Supplement of

Micro-spectroscopic and freezing characterization of ice-nucleating particles collected in the marine boundary layer in the Eastern North Atlantic

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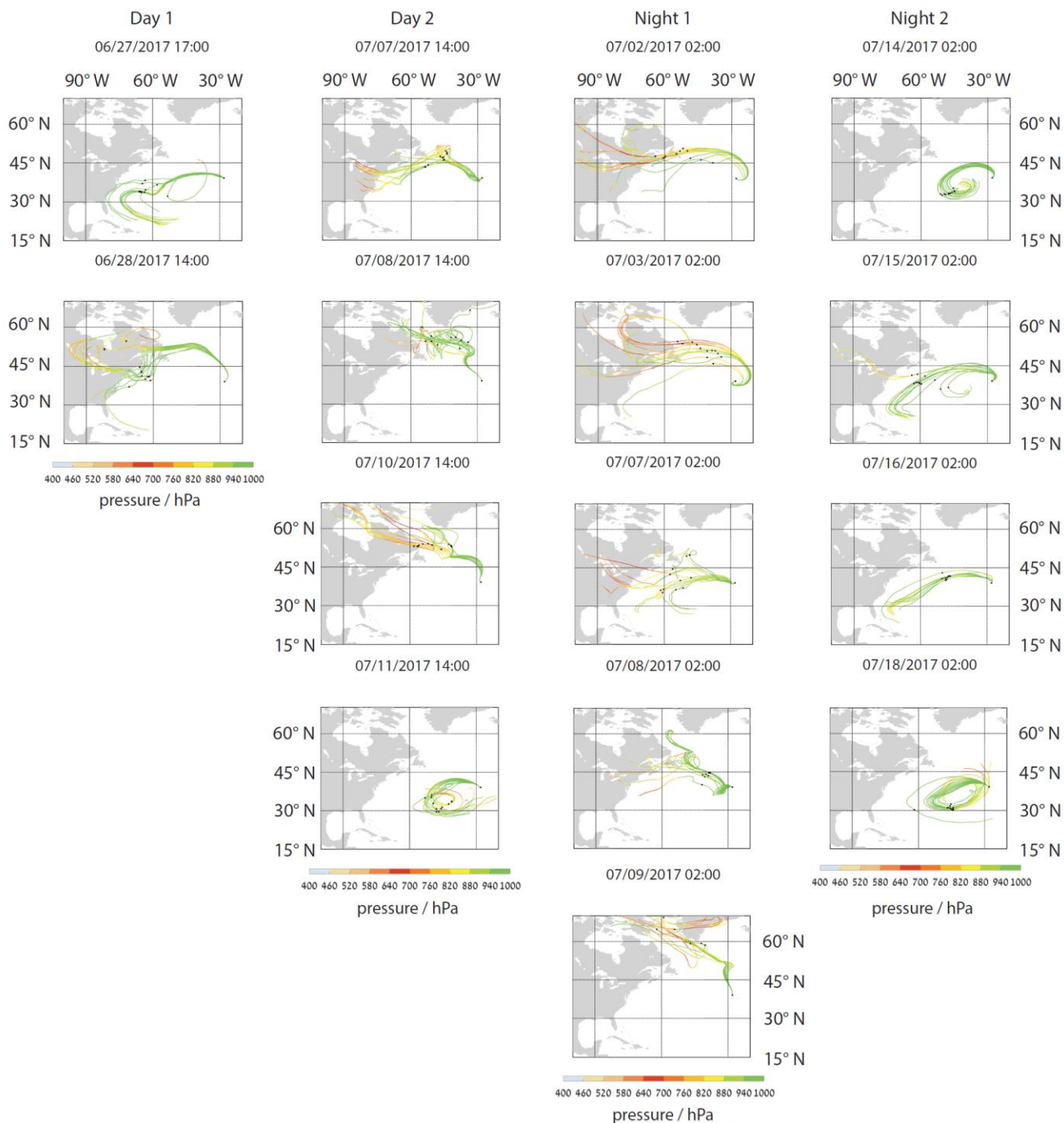
This Supplement comprises of six figures (S1-S6).

Figure S1: Sampling location on Graciosa Island, Azores, during the Aerosol and Cloud Experiments in the Eastern North Atlantic (ACE-ENA) field campaign (Wood, 2019) and image of ground sampling site (Wang et al., 2021).

Figure S2: Representative backward trajectories for examined daytime and nighttime samples for corresponding sampling periods. The date in the panel title marks the time from which the 10-day backward trajectories are calculated (black dot), and the additional black dots mark the trajectories' locations 5 days prior to their arrival at the sampling site. The trajectories are colored by their pressure level (hPa).

