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Ice-Nucleating Particles Near Two Major Dust Source Regions

Charlotte M. Beall[1,2], Thomas C. J. Hill[3], Paul J. DeMott[3], Tobias Köneman[2*], Michael Pikridas[4], Frank Drewnick[5], Hartwig Harder[6], Christopher Pöhlker[2], Jos Lelieveld[4,6], Bettina Weber[2**], Minas Iakovides[4], Roman Prokeš[7,8], Jean Sciare[4], Meinrat O. Andreae[1,2,9], M. Dale Stokes[1], Kimberly A. Prather[1,10]

[1]Scripps Institution of Oceanography, University of California San Diego, La Jolla, CA 92037, USA

[2]Max Planck Institute for Chemistry, Multiphase Chemistry and Biogeochemistry Departments, D-55128 Mainz, Germany

[3]Department of Atmospheric Science, Colorado State University, Fort Collins, CO 80523, USA

[4]Climate & Atmosphere Research Center, The Cyprus Institute, Nicosia, CY-1645, Cyprus

[5] Max Planck Institute for Chemistry, Particle Chemistry Department, D-55128 Mainz, Germany

[6]Max Planck Institute for Chemistry, Atmospheric Chemistry Department, D-55128 Mainz, Germany

[7]RECETOX, Faculty of Science, Masaryk University, Kotlarska 2, 611 Brno, Czech Republic

[8]Department of Atmospheric Matter Fluxes and Long-range Transport, Global Change Research Institute of the Czech Academy of Sciences, Belidla 4a, 60300, Brno, Czech Republic

[9]Department of Geology and Geophysics, King Saud University, Riyadh, Saudi Arabia

[10]Department of Chemistry and Biochemistry, University of California San Diego, La Jolla, CA, 92093 USA

*Now at Envicontrol GmbH, Waidmarkt 11, 50676 Köln, Germany

**Now at: Institute of Biology, University of Graz, 8010 Graz, Austria

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Correspondence to: Charlotte M. Beall, cbeall@ucsd.edu

Abstract

Mineral dust and sea spray aerosol represent important sources of ice nucleating particles (INPs), the minor fraction of aerosol particles able to trigger cloud ice crystal formation and, consequently, influence multiple climate-relevant cloud properties including lifetime, reflectivity, and precipitation efficiency. Mineral dust is considered the dominant INP source in many parts of the world due to its ice nucleation efficiency and its sheer abundance, with global emission rates of up to 4700 Tg a^{-1}. However, INPs emitted from the ocean surface in sea spray aerosol frequently dominate INP populations in remote marine environments, including parts of the Southern Ocean where cloud-resolving model simulations have demonstrated that cloud reflectivity is likely strongly controlled by INPs. Here we report INP concentrations measured in aerosol and seawater samples during Air Quality and Climate Change in the Arabian BAsin (AQABA), a shipborne campaign that spanned the Red Sea, Gulf of Aden, Arabian Sea, Arabian Gulf, and part of the Mediterranean. In aerosol samples collected within a few hundred kilometers of the first and second ranked sources of dust globally, the Sahara and Arabian Peninsula, INP concentrations ranged from 0.2 to 11 L^{-1} at -20 °C with observed ice nucleation site densities (n_s) 1-3 orders of magnitude below levels predicted by mineral dust INP parameterizations. Over half of the samples (at least 14 of 26) were collected during dust storms with average dust mass concentrations between 150 and 490 µg m^{-3} (PM_{10}). The impacts of heat and peroxide treatments indicate that organics were responsible for the observed ice nucleation (IN) -activity at temperatures ≥ -15 °C with proteinaceous (heat-labile) INPs frequently observed at higher freezing temperatures > -10 °C. Overall, results demonstrate that despite proximity to the Sahara and the Arabian Peninsula and the dominance of mineral dust in the aerosol sampled, existing mineral dust parameterizations alone would not skillfully represent the near-surface n_s in the observed temperature regime (-6 to -25 °C). The decreased n_s, and results demonstrating that organics dominated the observed IN activity > -15 °C, indicate that the IN-active organic species are limited compared to the mineral IN components of dust. Future efforts to develop or improve representations of dust INPs at modest supercooling (> -15 °C) would benefit from a characterization of the specific organic species associated with dust INPs. More generally, an improved understanding of the organic species associated with increased IN -activity and their variability across dust source regions would directly inform efforts to determine whether n_s-based parameterizations are appropriate for faithful representation of dust INPs in this sensitive temperature regime, whether region-specific parameterizations are required, or whether an alternative to the n_s approach is necessary.

