Author Response to RC2 for ACP manuscript EGUSPHERE-2024-2159 (Moore et al.)

Our responses to each comment are provided in blue text, with the reviewer comments in black. Line and figure numbers refer to the line numbers in the original submission, for consistency with the Reviewer comments. Quotations in red were added to the revised manuscript.

This work was well designed, the experiments and analysis were thoroughly carried out. I especially appreciate the detailed description of the experiments, which could be really helpful for other scientists in the community to conduct similar work. Overall, this study is excellent.

I have only one comment. The "less clear trend with wind speed" of INP concentration from IS measurements was attributed to "different averaging times, differences in inlet orientations or locations, or differences in the aerosol sampled". I have no objections about the possibility of these factors. However, there might be two other reasons, which could be somehow more important. First, for IS method the particles were formed from evaporation of seawater droplets, later collected on filters, and then washed off by water. The resulted solution should be very similar to the original seawater, except they might have different concentrations. It should have similar ice-nucleating properties as the seawater, which could be confirmed by the similar slopes of the INP spectra for filters in Figure 2a and those for seawater in Figure A7. Therefore, IS actually measured seawater-like solutions. As seawater used in experiments kept the same, it is understandable IS method did not see the trend as CFDC method. Second, the two methods measured INP for particles with different sizes. CFDC measured polydispersed particles. The different size distribution of particles generated by different wind speed would of course result in different INP concentrations. However, the IS equivalently measured several groups (meaning each dilution) of monodispersed seawater-droplets generated particles. For each group (one dilution), the droplets had similar size and might have similar INP properties. If you maintain the concentration of solution by using different amount of water to wash the filters according to the mass collected, you might get even less clear trend with wind speed for IS measurements.

Response: We thank the reviewer for reading the manuscript and providing thoughtful comments. Our apologies, but we do not follow the additional reasons provided for the discrepancy between the IS and CFDC relationships with wind speed. Both the IS and CFDC sampled the same aerosol at the same time, which was produced by wave breaking and bubble bursting within the SOARS channel. Both sampled polydisperse SSA, although differences in the inlets used by the CFDC and IS are hypothesized to be responsible for some of the differences in INP concentration seen between the two instruments, with inlet losses at high wind speed potentially responsible for the unclear relationship between the IS INP concentrations and wind speed. Good agreement between the IS and CFDC at the same temperature is expected and has been seen in laboratory and field measurements in multiple locations and with different aerosol types, including marine aerosol (e.g. DeMott et al., 2016). Some examples of detailed intercomparisons between different INP measurement methods, including both CFDC and IS, can be found in DeMott et al. (2017) and DeMott et al. (2018). One difference between the CFDC and IS measurements is that due to instrument constraints, the CFDC only measures INPs <2.4 μm (50% aerodynamic diameter cut size), whereas the IS is only limited by inlet transmission, which varies based on the specific set up used. Since the CFDC cannot measure the

largest INPs, it is possible, and has been observed in some cases, that the IS concentrations are higher than the CFDC, but they should still show a similar relationship with wind speed. Neither of these were observed during CHAOS.

The source of aerosol for both INP measurements during CHAOS was seawater, and so INPs sampled by both the IS and CFDC are expected to have similar ice-nucleation properties to each other and the source seawater. Despite the seawater source being the same when the wind speed was varied, the concentration of aerosols increased due to enhanced SSA production, and the CFDC also observed an increase in INP concentration with wind speed. The CFDC was operated under water-supersaturated conditions (RH>100%) during CHAOS, to measure INPs active in the immersion freezing mode. The IS, which requires re-suspension of particles collected on filters in water prior to measurement, also measures INPs active in the immersion freezing mode. Although the measurement technique of the IS is different from that of the CFDC, the drop-freezing method (Vali 1971) of the IS also measures the full INP population present, the same as the CFDC. Since they sampled the same polydisperse aerosol and both measured INPs active in the immersion freezing mode, the IS was also expected to observe an increase in INP concentration with wind speed, the same as the CFDC.

References:

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