45

Ice formation monitored by optical microscope
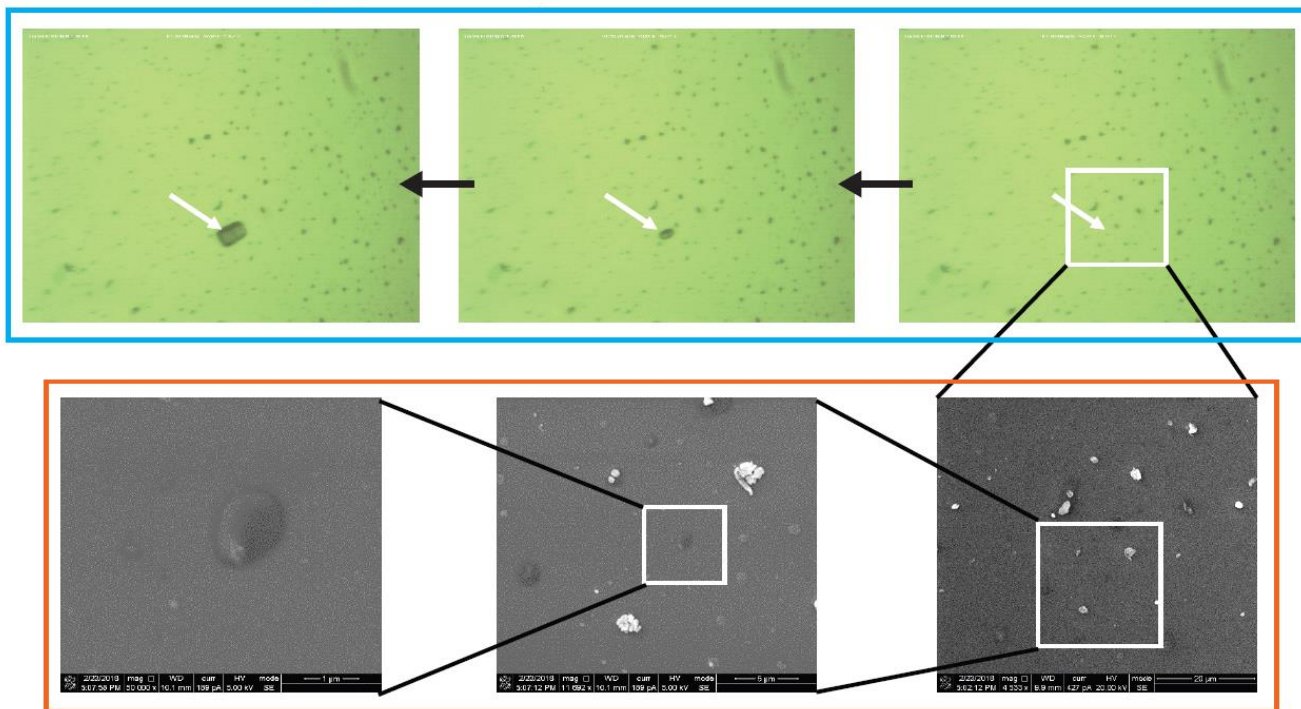
Ice-nucleating particle identification by scanning electron microscope

Figure S3: Process of ice-nucleating particle (INP) identification using multi-model instrument approach following Knopf et al. (2014).

