



*Supplement of*

**Physicochemical characterization of free troposphere and marine boundary layer ice-nucleating particles collected by aircraft in the eastern North Atlantic**

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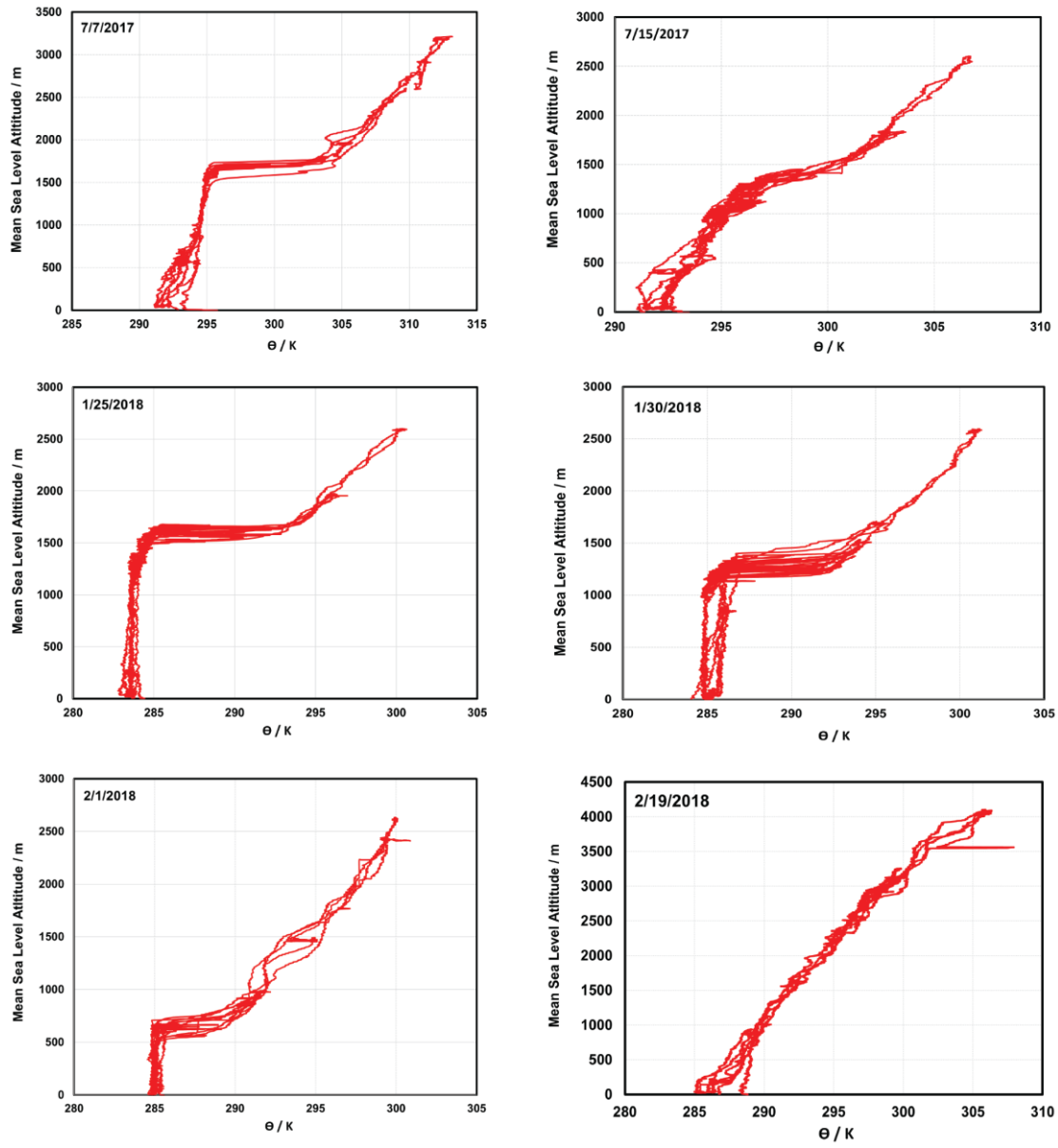
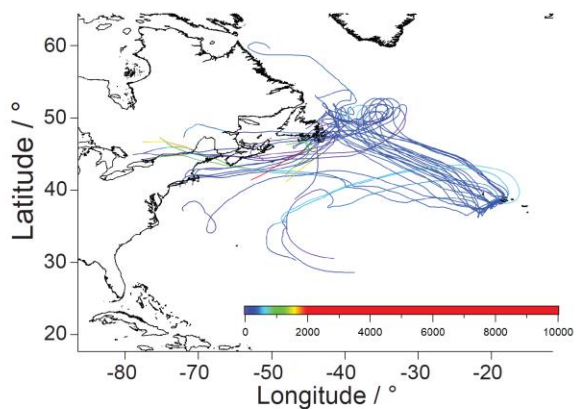
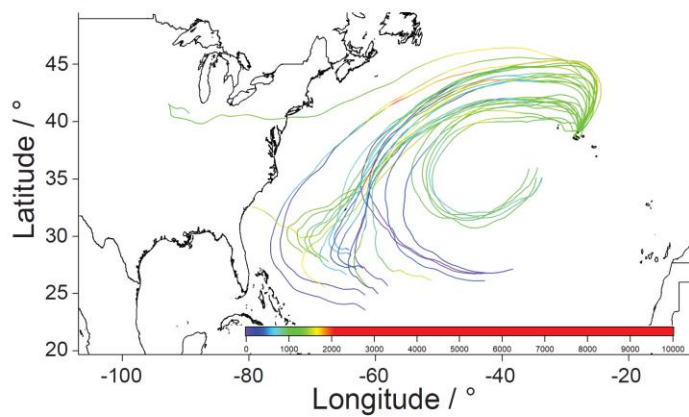


Figure S1: Flight altitude versus potential temperature ( $\Theta$ ) for each flight date as given in Table 1.

