



EGUsphere, referee comment RC2
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Comment on egusphere-2022-310

Anonymous Referee #2

Referee comment on "Modelling wintertime sea-spray aerosols under Arctic haze conditions" by Eleftherios Ioannidis et al., EGU sphere,
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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.

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 What was the purpose of using an outdated sea spray source function – Gong97, which is not even based on observations, but is rather a mathematical extension of an even older, although pioneering at a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 If sea spray model output is so outdated and diverging with observations, how can anyone trust inferences on marine organic matter, ssSO₄ and processes taking place in internally mixed particles?

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 paper is very long and well organized, but suffers from lack of focus. 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 other example is sulphate. Although there is a clear distinction between $ssSO_4$ and $nssSO_4$, the model comparison only deals with total SO_4 making all findings or inferences vague as those species come from entirely different sources. The authors devote a large section on neutralization factor, but use total sulphate in calculations despite the fact that only $nssSO_4$ is making aerosol acidic. By contrast, $ssSO_4$ is balanced by sea salt cations.

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.

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.

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.

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

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

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

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.

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.

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, ssSO₄, 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?

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.

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

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

Line 224. 20% of ss-SO₄ is not a small fraction.

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 ssSO₄ as 0.25xNa.

Line 259. What was the degree of neutralization considering the fact that NH₃ neutralizes the stronger sulphuric acid and only then the nitric acid?

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.

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.

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?

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

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).

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.

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.

Line 558. SSA - win 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?