Comment on egusphere-2022-310
Anonymous Referee #1

Referee comment on "Modelling wintertime Arctic Haze and sea-spray aerosols" by Eleftherios Ioannidis et al., EGUsphere,
https://doi.org/10.5194/egusphere-2022-310-RC1, 2022

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 Barrow 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 organics, 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.

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 sea-ice fractions.

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.

Major
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 Barrow 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-SO$_4^{=}$ 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.

Minor comments
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,
3. Line 16 – Maybe reduced biases, instead of improved biases.
4. Line 31 – “mid-latitude”
5. Line 55 – Also important to further knowledge about the vertical extent of SSA.
6. Line 110 – Instead of “All the various processes”, maybe “The well-known processes” or
“Basic processes”
7. Line 112-113 – What about dust? It can be an important factor in the Arctic.
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.
9. Line 224 – “long-raNge”
10. Lines 228-229 – The sea around Alert is also frozen in winter.
11. Figure 2 – I suggest only showing the weekly-averaged model points to enable the
scale be expanded to show the proper comparisons better.
12. Figure 6 and like figures – Can you place the labels horizontally above or below the
globes?
13. Figure 3 - What is the difference between the blue crosses and the grey dots in SO$_4^{=}$
in Figure?
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.
15. Line 407 - It is not so easy to see this for NH$_4^{+}$, since the concentrations of NH$_4^{+}$ are
quite small. What is the uncertainty in the NH$_4^{+}$ measurements?
16. Line 415 and Figure 4b - It is very difficult to tell from this figure that lower
concentrations are better represented by HEM_NEW.
17. Line 418 – Modelled sub-micron OA...
18. Line 429 – parenthesis after “2” not needed.
19. Line 455 – “difficulties in capturing sub-micron SSA and nss-SO4= during wintertime…”
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…”?
21. Figure 7 caption – “sub-micron aerosol mass concentrations” instead of aerosol mass concentrations for sub-micron”.
22. Line 514 – not be expected
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.
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 …”
25. Line 667 – “not a significant source of SSA”
26. Lines 726-728 - This is misleading, unless you can show that submicron aerosol at other sites compares well.
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.
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.