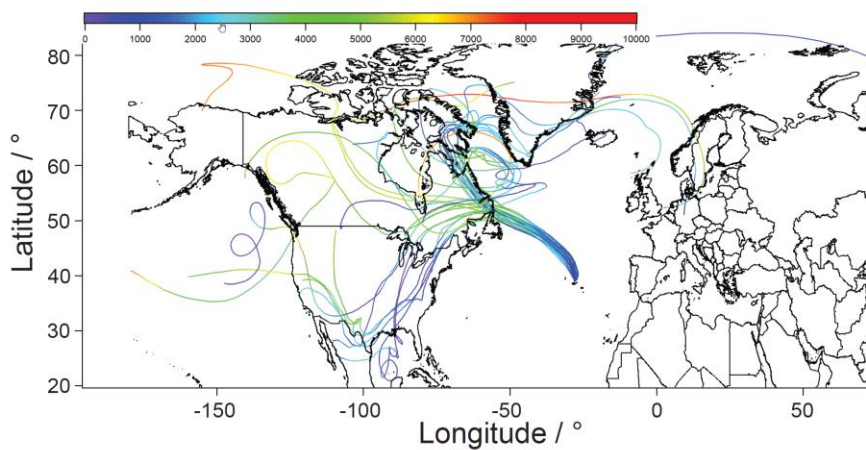
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7/15/2017 S-FT1

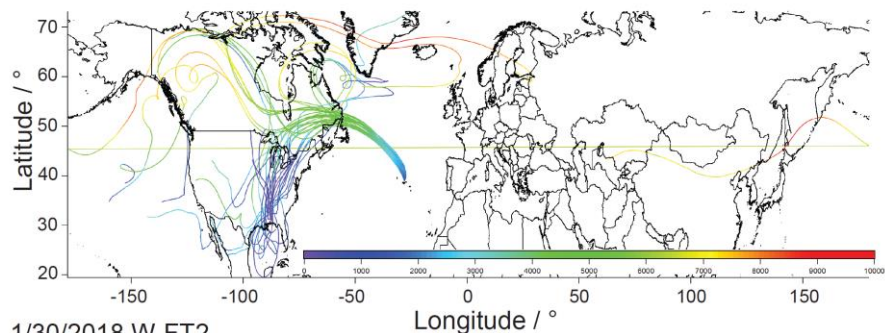


1/25/2018 W-MBL1-DI

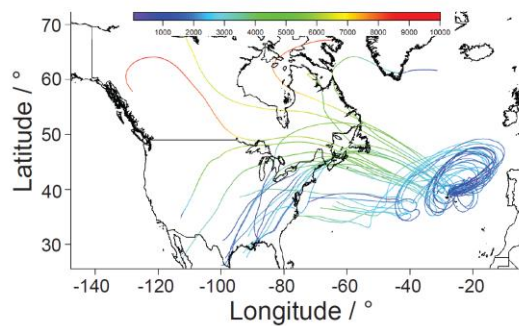


**Figure S2: Representative 10-day backward trajectories for examined airborne-collected marine boundary layer (MBL) and free troposphere (FT) particle samples starting at given sampling altitude. The trajectories are colored by their altitude (m).**

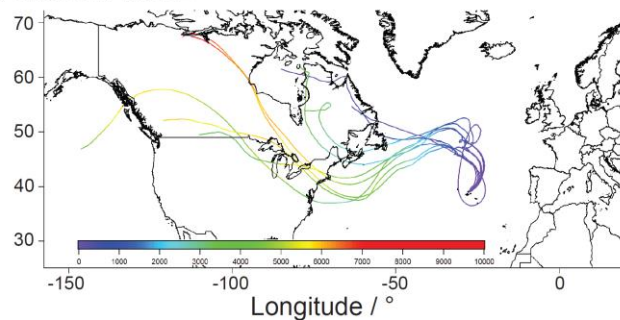
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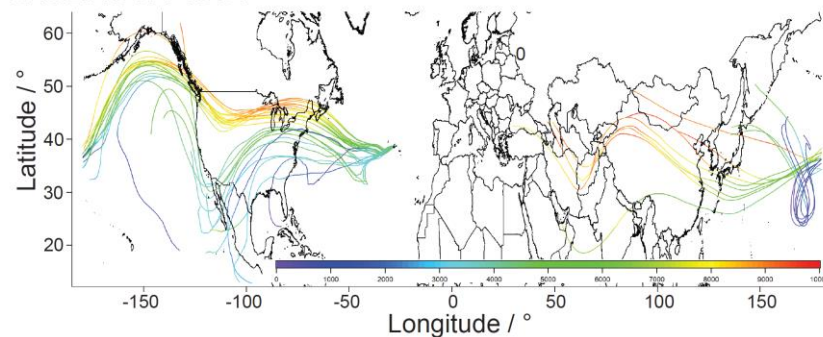
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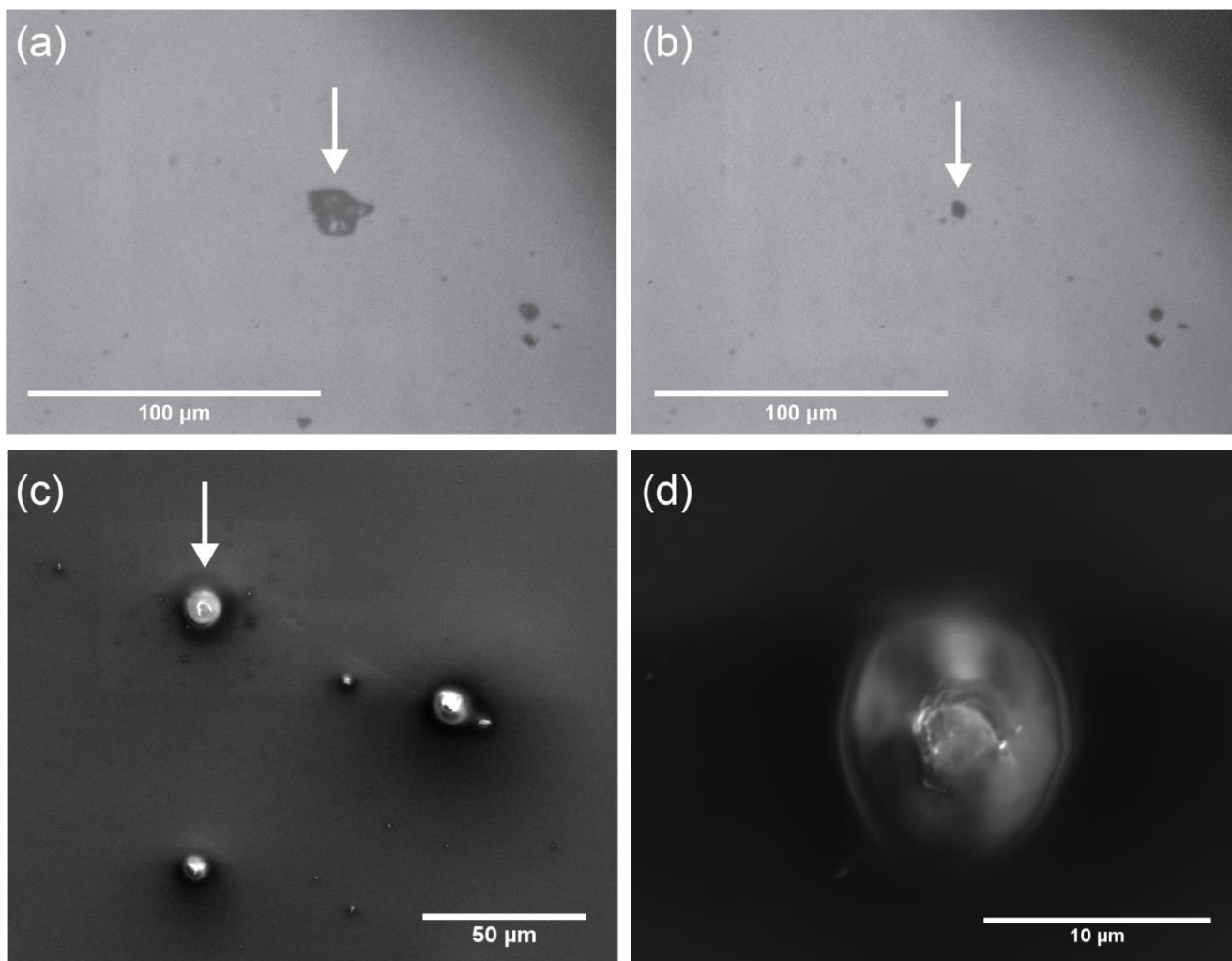
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2/19/2018 W-FT3-DI