4

Day 1

	OCECIN
	OCEC
	OCIN
	OC
	background

Day 2

	OCECIN
	OCEC
	OCIN
	OC
	background

Night 1

	OCECIN
	OCEC
	OCIN
	OC
	background

Night 2
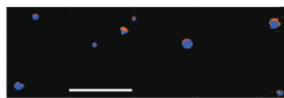

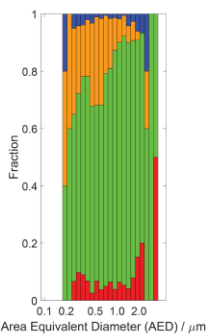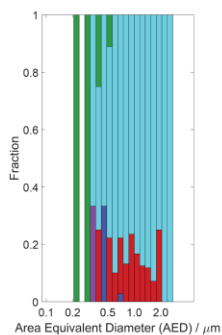|	OCECIN
	OCEC
	OCIN
	OC
	background
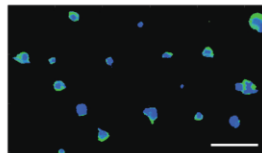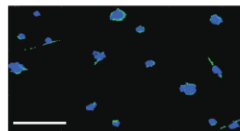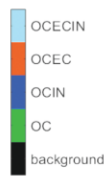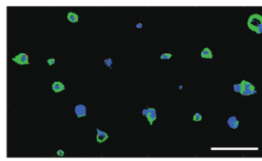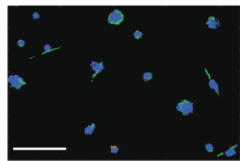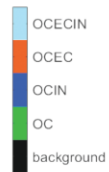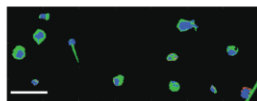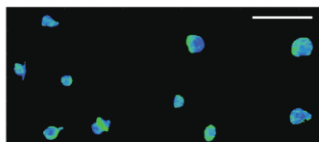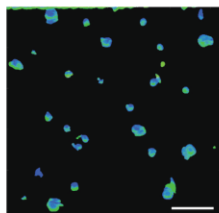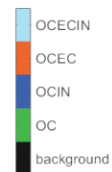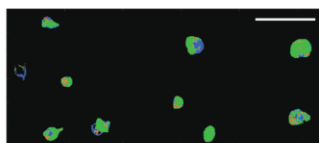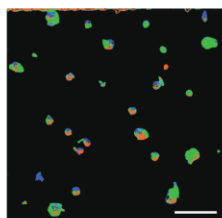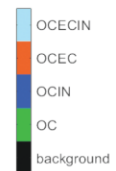
Figure S4: Representative false color particle maps derived by STXM/NEXAFS for the daytime and nighttime samples. The upper panels for each sample provide particle population mixing state where IN - inorganic, EC - elemental carbon, OC - organic carbon. The corresponding lower panels indicate the range in organic volume fraction (OVF) per particle, where organic dominated particles are in green (organic mass is > 80%) and inorganic dominated particles are blue (inorganic mass is > 80%). Scale bar indicates 5 µm.
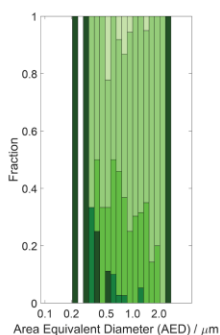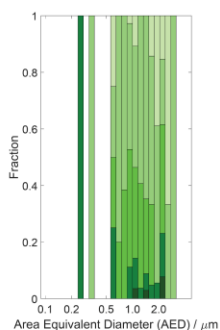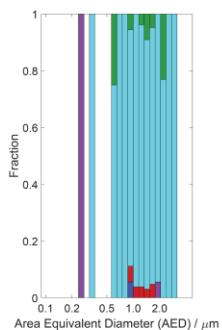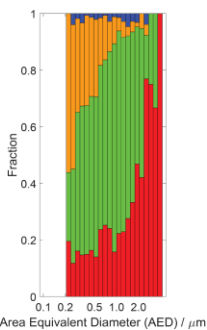
Day 1   SEM - Cluster   STXM - Mixing State   STXM - Organic Volume F
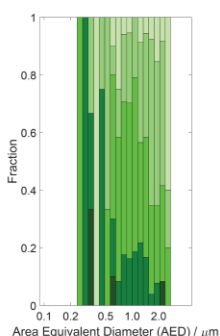
Day 2

Night 1
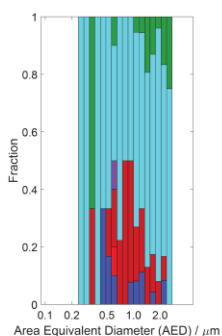
Night 2

Legend:
SS$_{proc}$/dust/org   SS$_{proc}$/dust
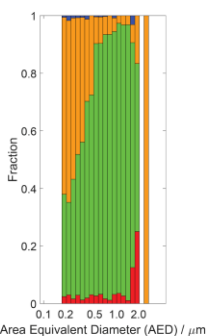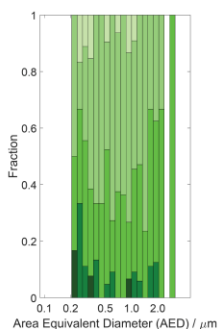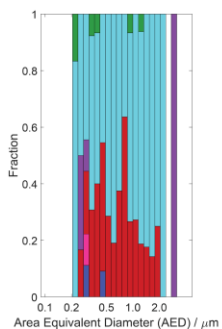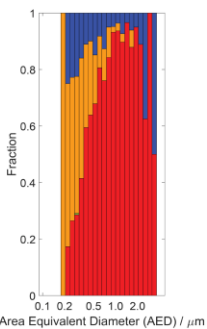Sea salt   Organic/Cl

IN   OCEC
ECIN   OCIN
EC   OC
OCECIN

80-100%   20-40%
60-80%   <20%
40-60%

Axis labels (repeated per panel): Fraction (y-axis); Area Equivalent Diameter (AED) / μm (x-axis); 0.1, 0.2, 0.5, 1.0, 2.0

