EGUsphere, referee comment RC2
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Comment on egusphere-2022-561

Anonymous Referee #2

Referee comment on "Marine nitrogen fixation as a possible source of atmospheric water-soluble organic nitrogen aerosols in the subtropical North Pacific" by Tsukasa Dobashi et al., EGUsphere, https://doi.org/10.5194/egusphere-2022-561-RC2, 2022

The paper by Dobashi et al. explores an important subject of secondary aerosol formation by nitrogen containing precursors over the open North Pacific. These type of studies are rare and difficult to come by and thus deserve to be published. The difficulty with the marine atmosphere research, especially in the Northern Hemisphere, is that it is often impacted by anthropogenic emissions via direct continental outflow or remnant terrestrial background. Multiple lines of evidence are, therefore, needed to rule out such impact and link atmospheric measurements to marine biological processes. The authors made a large effort in analysing multiple tracers or species, but have not discussed the results in coherent and comprehensive manner. At the moment the paper is more like a measurement report without an in-depth analysis of underlying mechanisms or processes and often overselling correlation or similarity for a causal link. I encourage the authors to play devil's advocate and take an opposite side to assess if their argument can be falsified using same data. Therefore, a discussion where all evidence would be weighted pro and against is badly lacking. It may well be that the authors can compile compelling argument for their proposed link between nitrogen assisted secondary aerosol formation and nitrogen fixation in biologically active sea water, but a fair account should be given for the so called "null hypothesis" that observations were coincidental. It is also unclear how further studies should be geared in answering the remaining questions as a simple repetition will deliver same result.

Comments as they appear in the text:

Abstract, line 22 "The result suggests..." (at best) and does not indicate, because the suggested link is only a proposition.

Page 2, line 31. The manual operation of sampling clearly helped to avoid own ship emissions, but what about transcontinental/continental outflow pollution? Air mass

trajectories suggest no contact with land, but they typically do not rule out entrainment of pollution from decoupled boundary layer or free troposphere. Were there any tracers like black carbon measured to rule out anthropogenic impact and confirm marine origin of the sampled air mass? Note that higher WSON concentration observed over ENP where trajectories originated close to the continental boundary.

Page 2, line 37. Size segregated samples were collected every 48hours, but there were 51 fine and 9 coarse particles samples. More confusing are similar percentages of total samples collected. Please clarify why many coarse samples were missing in which case their percentage could not be as high.

Page 3, line 23. Were SSW samples collected overboard with an acid cleaned bucket or using onboard plumbing system into acid cleaned bucket? In any case what was the approximate depth of SSW samples?

Page 4, line 14. NH4 and NO3-N point to anthropogenic origin of nitrogen. NO3 in coarse particles results from chloride depletion by HNO3, while ammonia reduced ammonium ion is reacting with sulphate of whatever origin. Despite relatively low concentration of all inorganic species, their origin is pointing towards anthropogenic sources. It should also be noted that natural ammonia source over the oceans is almost negligible (unless the authors have evidence it is otherwise). I think that such discussion is essential, otherwise paper would look like a report instead of a scientific paper.

Page 4, line 22. Is the similar longitudinal distribution of chlorophyll coincidental or causal? Similarity or regression cannot prove causal relationship unless more substantial support is provided and thoroughly discussed. One way to look at it is to use some specific anthropogenic pollution tracers showing no connection or opposite relationship with WSON and inorganic species.

Figure 3 is as useful as can be misleading as it contains only 17 of 51 fine aerosol samples and contains only 3 points with WSON >10ngN/m3. Most of the marine origin deltaC13 points (20-24 permille) probably belong to WNP region, so the highest value is of those 8 samples where WSON was >10ngN/m3. If those 8 points were eliminated, ENP and WNP regions become indistinguishable.

Figure 4. Absence of the correlation between MSA and WSON could be due to the temperature impact which favours SO4 formation in subtropical latitudes instead of MSA. Have the authors looked at SO4 to WSON correlation. If the source of sulphur and nitrogen species is biogenic, they should correlate to some extent, because 48hour sampling has a footprint of ~1000-1500km and biogenic processes overlap even if underlying mechanisms differ somewhat.

Page 5, line 11. The authors correctly discuss primary versus secondary origin of WSON species, but secondary formation does not necessarily is of marine origin. The real challenge is to rule out anthropogenic sources of gaseous precursors leading to secondary WSON.

Page 5, line 24. Why would N2 fixation result primarily in inorganic dissolved nitrogen and not organic like amines?

Page 6, line 18. Different ratio can arise from preferential production as well as from remnant anthropogenic source. Despite coming back to the same argument of anthropogenic origin I do not object authors argument about biogenic origin of WSON, but rather encourage the authors to look for more direct evidence in discounting anthropogenic origin.

Page 6, line 27. What about amines as a WSON species?

Page 6, line 40. Discussion section is recommended to discuss all information presented in previous chapters and discussing them simultaneously in a comprehensive manner. I encourage the authors of playing devil's advocate for their own advantage in taking critical view to their own data.