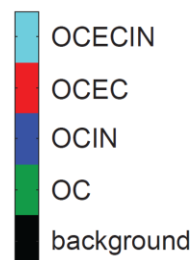
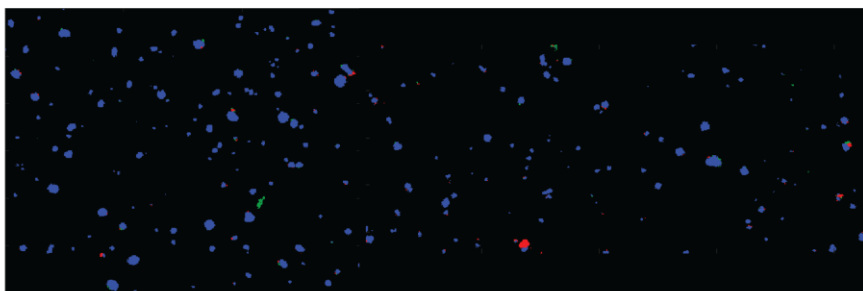


**Figure S3: Representative 10-day backward trajectories for examined airborne-collected marine boundary layer (MBL) and free troposphere (FT) particles starting at given sampling altitude. The trajectories are colored by their altitude (m).**

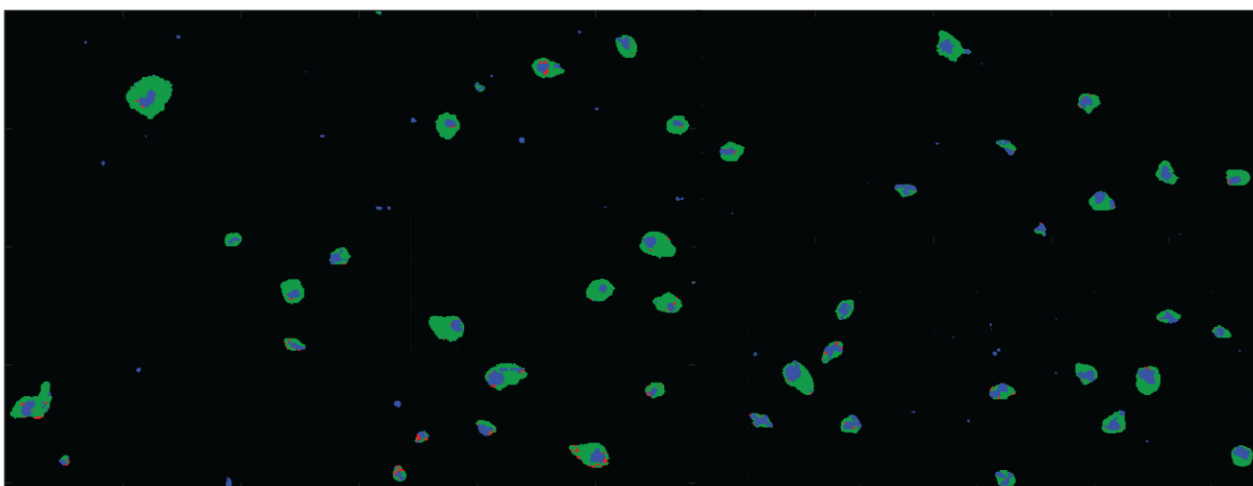


**Figure S4:** Procedure to identify the ice nucleating particle (INP) from optical microscope images (panels a and b) in scanning electron microscopy (panels c and d). Panel (b) shows the residual particles after ice crystal sublimation. White arrow indicates the INP.

