

Reviewer #2:

General comment

This study evaluated if the marine biological activity present in the North Pacific Ocean, Bering Sea, and Arctic Ocean can impact biological aerosol particles formation, and the distribution of CCNs and INPs. To answer this, a large set of measurements were performed between September 27 and November 10, 2019 in an oceanographic cruise. The author found that bioindicators, fluorescent particles, INP, and CCN correlate positively, indicating that marine biota contribute substantially as a source of bioaerosols and cloud formation via INP and CCN over the remote Arctic Ocean during periods of high biological activity. The results are of high importance for this remote, valuable, and uncertain region. The measurements were properly performed and the data analyses are correct and consistent. Although the is well-written several parts can be improved. The present manuscript can be accepted for its publication in ACP after the following comments are properly addressed.

We would like to thank the reviewer for providing valuable comments and insights on our work. Our responses are given below. All changes are shown in color in the revised manuscript.

Major comments:

1. All the correlations indicated along the manuscript need to be defined as statistically significant or not.

We have added the P-values for data with correlations larger than 0.5, to show statistical significance in Tables 2 and 3.

2. The discussion of the present results with literature data needs to be improved avoiding redundancy and trying to be concise and clear.

We removed figures and texts about aerosol number concentration and number-size distribution obtained by SMPS, to focus on the CCN and INPs discussions. Overall, the discussion of relationship between bioindicators derived from marine ecosystems, their correlation with fluorescent bioaerosols and particles activated as CCN and INP has been revised to be more concise, particularly in Section 3.4.

3. Some figures need to be improved.

We have carefully reviewed and made revisions for clarity.

Minor comments:

P1, L23: Why the CCN concentration for the Arctic Ocean is indicated as a single value (36 cm^{-3}) and not as a range?

For the characterization with air mass, we have classified periods dominated by terrestrial and Asian continental influences (P1 and P5), periods dominated by marine and biogenic sources (P2 and P4), and periods in the Arctic Ocean (P5). The averaged N_{CCN} in the Arctic

Ocean is described as a single value because it is derived only from a single P3 period. See Table 1 for full ranges and standard deviations.

P1, L26-27: “The averaged INP concentration (N_{INP}) measured at temperatures of -18 and -24 °C with marine sources was 0.01 – 0.09 and 0.1 – 2 L $^{-1}$, respectively” Please indicate the region or period you are referring to.

We corrected in the revised manuscript.

“The averaged INP concentration (N_{INP}) measured at temperatures of -18 and -24 °C with marine sources in the North Pacific and Bering Sea was 0.01 – 0.09 and 0.1 – 2.5 L $^{-1}$, respectively, and that over the Arctic Ocean was 0.001 – 0.016 and 0.012 – 0.27 L $^{-1}$, respectively.” (Page 1, Lines 28-30)

P3, L6: I am not sure that the term “biomaterials” is appropriate. Please reconsider this.

We changed the word “biomaterials” to “microorganism and biological substances” in the revised manuscript. (Page 3, Line 10)

P6, L20: I am not sure that the term “number densities of INPs” is appropriate. Please reconsider this.

We changed the word “number densities of INPs” to “number concentrations of INPs” (Page 6, Lines 24)

P7, L15: should “P3, 11–27 October 2019” be “P3, 10–27 October 2019” as indicated in Table 1?

Yes, exactly. We corrected as suggested in the revised manuscript.

P8, L20, L26 and a long the text: I think it would be more appropriate to call “bloom” and “autumn bloom” as “phytoplankton bloom”.

We changed the words “bloom” and “autumn bloom” to “phytoplankton bloom” in the revised manuscript.

P11, L2-3: “suggesting the possibility of dependence on phytoplankton communities in the different oceanic regions”. What is reported in the Literature?

We added the reference (Taylor et al., 2014) in the revised manuscript.

P12: In line 13 “Previous studies” is mentioned; however, only Park et al. (2020) is cited. Please add more studies.

This description is removed because we have excluded a figure and discussion on the aerosol number-size distribution and aerosol concentrations in the revised manuscript.

P13, L6: I think “such as dicarboxylic acid (i.e., oxalic acid)” should be “such as oxalic acid”.

We corrected as suggested in the revised manuscript.

Section 3.4. The discussion related to CCNs is very confused and hard to follow. Please improve this part.

In Section 3.4, we have excluded data and discussion on the aerosol number concentrations and number-size distribution in the revised manuscript to focus on the CCN and INPs discussions. We also revised the description of CCN parameterization briefly.

P15, L14: “INP formation”. Aerosol particles can form but not an INP. An aerosol particle is capable or not to act as INP.

We changed the word “INP formation” as “INP activation” in the revised manuscript.

(Page 15, Line 13)

P17, L3: Why κ value for the accumulation mode is indicated as a single value (0.57) and not as a range?

This description is based on the literature (Gong et al., 2019), not our results, but κ value for the accumulation mode is single parameter because the CCN activation diameter in the accumulation mode is obtained at 0.08% SS only.

P17, L23-29: I have the impression that this paragraph is repetitive.

We corrected in the revised manuscript.