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1 Introduction

Ice-nucleating particles (INPs) modulate the temperature and relative humidity at which ice particle formation occurs in the atmosphere and thus are a key factor that controls ice-phase partitioning in clouds. As initiators of ice formation and related phase-partitioning processes, INPs affect multiple cloud properties and exert a strong influence on cloud lifetime, reflectivity and precipitation efficiency (e.g. Lohmann and Feichter, 2005; Vergara-Temprado et al., 2018).

Globally, desert dust is likely the most abundant aerosol type by mass (Kinne et al., 2006; Kok et al., 2021). Furthermore, multiple studies have demonstrated that mineral dust is the dominant ice-nucleating (IN) species in many parts of the world based on observations (Ardon-Dryer and Levin, 2014; Boose et al., 2016; DeMott et al., 2015a; Price et al., 2018) and modeling of global INP distributions (Burrows et al., 2013; Hoose et al., 2010; Murray et al., 2012; Vergara-Temprado et al., 2017). Annual global dust emission rate estimates range between 400 and 4700 Tg a^{-1} (Huneeus et al., 2011; Kok et al., 2021). Of the global dust loading in the atmosphere (20-29 Tg), North African source regions are estimated to contribute ~50% (11-15 Tg), and the Middle East and Central Asian source regions account for the bulk of the remainder, ~30% (7.7 Tg) (Kok et al., 2021). Analysis of satellite products indicates that dust emissions rates are increasing over the Middle East at a rate of 15% a^{-1} (Klingmüller et al., 2016; Yu et al., 2018).

While Hoose and Möhler (2012) showed that mineral dust INPs generally activate ice crystals at freezing temperatures < -15 °C, dust containing K-feldspar has been shown to nucleate ice at much warmer temperatures, up to -4 °C (Atkinson et al., 2013; Harrison et al., 2016; Niedermeier et al., 2015; Wex et al., 2014; Whale et al., 2015; Zolles et al., 2015). K-feldspars represent up to ~24% of Saharan and Asian dusts by mass (Nickovic et al., 2012). However, knowledge of the abundance and the available surface fraction of aerosolized K-feldspar would be necessary to evaluate the IN efficiency of dust at temperatures > -15 °C (Kanji et al., 2017).

Though mineral dust is considered to be the dominant INP source in many regions, multiple modeling and observational studies suggest that marine INPs are frequently dominant in remote ocean regions in air masses with low concentrations of terrestrial aerosol (McCluskey et al., 2018b, 2018c; Vergara-Temprado et al., 2017; Wilson et al., 2015; DeMott et al., 2016). Using a global aerosol model to simulate marine organic and K-feldspar INP populations, Vergara-Temprado et

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al. (2017) showed that the relative contribution of marine organic vs. dust INPs in remote regions varies seasonally, and that marine organic INPs frequently outnumber K-feldspar INPs (up to 100% of the simulated days in the Southern Ocean during summer). Results from a follow-on cloud-resolving model study showed that Southern Ocean cloud reflectivity is strongly modulated by INP concentrations, indicating that accurate estimates of the radiative energy budget in the Southern Ocean likely require improved and reliable representation of both dust and marine organic INPs (Vergara-Temprado et al., 2018). By generating isolated nascent sea spray aerosol over a range of biological conditions, mesocosm studies have shown that marine INPs are comprised of two classes: a dissolved organic carbon (DOC) type composed of IN-active molecules and a particulate organic carbon (POC) type linked to the death phase of phytoplankton blooms (McCluskey et al., 2017, 2018a).