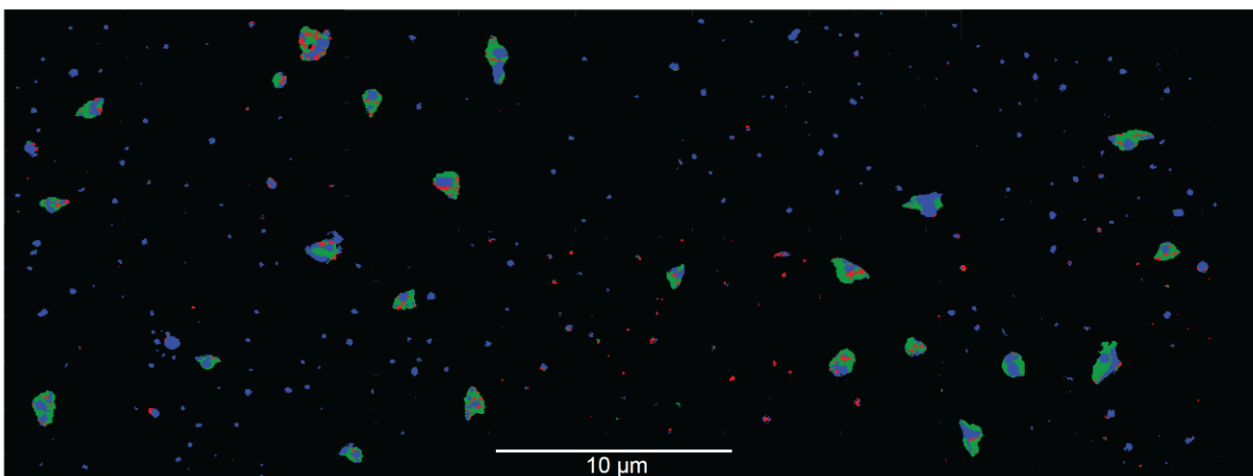
07/07/17 S-MBL1



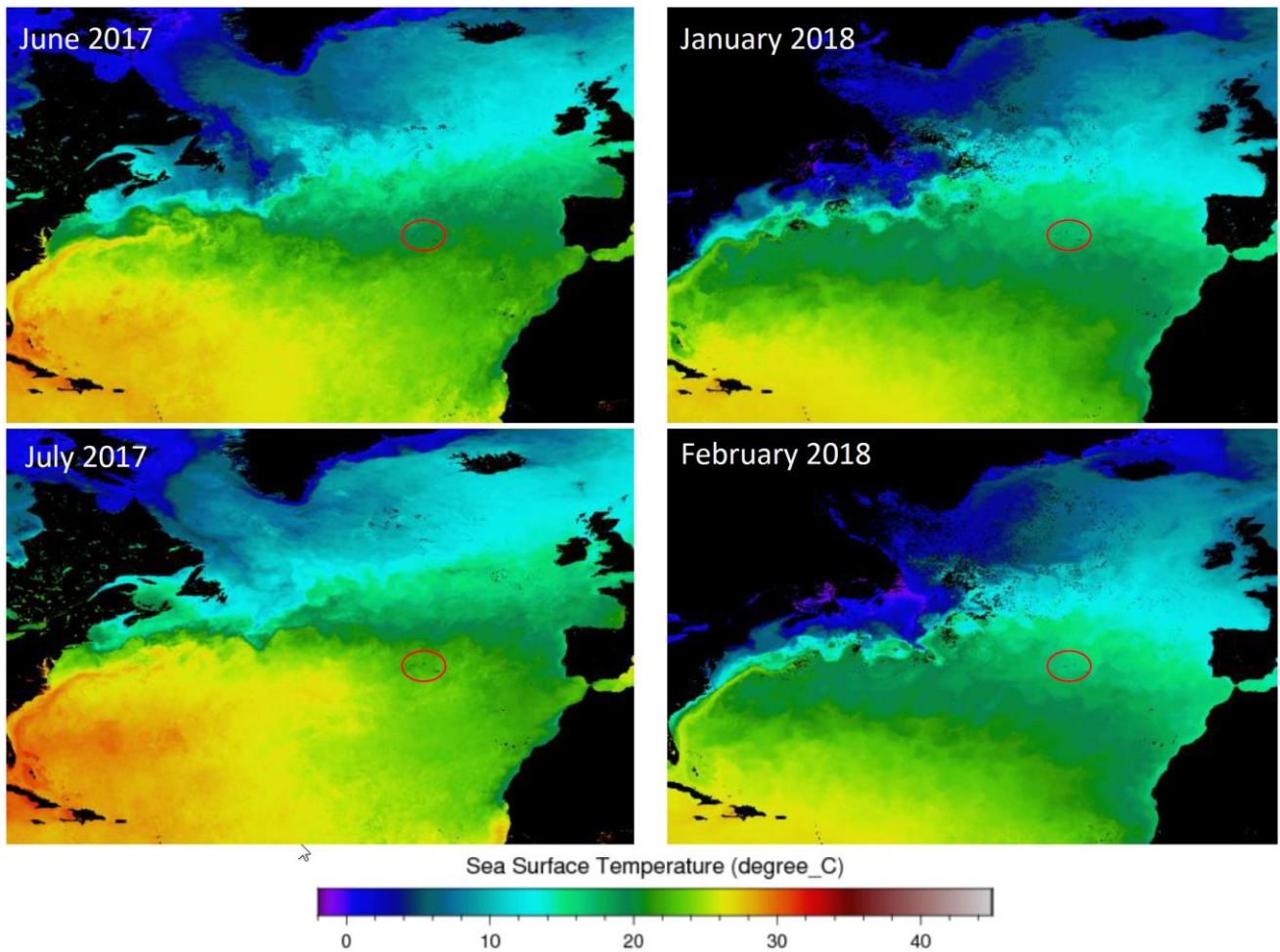
01/25/18 W-MBL1-DI



02/01/18 W-MBL2



**Figure S5: Representative particle mixing state analysis of particle samples collected in the marine boundary layer (MBL) where IN - inorganic, EC - elemental carbon, and OC - organic carbonaceous. The scale bar holds for all panels.**



**Figure S6: Monthly mean sea surface temperature derived by Aqua MODIS (Moderate Resolution Imaging Spectroradiometer) during daytime at 11  $\mu\text{m}$  and 4 km resolution (NASA Goddard Space Flight Center, 2019). Red circles indicate location of Azores Islands.**

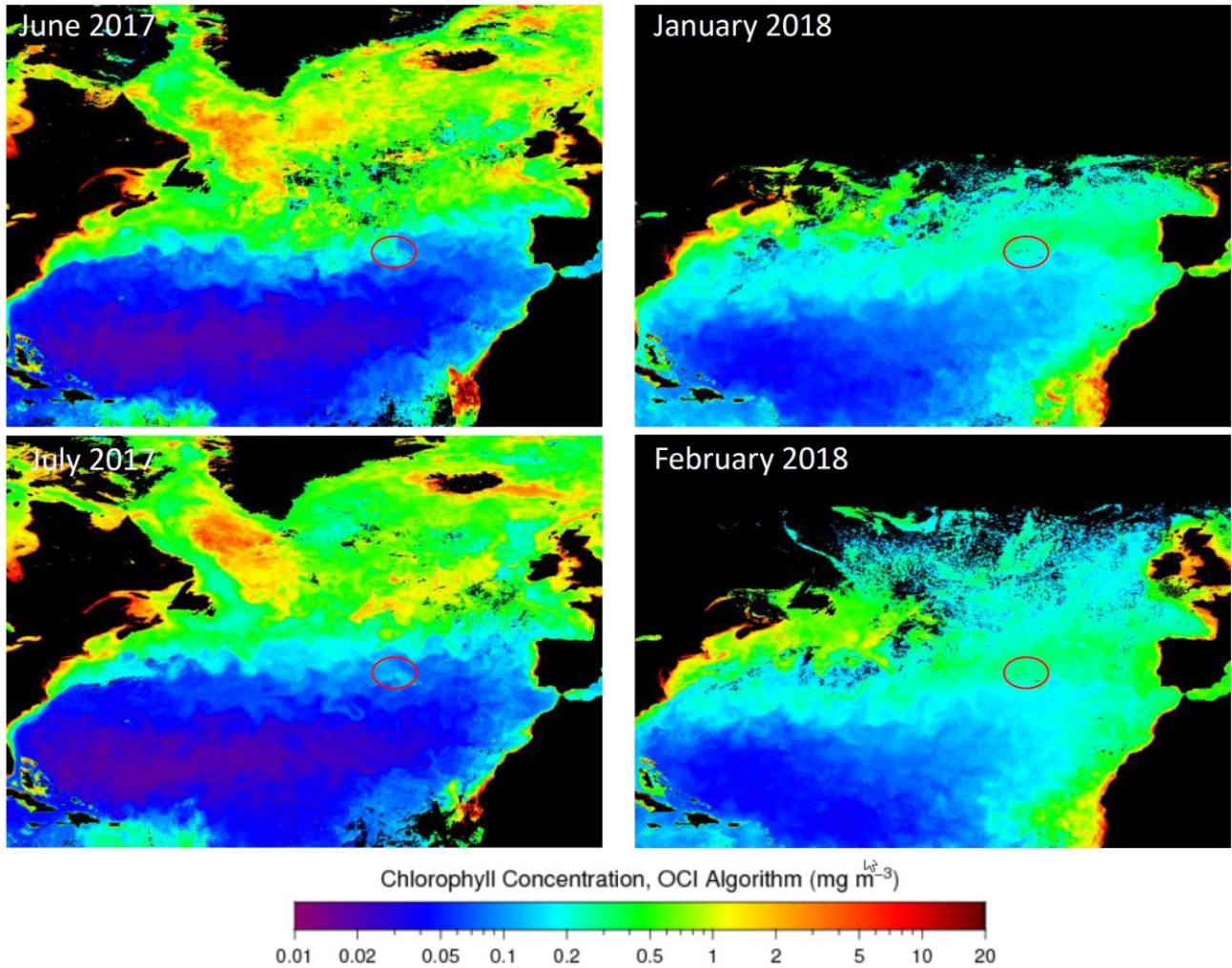
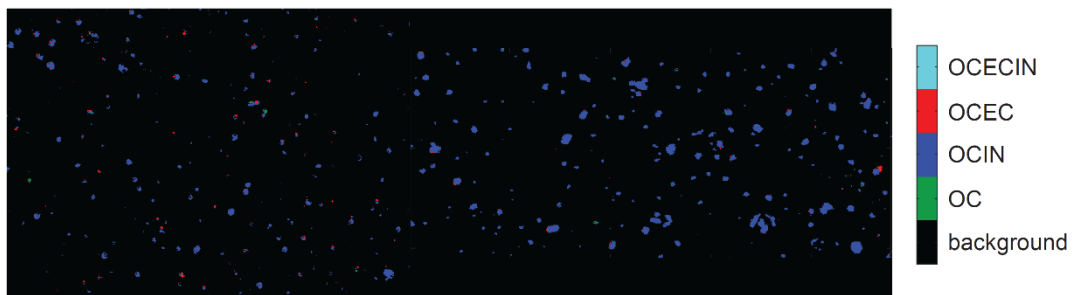