“It is also unique to find that simultaneously observed NCCN and NINP showed positive correlations (R: 0.95–0.97, Table 3), suggesting that enhanced marine biological sources and the formed fluorescent marine bioaerosols contribute to the increases of both CCN and INPs during a phytoplankton bloom, despite the reduced CCN activity due to the less hygroscopicity. This is consistent with the results from microcosm experiments, which suggest that OM coating on SSAs reduces the surface tension, enhances the emission flux of the particle number concentration, and thus the CCN concentration (Alpert et al., 2015; Ito et al., 2023).” (Page 17, Lines 14-20)

P18, L20: Why averaged NCCN at 0.4 % SS for the Arctic Ocean is indicated as a single value (36 cm⁻³) and not as a range?

For the characterization with air mass, we have classified periods dominated by terrestrial and Asian continental influences (P1 and P5), periods dominated by marine and biogenic sources (P2 and P4), and periods in the Arctic Ocean (P5). The averaged N_{CCN} in the Arctic Ocean is described as a single value because it is derived only from the P3 period. See Table 1 for full ranges and standard deviations.

Tables 2 and 3: indicate if the correlations are statistically significant or not.

We have added the P-values for data with correlations larger than 0.5, to show statistical significance in Tables 2 and 3.

Figure 1: The can different periods P1 to P5 be distinguished in the cruise track?

The cruise track during the P1 and P5 periods overlapped but the segments can be recognized from trajectory start points in each panel. We believe that this is clearly depicted in Figure 1 with separate panels.

Figure 2: Add labels to panel a, b, c, and d. Also, change the color for EC and NO₃ as they look very similar.

We corrected as suggested in the revised manuscript.

Figure 4: I am not sure if panels c and d can be more useful with scatter plots?

The plot of fluorescent particles versus wind speed is presented and discussed in Fig. 5 and here we would like to keep Fig. 4c and Fig. 4d to check the response of the fluorescent particle concentrations to the local wind speed in time series.

Figure 7: y-axis in panel f should go in log scale and y-axis in panel (f) should read "INP concentration".

We corrected as suggested in the revised manuscript (now Fig. 6f).

Technical comments:

P2, L5: Add a reference after "processes".

We added a reference (Ramanathan et al., 2001) in the revised manuscript.

P2, L10: Add a reference after "(INPs)".

We added a reference (Brooks and Thornton, 2018) in the revised manuscript.

P2, L19: "(TEP and protein-containing Coomassie stainable particle (CSP)" It seems that a bracket is missing.

We corrected it in the revised manuscript.

P3, L1: Add a reference after "INPs".

We added references (Sun and Ariya, 2006; Murray et al., 2012) in the revised manuscript.

P3, L3-4: "Some CCN grow to giant CCN". Is it not possible to have a primary GCCN?

We corrected this sentence as follows:

"Some particles may form giant CCN and be activated as cloud droplets of larger size" (Page

3, Lines 7-8).

P3, L11: Add a reference after “observations”.

We added a reference (Xie et al., 2021) in the revised manuscript.

P5, L14: Add a reference after “state”.

We added a reference (Furutani et al., 2008) in the revised manuscript.

P8, L13: I think “...contents and suggesting...” should be “...contents, suggesting...”

We corrected as suggested in the revised manuscript.

P9, L12: “total” of what?

P9 L20: “fraction” of what?

We corrected this sentence as follows:

“representing a 2.5 % fraction of the total particles.” (Page 10, Lines 5-6)

“the fraction of fluorescent particles in the total particles was 7 %.” (Page 10, Line 14)

P11, L9: Add a reference after “ocean”.

We added the references (Russell et al., 2010; Engel et al., 2017; Park et al., 2019) in the revised manuscript.

P11, L29: “study” should be “studies”.

We corrected as suggested in the revised manuscript.

P13, L4: Add a reference after “range”.

We added the references (Facchini et al., 2008; Ovadnevaite et al., 2011) in the revised manuscript.

P13, L32 to P14, L4: Please improve the punctuation here. This is a very long paragraph.

We corrected this sentence as follows:

“CCN activation during summer was affected largely by the less hygroscopic OM associated with marine and terrestrial biological activity, while CCN activation during winter was mainly determined by highly hygroscopic components such as sea salt with limited biological activity (Kawana et al., 2022a).” (Page 14, Lines 15-17)

P14, L19: “(Fig. 8d)” should be “(Fig. 7d)”

We corrected as suggested in the revised manuscript.

We now numbered as Figs.6d because the original Fig 6 has been deleted in the revised manuscript.

P14, L25: I think “lower than elsewhere (i.e., North Pacific Ocean).” Should be “lower than in the North Pacific Ocean.”

We corrected as suggested in the revised manuscript.

P14, L30: Add a reference after “INPs”.

We added a reference (Welti et al., 2020) in the revised manuscript.

P15, L17: I think “but still significant” should be “but still strong” or something similar.

We changed the word “still significant” as “still high” in the revised manuscript. (Page 15, Line 29)

Figure 4: I think “(black line) and fluorescent” should be “(black line), fluorescent”

We corrected as suggested in the revised manuscript.

Reference:

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Murray, B. J., D. O’Sullivan, J. D. Atkinson, M. E. Webb.: Ice nucleation by particles immersed in supercooled cloud droplets, *Chem. Soc. Rev.*, 41, 6519-6554, 2012.

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