Parameterizations for both marine and mineral dust populations are commonly implemented in atmospheric models to estimate dust and marine INP concentrations. There are multiple existing mineral dust INP parameterizations used to estimate their concentrations in aerosolized desert dust, some based exclusively on laboratory measurements (e.g., Niemand et al., 2012; Ullrich et al., 2017), and others derived from a combination of laboratory and field measurements (DeMott et al., 2015). There are, additionally, multiple mineral-specific INP parameterizations including illite (Broadley et al., 2012), kaolinite (Welti et al., 2012), quartz (Harrison et al., 2019) and K-feldspar (Atkinson et al., 2013). The parameterizations by Ullrich et al. (2017, hereafter, "U17") and Niemand et al. (2012, "N12") were developed using dust samples from multiple deserts, and both found little variability in the IN activity between dusts from locations as disparate as the Sahara and Asia. DeMott et al. (2015, "D15") found agreement between their observations-based parameterization and N12, supporting the validity of laboratory-based parameterizations. Results in D15 also confirmed the conclusions of N12 and U17: that to first order, dusts from distinct regions can be parameterized as a single particle type. The D15 parameterization has been considered to be representative of dust that has undergone atmospheric photochemical and oxidative processes in transport (i.e., "aged" dust), because the parameterization was derived from observations made far (1000s of kilometers) from the dust emissions sources (Boose et al., 2016).

By contrast, few studies report INP measurements near (e.g., $<$ 1 day of transport) a major dust source, and the lack of observations near dust source regions inhibit the evaluation of the ability

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of existing dust INP parameterizations to represent nascent dust populations (Boose et al., 2016; Gong et al., 2020; Price et al., 2018). INP observations are particularly lacking for the sensitive temperature regime > -20 °C. Boose et al. (2016) found that D15 overpredicted INPs observed during Saharan dust events at a location within 100s of km of the Sahara (Izaña, Tenerife, Spain) by 2-3 orders of magnitude, suggesting that aging may lead to increased IN efficiency in mineral dust and that D15 may be less representative of nascent dust. These conclusions were supported by Conen et al. (2015), who found that concentrations of INPs at -20 °C measured during Saharan dust events were one order of magnitude higher at Jungfraujoch in the Swiss Alps than in Izaña, where dust events had occurred 1-7 d prior to reaching Jungfraujoch. Gong et al. (2020) measured INPs in a variety of atmospheric and seawater sample types at Cabo Verde and determined mineral dust to be the dominant source of INPs observed in the atmosphere but found that INPs with freezing temperatures > -10 °C were likely biological. At altitudes between 30 and 3500 m in the same region, Price et al. (2018) found that measured concentrations of INPs ranged two orders of magnitude at a given temperature, and that the observed concentrations related to the atmospheric dust loadings.

Recently, multiple studies have provided new, much-needed observations of ambient atmospheric INPs in marine environments (DeMott et al., 2016; Hartmann et al., 2020; McCluskey et al., 2018b, 2018c; Yang et al., 2020) where data was historically lacking and, consequently, an impediment to achieving predictive understanding of global INP distributions (Burrows et al., 2013). There are now two parameterizations available for the estimation of atmospheric concentrations of marine INPs emitted from the ocean surface: Wilson et al. (2015), which estimates cumulative INPs from total organic carbon (TOC) concentrations in simulated SSA, and McCluskey et al. (2018), which estimates ice nucleation site density (n_s) from aerosol surface area. Wilson et al. (2015) and McCluskey et al. (2018) derived marine INP parameterizations from field measurements of INPs in Atlantic and Arctic Ocean sea surface microlayer samples and pristine SSA samples over the North Atlantic Ocean, respectively.