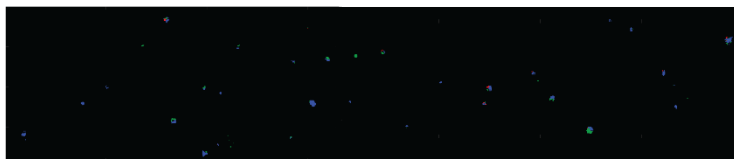
Figure S7: Monthly mean chlorophyll a concentration derived by Aqua MODIS (Moderate Resolution Imaging Spectroradiometer) at 4 km resolution (NASA Goddard Space Flight Center, 2022). Red circles indicate location of Azores Islands.



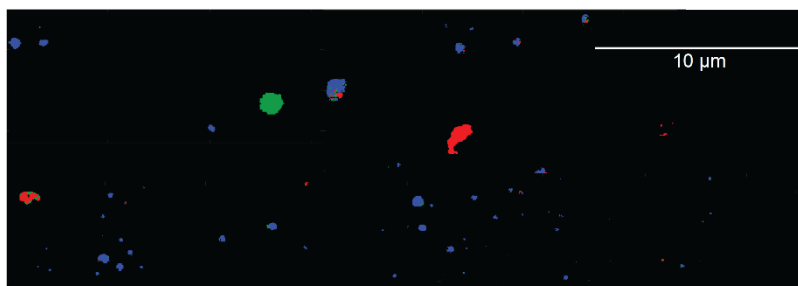
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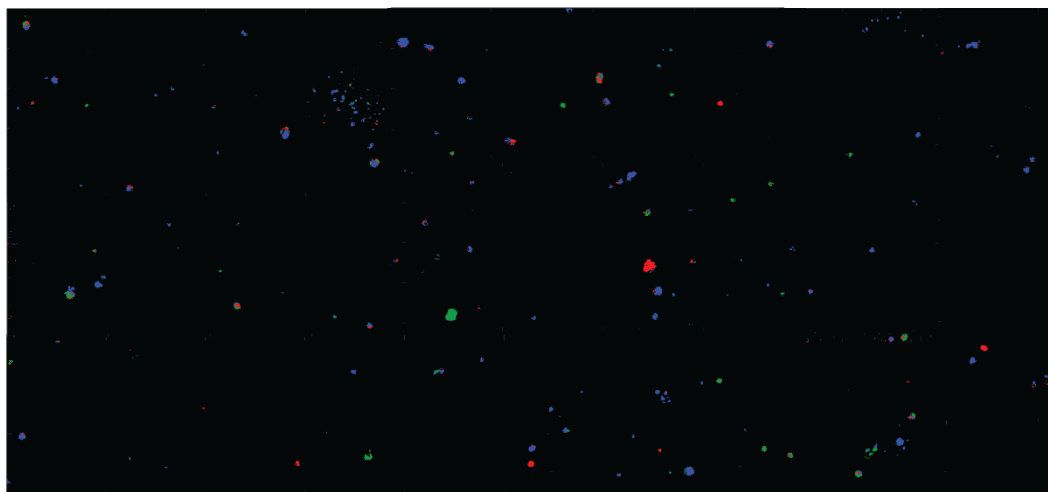
01/25/18 W-FT1-DI



01/30/18 W-FT2

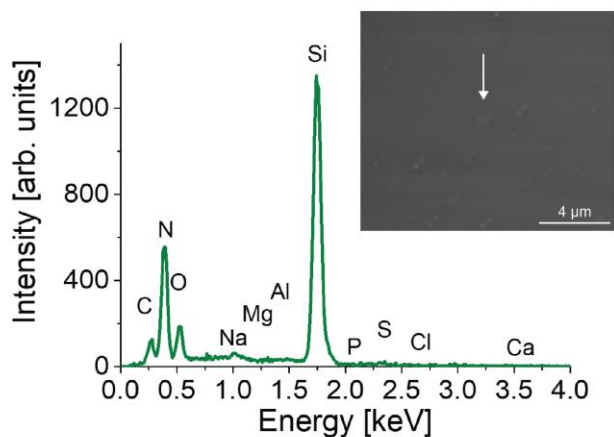


02/19/18 W-FT3-DI

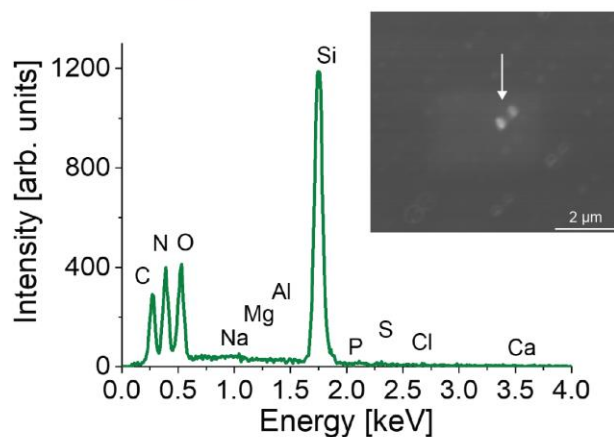


**Figure S8: Representative particle mixing state analysis of particle samples collected in the free troposphere (FT) where IN - inorganic, EC - elemental carbon, and OC - organic carbonaceous. The scale bar holds for all panels.**

