the Arctic with available observations of these aerosols. When f is equal to 1 then aerosols are assumed to be neutralized, while when $f < 1$ then aerosols are acidic, and more acidic when f is closer to zero (Fisher et al., 2011). In general, higher molar concentrations were observed for nss-SO_4^{2-} compared to NO_3^- and NH_4^+ . Table D1 shows f for observations and the two 100km simulations at the different sites. Overall, modelled f increases due to the improved treatment of SSA and the associated influence on NO_3^- via heterogeneous reactions. Since aerosols are assumed to be internally mixed in the model, NH_4^+ and nss-SO_4^{2-} mass concentrations also vary between the two simulations. Thus, aerosols in HEM_NEW tend to be less acidic (e.g. at Alert and Villum), due to NO_3^- decreases in the coarse-mode/TSP size range. This leads to better agreement with the observed f at Alert, in particular. At Villum, observed aerosols are less acidic than in the model. This could be due to the fact that the model has more NH_4^+ compared to the observations. Only small changes are found at Utqiagvik between the two runs, and the model tends to have aerosols which are slightly more acidic (super-micron) and less acidic (sub-micron) compared to the observations. The small increase in model sub-micron f at Utqiagvik could be due to the increase in sub-micron NO_3^- and insignificant changes in NH_4^+ and nss-SO_4^{2-} . Differences with the observed values could be explained by underestimation of nss-SO_4^{2-} at this site. The calculated f for observations could also be biased low (too acidic), since some of the NO_3^- and SO_4^{2-} are present as Na_2SO_4 and NaNO_3 in the atmosphere, which are not measured.”

Line 278. The authors missed to mention earlier pioneering studies of Blanchard 1976, O'Dowd et al. 2004 Nature and later Ovadnevaite et al. 2011 GRL, 2014 JGR. The authors have a justified liberty of choosing parameterization, but the acknowledgement of earlier studies is advisable.

Thank you for pointing out these studies, most of which we were already aware of. We have added these references to the paper (new Sections 4.2.1 and 4.2.3). Please also see our other replies to related points.

Line 316. Wind speed is indeed an oversimplified dependency most importantly not accounting for increasing and decreasing wind speed relationships. This aspect as well SST dependence together with a new sea salt source function accounting for the sea state was introduced by Ovadnevaite et al. in 2014 ACP.

We now reference this study in the text as noted above. In general, we had only mentioned studies focusing on autumn and winter, and the Arctic and/or polar regions. However, it is indeed important to acknowledge studies focusing on lower latitudes and over the Atlantic and Pacific Oceans, areas where Arctic SSA may originate. Apart from studies focusing on the Arctic studies relevant to other areas were cited in the submitted manuscript, namely Revell et al. 2019 (Southern Ocean), Frey et al. 2020 (Antarctica), Goddijn-Murphy, et al. 2011 (northeast Atlantic), Clarke et al. 2006 (Hawaii), Callaghan et al. 2013 (east coast of USA), and Barthel et al. 2019 (Europe, northeast Atlantic, Africa).

Line 331. Na, Cl, ss-SO₄ and OM does not capture full sea spray as Mg, Ca, K are missing. How was the ss-SO₄ fraction of 9.9% obtained, especially that 7% is arising from basic sea water composition?

K⁺ and Mg²⁺ are not explicitly modelled in MOSAIC as they are usually present in smaller amounts compared to other cations. In MOSAIC, K⁺ and Mg²⁺ are represented by equivalent amounts of Na⁺ (Zaveri et al. 2008). The percentage ss-SO₄²⁻ of 9.9% is due to the way the fractions of different components of the SSA emissions are estimated in WRF-Chem. In the base version of WRF-Chem, SSA mass is assumed to be NaCl, first the fraction of Na⁺ was calculated and the fraction of Cl⁻ as 1 – fraction of Na⁺. Then, the fraction of ss-SO₄²⁻ was estimated as the fraction of Na⁺ multiplied by 0.252. This is explained more clearly in the revised version in the new Section 4 (lines 312-315):

“We find that, on average, the mass fraction of ss-SO₄²⁻ emissions in our simulations is around 9.9% of the total SSA emissions. This can be compared with the CMAQ model where the ss-SO₄²⁻ emissions are estimated to be 7% of the total SSA emissions (Kelly et al., 2010).”

Line 356. Basic inorganic chemistry mandates that NH₃ is first neutralising sulphuric acid (stronger acid) and producing either ammonium bisulfate or sulphate. Only a leftover producing ammonium nitrate which is pretty stable in polar areas.

We added the following in the revised manuscript (lines 425-427):

“Temporal variation in NH₄⁺ during January and February 2014, generally, follow nss- SO₄²⁻ as NH₄⁺ preferentially forms ammonium bisulfate and, to a lesser extent, ammonium sulfate in the particle phase (Schmale et al., 2022), and they have common anthropogenic origins.”

Line 360. f=1 is representing fully neutralized aerosol, not "more". More importantly, only nss-SO₄ (and NO₃) should be used for assessing neutralization, because ss-SO₄ is balanced by other cations in sea water (salt).

Thank you for noting this. This has been corrected, and only nss-SO₄²⁻ is used for both observations and model results. This discussion has been moved to APPENDIX D to reduce the length of the paper and referred to briefly in Section 5.

Line 378. Sulphate and nitrate mass is conserved and has nothing to do with chlorite depletion reactions as model simulation do not output masses of specific salts like Na₂SO₄ or NaNO₃ to attempt Na mass balance.

In the current version of WRF-Chem, Na₂SO₄ or NaNO₃ these salts are included in the model as products from heterogeneous reactions, but are not standard output. Also, MOSAIC assumes aerosols are internally mixed in each bin. We added the above information in the revised Section 2 (lines 118-122). We now discuss the neutralized factor in APPENDIX D (lines 775-790).

Line 480. If model underestimates SSA in general, higher dry deposition only makes matters worse. Clearly larger dry deposition is studied not for the better agreement, but simply for theoretical reasons. Better justification and context is needed in this section.

Please see our earlier reply. This part was intended as a sensitivity test. However, we removed this part from the paper to make it shorter.

Line 558. SSA - wind speed relationship is nonlinear (power of 2-3) with Gong97 on a high end of values. What is the purpose of comparing with linear dependences?

As a sensitivity test, we investigated the influence of using a higher wind speed dependence on sub-micron SSA in the 20km simulations over northern Alaska. This is based on data collected by Russell et al. (2010) in the Arctic, even if data in this region is very limited. They found a linear relationship between sub-micron Na⁺, OM, and wind speed based on data collected during the Arctic leg of the ICEALOT cruise. This point has been clarified in the text (lines 528-530).

“In a sensitivity run, results from RUS10 are used to include a higher wind speed dependence for sub-micron SSA. This linear dependence differs from the power dependencies included in G97, SAL14, and other studies, but is based on empirical relationships determined from analysis of data collected in the Arctic.”

Below we provide the new references we added in the revised manuscript:

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