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Figure S5: Size-resolved fractional particle composition for particle samples determined by micro-spectroscopic single-particle analysis as a function of area equivalent diameter (AED). From left to right: first column: CCSEM/EDX derived particle-type classes as 'processed sea salt with mineral dust and organic matter', 'sea salt', 'processed sea salt with mineral dust' and 'organic matter with chlorine'. Second column: STXM/NEXAFS derived particle mixing state: IN - inorganic, EC - elemental carbon, OC - organic carbon. Third column: organic volume fraction (OVF).
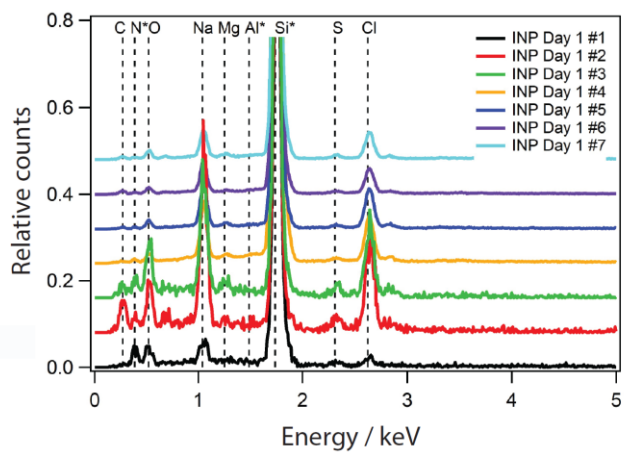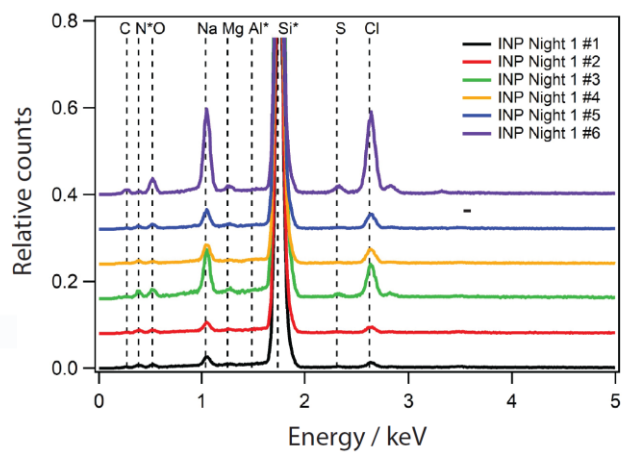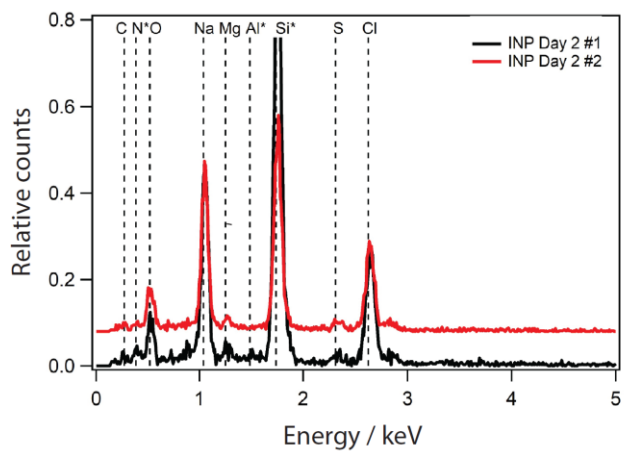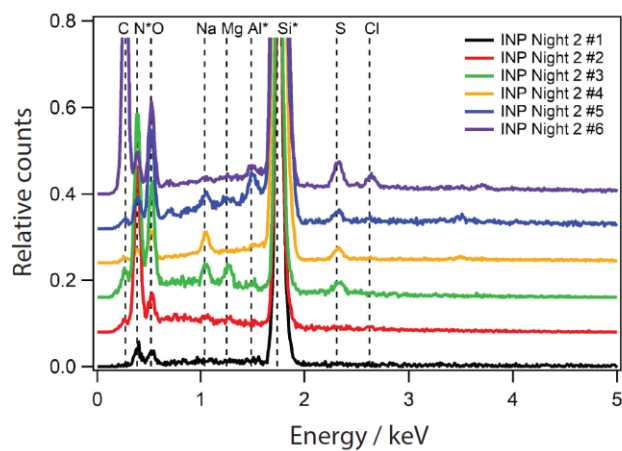
85

90

95

100

105

Figure S6: EDX spectra of identified INPs present in daytime and nighttime samples given in Fig. 6 and Table 3. *corresponds to the signal from the substrate (Si_3N_4 coated silicon wafer chips) and the chamber/holder (Al).

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References

Knopf, D. A., Alpert, P. A., Wang, B., O'Brien, R. E., Kelly, S. T., Laskin, A., Gilles, M. K., and Moffet, R. C.: Microspectroscopic imaging and characterization of individually identified ice nucleating particles from a case field study, J. Geophys. Res., 119, 10365-10381, 10.1002/2014JD021866, 2014.

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Wood, R.: Eastern North Atlantic, DOE/SC-ARM-14-006, 2019.