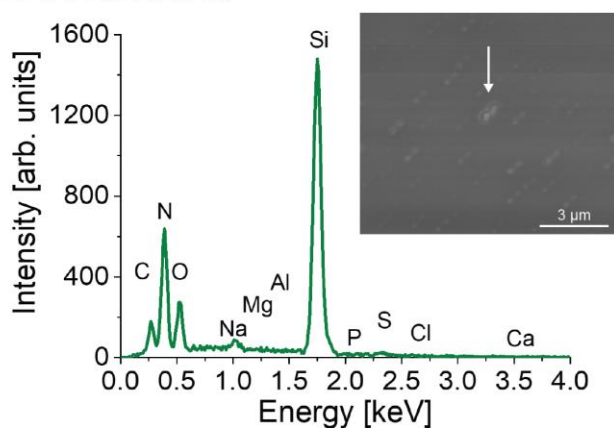
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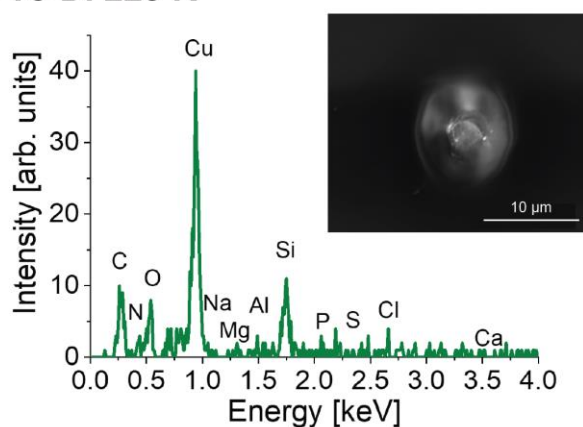
W-FT1-DI 223 K



W-FT1-DI 223 K



W-FT3-DI 223 K



W-FT3-DI 223 K

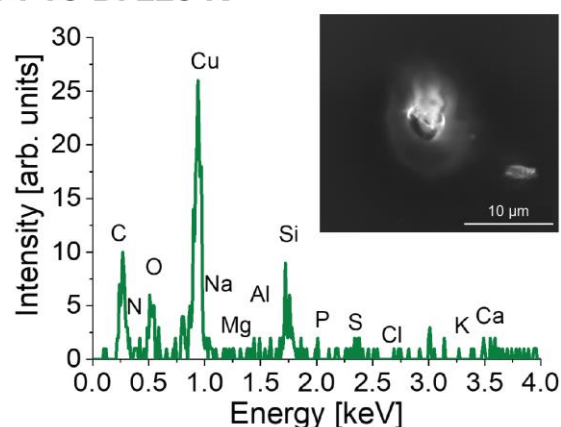
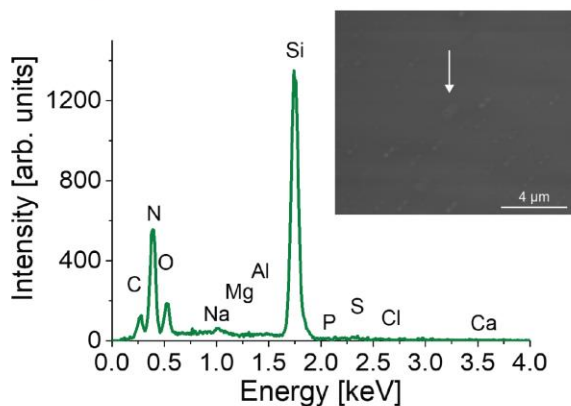
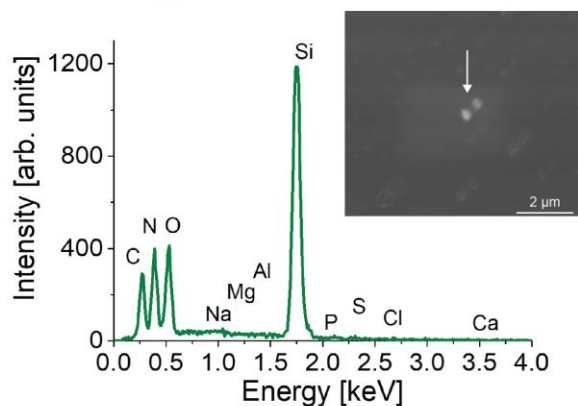


Figure S9: EDX spectra of identified INPs from particles samples collected during winter under free troposphere (FT) conditions with corresponding SEM obtained particle image. Si and N signals stem from the substrate ( $\text{Si}_3\text{N}_4$  coated silicon wafer chips) and the chamber/holder (Al).

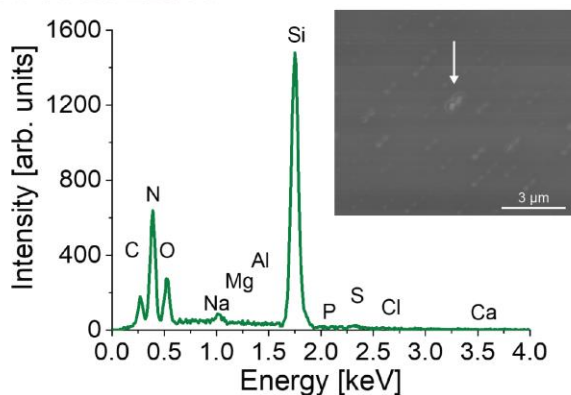
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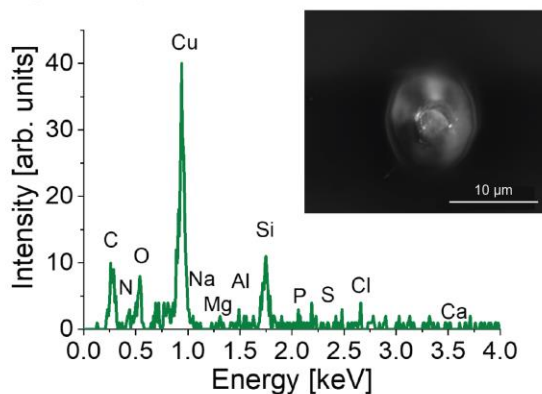
W-FT1-DI 223 K