Here, we report observations of INPs measured in air masses influenced by both desert dust and marine aerosol (Edtbauer et al., 2020) in close proximity to the two greatest global dust aerosol sources: the Sahara (#1) and the Arabian Peninsula (#2) (Kok et al., 2021). INP concentrations were measured in 26 aerosol samples collected during Air Quality and Climate Change in the

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Arabian BAsin (AQABA), a shipborne campaign which took place July – August 2017 on a transect that spanned the `central and eastern parts of the Mediterranean, the Red Sea, the Gulf of Aden, the Arabian Sea and Arabian Gulf. Observed n_s were compared to dust and marine INP parameterizations, and the contributions of heat-labile (e.g., proteinaceous) and organic compounds to observed INP populations were assessed via heat and peroxide treatments. Finally, the potential INP source strengths of subsurface seawater (SSW) were assessed and compared with SSW INP measurements from prior studies of remote and coastal seawater.

2 Methods

2.1 Project Overview

The AQABA campaign was conducted from 25 June to 3 September 2017 onboard the RV *Kommandor Iona*. The research voyage was conducted in two transects: the first leg beginning in La-Seyne-sur-Mer, France, heading through the Suez Canal, around the Arabian Peninsula and ending in Kuwait, and second leg a return transect via the same route (Fig. S1-S2). The campaign supported a large suite of on- and offline aerosol and gas-phase measurements (Bourtsoukidis et al., 2019, 2020; Celik et al., 2020; Edtbauer et al., 2020; Eger et al., 2019; Friedrich et al., 2021; Pfannerstill et al., 2019; Tadic et al., 2020; Wang et al., 2020).

2.2 Aerosol and Trace Gas Measurements

Aerosol size distributions were measured using an Optical Particle Spectrometer (OPC, Grimm model 1.109) and a Fast Mobility Particle Spectrometer (FMPS, TSI model 3091). The OPC measures particles in the size range 0.25 – 32 μm, and the FMPS measures particles with sizes between 5.6 nm and 560 nm. The inlet for the aerosol instrumentation was located at the top of a measurement container at a distance of about 25 m from the INP filter sampling unit. To avoid condensation in inlet lines, aerosol samples were passed through a drying system, which reduced ambient relative humidity to an average value of ≈ 40 % in the measurement container. Two additional measurements provided aerosol data from which a filter flag intended to identify and eliminate stack emissions was derived: particle number concentrations as measured by a Condensation Particle Counter (CPC, TSI model 3787) and black carbon concentrations (Aethalometer, Magee AE33). The filter flag, based on short term variation in particle number

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concentration, black carbon concentration, wind direction and speed, was applied to all aerosol data so that samples contaminated by stack emissions could be identified.

Particle surface area concentrations were derived from FMPS and OPC measurements as follows. Geometric diameters were estimated from the measured mobility diameters (FMPS) and optical particle diameters (OPC). Aerosols were assumed dry at sampling conditions following the drying system described above. To convert optical particle diameters into geometric diameters, it was assumed that all coarse particles (d_p > 3000 nm) were composed of sea salt and dust with a mass ratio of 25% to 75%, and for the respective refractive indices and shapes the measured optical particle diameters were converted into geometric diameters**.** Fine particle (d_p < 700 nm) sizes were converted from d_{opt} into d_{geo} using the optical properties calculated from the PM_1 chemical composition as measured by an Aerosol Mass Spectrometer (Aerodyne HR-ToF-AMS), assuming spherical particles. For particles in the intermediate size range (700 – 3000 nm), log-linear interpolation of optical and spherical properties was applied for conversion of optical into geometric particle diameters. The mobility diameters measured by the FMPS were considered equivalent to the geometric diameter, assuming spherical particles. From the resulting particle size distributions, particle surface area was calculated for each size bin. Total particle surface concentrations were determined by integrating the surface area distribution for particles up to 10 µm. The overall uncertainty of derived particle surface area concentrations is estimated to be 30%.

The water-soluble fraction of total suspended particles (TSPs) was monitored with hourly resolution using a Monitor for AeRosols and Gases in Ambient Air, MARGA (Metrohm Applikon model S2, Herisau, Switzerland). Sea salt concentrations were estimated by scaling measured soluble Na^+ concentrations by 3.27 following Manders et al. (2009). Hourly composition data was linearly interpolated for 4 samples where 1-3 hours (of 7-24 hours total sampling time) was missing (Fig. S3).

