

## **General comments:**

In this paper, the authors present their findings on the ice-nucleating particle (INP) characteristics of the Sea Spray Aerosol (SSA), which were generated from a Marine Aerosol Reference Tank (MART). Additionally, the researchers explored the impact of atmospheric aging on these characteristics. Interestingly, they found no observable effect of the atmospheric aging. On the whole, the study is methodologically sound. However, I must express three major concerns as well as a few specific issues related to the content, which I will delve into more deeply below. Despite these concerns, the study remains intriguing and offers valuable contributions to the broader scientific community's understanding of the INP characteristics of SSA. There is no doubt that with necessary revisions, the work will be worthy of publication. Nonetheless, it is imperative to note that major revisions are required to elevate the study to its full potential.

## **Three major issues:**

### **1. The freezing mechanism at temperature $< 220\text{K}$**

The elucidation of the nucleation mechanism in SSA remains a significant and yet unresolved scientific query. The authors of the current study, intriguingly, appear to circumvent direct discussion of the low-temperature nucleation dynamics of SSAs. They opt instead to vaguely encapsulate the complex phenomena using the generic term 'heterogeneous freezing.' The data put forth in this paper, particularly as illustrated in Figure 5, presents a compelling view. It appears to document a transition from homogeneous to heterogeneous nucleation as temperatures descend towards 220 degrees. Nucleation observed under these chillier conditions within a range spanning from water to ice saturation. This behavior should ideally be defined as deposition nucleation, however, Figure 5 sheds light on the temperature interval wherein the pore condensation freezing (PCF) manifests itself. A striking alignment is observed between the nucleation occurring below 215 degrees and the PCF. This concurrence seemingly substantiates the notion that SSA nucleation under colder conditions could indeed be characterized by the PCF.

### **2. The phase state of the SSA**

The main objective was to compare the INP characteristics of the pure and aged SSA, however, the reviewer was concerned the state of particles could influence the results. In this study, the measurement of humidity was performed before the coil cold trap, maintaining a controlled relative humidity at 10% under ambient temperature conditions. However, this level of water vapor pressure can escalate from a few thousand to tens of thousands supersaturation with respect to ice at 220K. Consequently, it is imperative for the authors to consider the dwell time within the coil cold trap and the Continuous Flow Diffusion Chamber (CFDC). Furthermore, it would be beneficial to generate estimations of the phase state prior to its entry into the CFDC.

### **3. The setup of the experiment**

Currently, the sample air directly enters the oxidation flow reactor after exiting from the MART instrument. It is suggested that the sample air should be dehumidified before

passing through the oxidation flow reactor. This is because, during liquid-phase oxidation, the crystallization of SSA solution droplets into crystals may not affect its surface structure. However, if oxidation occurs in the solid phase, the pores on the surface of the SSA particles might be filled, thereby affecting its ice nucleation properties.

### **Specific comments:**

P1, Line 16: I suggest use the supersaturation with respect to ice instead of RH to evaluate the onset of the ice nucleating forms.

P3, Sec 2.1: The artificial seawater was filter through a TOC+HEPA filter to remove the insoluble particles, what about the natural seawater? There might be some dust and biological particles inside the natural seawater which have influence on the INP measurement.

P5, Line 151-152: I was wondering why there were so many particles during the “blank experiment” with DI water. Does that mean there were contamination of the MART and sampling tubes? Thus, I strongly suggest do “blank experiment” before and after each experiment.

P5, Line 155: TSI models 3080, remove the 3081 and 3010, which is the model of DMA and CPC.

P5, Line 160: This paragraph is confusing and very hard to understanding.

Line 195-295, when discussing the influence of organics, some studies (Ignatius, et al., 2016; Knopf et al., 2018; Tian et al., 2022) found organic aerosol (likely secondary) could be glassy (Koop et al., 2011) and efficient heterogeneous ice nuclei under the condition of low RH, which could be referenced to support the point that organics itself may serve as INP.

P7, Sec2.4: The IS show the mixed phase regime (-38–0 °C) INP concentration which inconsistent with the main theme of this study, and the results was shown in the supplement. The author need to carefully consider whether to retain this section of content.

### **References:**

- Koop, T., Bookhold, J., Shiraiwa, M., Pöschl, U. Glass transition and phase state of organic compounds: dependency on molecular properties and implications for secondary organic aerosols in the atmosphere. *Phys. Chem. Chem. Phys.*, 2011, 13, 19238-19255.
- Knopf, Daniel A., Alpert, Peter A., Wang, B. B. The role of organic aerosol in atmospheric ice nucleationL: A review. *ACS Earth Space Chem*, 2018, 2, 168-202. DOI: 10.1021/acsearthspecechem.7b00120.
- Tian, P., Liu, D. T. Bi, K. et al. Evidence for anthropogenic organic aerosols contributing to ice nucleation. *Geophysical Research Letters*, 49, e2022GL099990.

<https://doi.org/10.1029/2022GL099990>.

Ignatius, K., Kristensen, T. B., Jarvinen, E., et al. Heterogeneous ice nucleation of viscous secondary organic aerosol produced from ozonolysis of  $\alpha$ -pinene. *Atmos. Chem. Phys.*, 2016, 16, 6495-6509.