W-FT1-DI 223 K



W-FT3-DI 223 K



W-FT3-DI 223 K

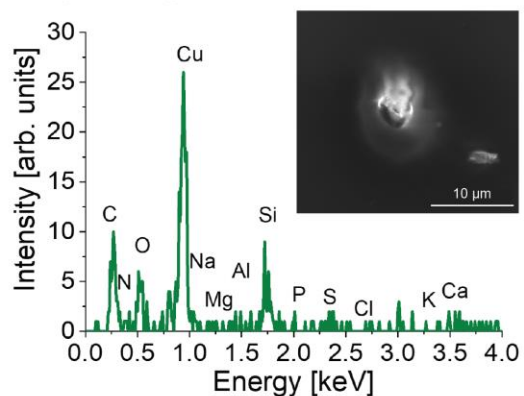


Figure S10: EDX spectra of identified INPs from particles samples collected during winter under free troposphere (FT) conditions with corresponding SEM obtained particle image. Si and N signals stem from the substrate ( $\text{Si}_3\text{N}_4$  coated silicon wafer chips) and the chamber/holder (Al).

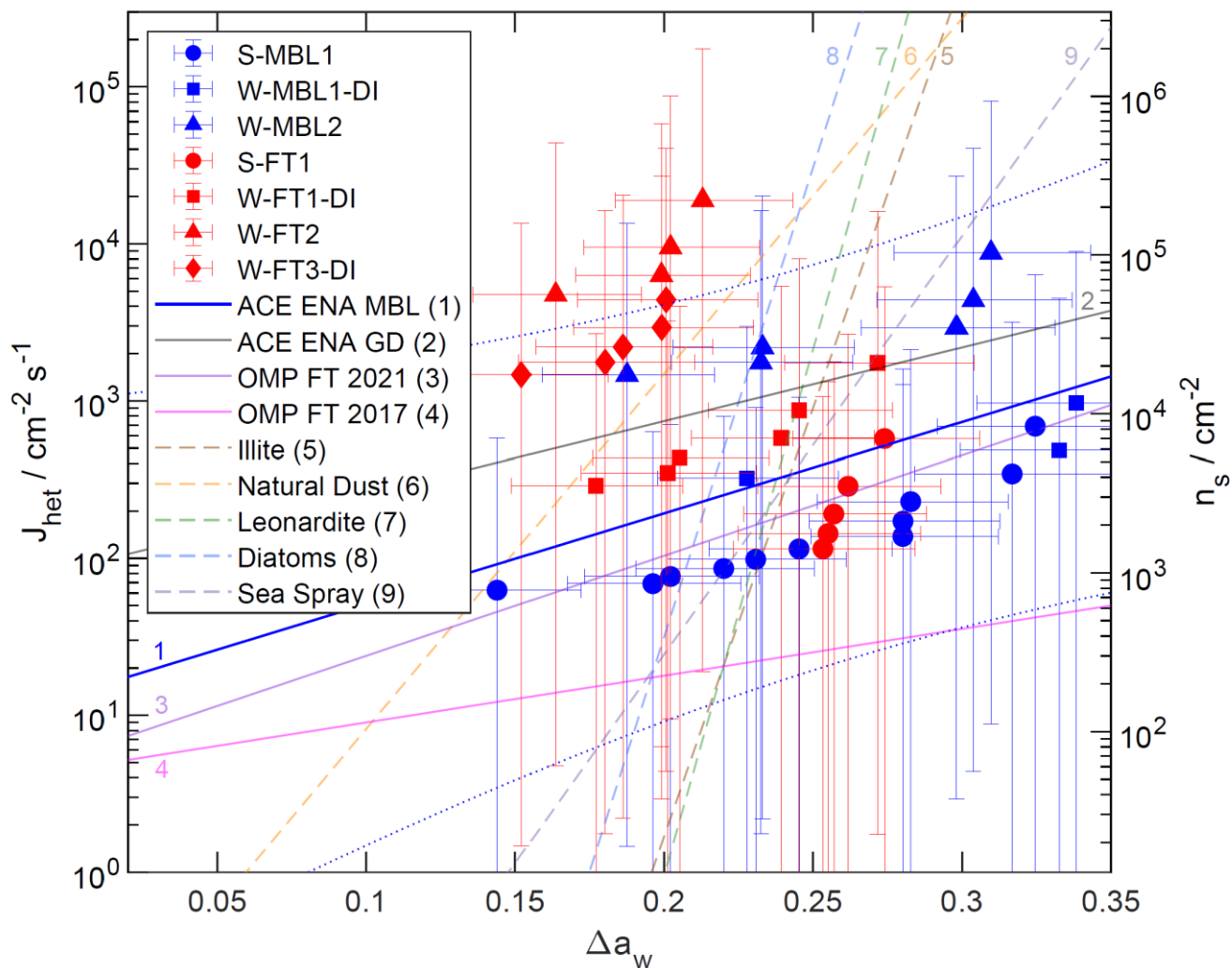
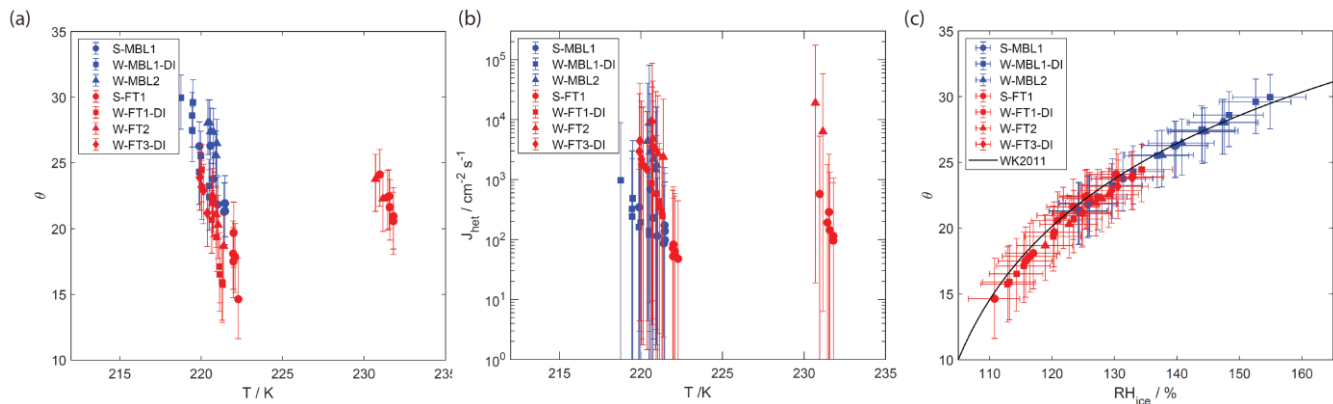


Figure S11: Immersion freezing (IMF) data of examined ACE-ENA particle samples (solid symbols) and of previous studies (colored lines) as given in legend. Heterogeneous ice nucleation rate coefficients ( $J_{\text{het}}$ ) and ice nucleation active sites (INAs) density ( $n_s$ ) are presented as a function of the water activity criterion  $\Delta a_w$ . Error bars include uncertainties in temperature, humidity, and surface area. Blue solid line represents a linear regression to the newly derived MBL IMF data. Solid black, magenta, and purple lines represent  $J_{\text{het}}$  and  $n_s$  IMF derived from accompanying ACE-ENA ground site INP measurements (ACE-ENA GD), from the Observatory of Mountain Pico (OMP) measurements under free tropospheric (FT) conditions in the Azores on a neighboring island (PMO FT 2017, 2021; (China et al., 2017; Lata et al., 2021)). Please note that only  $J_{\text{het}}$  was reported for OMP FT 2021 (Lata et al., 2021). Water activity-based IMF  $J_{\text{het}}$  and  $n_s$  for other INP types are given as dashed colored lines as indicated in legend.



**Figure S12: Deposition ice nucleation (DIN) data of examined ACE-ENA MBL and FT particle samples in blue and red colors, respectively. (a) Heterogeneous ice nucleation rate coefficients ( $J_{het}$ ) as a function of temperature. (b) Contact angles ( $\theta$ ) corresponding to  $J_{het}$  values shown in (a). (c)  $\theta$  values for relative humidity with respect to ice ( $RH_{ice}$ ) under which DIN was observed. Solid line represents the DIN parameterization by Wang and Knopf (2011).**

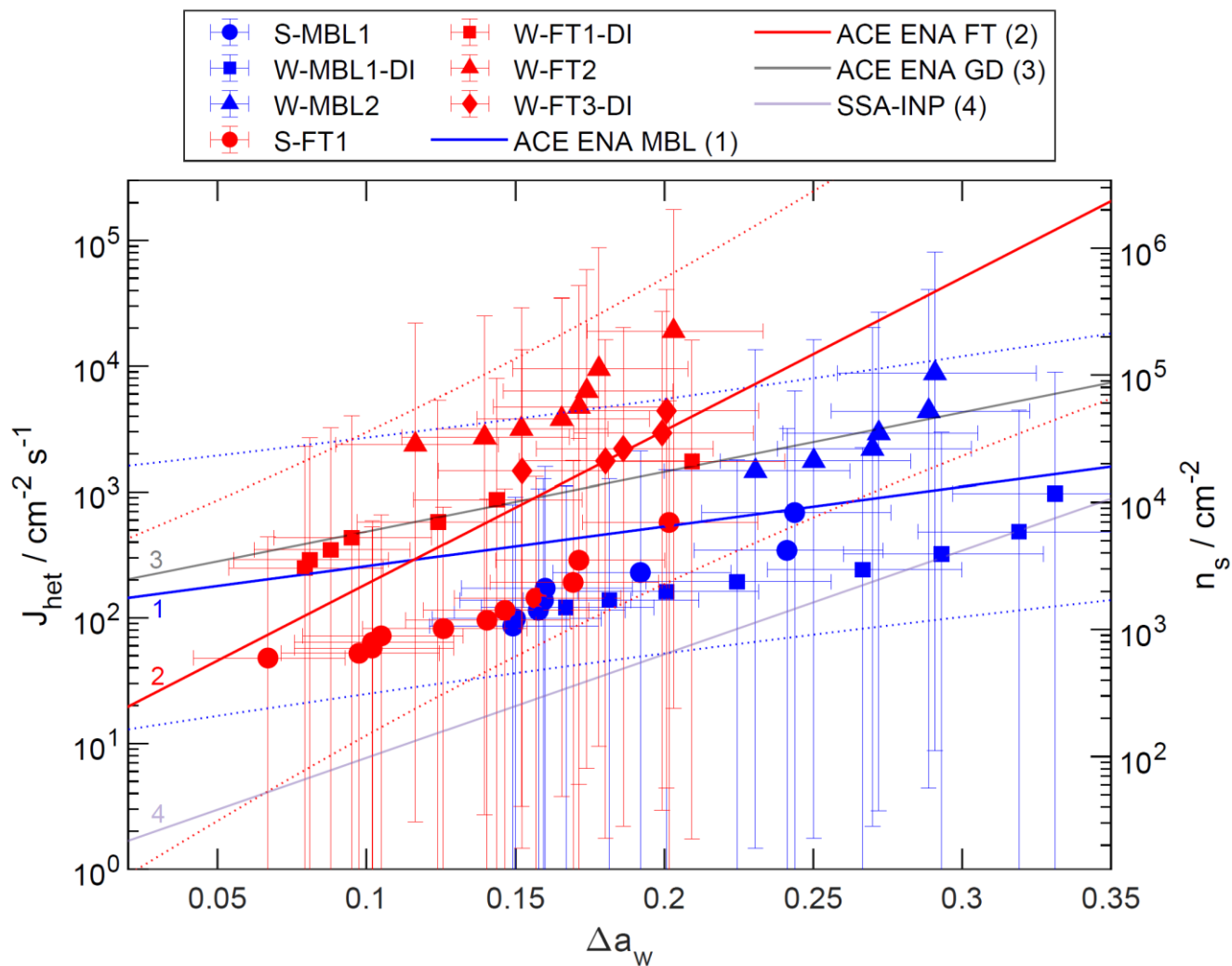


Figure S13: Deposition ice nucleation (DIN) data of examined ACE-ENA MBL and FT particle samples (blue and red symbols, respectively). Heterogeneous ice nucleation rate coefficients ( $J_{\text{het}}$ ) and ice nucleation active sites (INAS) density ( $n_s$ ) are presented. Error bars included uncertainties in temperature, humidity, and surface area. Blue and red solid and dotted black lines represent linear regression fits and associated fit uncertainties for MBL and FT particle samples, respectively. Black line represents the DIN ABIFM parameterization derived from ground site ACE-ENA INP measurements. Purple line represents the water-activity based DIN sea spray aerosol parameterization from Alpert et al. (2022).

## References

NASA Goddard Space Flight Center, Ocean Ecology Laboratory, Ocean Biology Processing Group. Moderate-resolution Imaging Spectroradiometer (MODIS) Aqua Global Level 3 Mapped SST. Ver. 2019.0. PO.DAAC, CA, USA. Dataset accessed [2023-03-21] at <https://doi.org/10.5067/MODSA-MO4D9>.

NASA Goddard Space Flight Center, Ocean Ecology Laboratory, Ocean Biology Processing Group. Moderate-resolution Imaging Spectroradiometer (MODIS) Aqua Chlorophyll Data; 2022 Reprocessing. NASA OB.DAAC, Greenbelt, MD, USA. doi: 10.5067/AQUA/MODIS/L3B/CHL/2022. Accessed on 03/21/2023