Nitric oxide (NO) concentrations were measured using a commercially available two-channel chemiluminescence monitor, CLD 790 SR (ECO Physics AG, Dürnten, Switzerland). During the AQABA campaign, the CLD 790 SR, MARGA, FMPS, OPC, HR-ToF-AMS, CPC and

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Aethalometer were operated within laboratory containers on the main deck of the research vessel. The NO measurements were used to prevent stack sampling during INP collection (see Sect. 2.4).

2.3 Dust Mass Concentrations from MERRA

Area-averaged hourly dust surface mass concentrations along the cruise track were obtained from the (0.5 x 0.625 °) Modern-Era Retrospective analysis for Research and Application, version 2 (MERRA-2; Gelaro et al., 2017). MERRA-2 uses the GEOS-5 Earth system model (Molod et al., 2015; Rienecker et al., 2011) with 72 vertical layers between the surface and 0.01 hPa (~ 80 km) and the three-dimensional variational data assimilation Gridpoint Statistical Interpolation analysis system (Kleist et al., 2009; Wu et al., 2002). It simulates 5 types of aerosols (dust, sea salt, sulfate and black and organic carbon) using the Goddard Chemistry, Aerosol, Radiation, and Transport (GOCART) model (Chin et al., 2002; Colarco et al., 2010). Dust emissions and deposition rates in MERRA-2 are estimated by summing the emissions and deposition rates across GOCART simulated dust particles between 0.1 - 10 µm in size (dry diameter) (Gelaro et al., 2017). Dust emissions are constrained by wind-driven erosion over the source locations, which are identified from the topographic depression map (Ginoux et al., 2001). Aerosol observations are derived from various satellite products and are jointly assimilated within GEOS-5 with meteorological observations (Buchard et al., 2017). MERRA-2 has been shown to successfully reproduce the interannual variability of North-Atlantic dust transport. Additionally, the improved aerosol assimilation scheme in MERRA-2 was shown to have a positive impact on the representation of long-range dust transport from the Sahara compared to prior versions (Buchard et al., 2017).

2.4 Measurement of Ice Nucleating Particles

Ambient aerosol sampling for offline measurement of INPs was conducted from 5 Jul – 31 Aug 2017 on the *Kommandor Iona's* wheelhouse top (platform above the bridge), ~25 m from the online aerosol measurements inlet and ~35 m from the ocean surface (Fig. S4). Sampling locations along the cruise transect corresponding to each aerosol sample are shown in Figs. S1-2.

Aerosol samples were collected over 3-28 hour periods on polycarbonate filters (47 mm diameter, 0.2 µm pore-size, Whatman® Nuclepore, Chicago, Illinois, USA) placed in open-face Nalgene® Analytical Filter Units (Waltham, Massachusetts, USA). Aerosol sampling flow rates through the

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filter units were set to 10-13 Lpm using a MassStream™ mass flow controller (Bethlehem, PA, USA) connected inline with a rotary vane pump (Thomas QR-0100, Gardner Denver ©, Monroe, LA, USA). To decrease exposure to stack emissions, the pump was automated to switch off when online measurements of NO exceeded one standard deviation above the average background concentration for over 1 minute (~ 0.4 ± 0.8 ppb). Comparing the stack contamination filter flag for aerosol measurements (Sec. 2.2) with INP sampling periods additionally indicates no influence of stack emissions on INP filter samples.

Prior to sampling, filters were cleaned by soaking in 10 % H_2O_2 for 10 minutes followed by rinsing three times with deionized water, the last rinse further "polished" by passage through a 0.1 µm pore-size syringe filter (Puradisc, Whatman ®, Maidstone, U.K). Filters were pre-loaded into filter units in a laminar flow hood to further minimize contamination from handling. After collection, each aerosol filter was placed in a 60 mm diameter sterile Petri dish (Life Science Products, Frederick, Colorado, USA) using pre-cleaned acetyl plastic forceps (Fine Science Tools, Foster City, California, USA), sealed with Parafilm and stored frozen (-20 °C). Samples were shipped in a dry shipper via Cryoport® High Vol Shipper at -180 °C and upon arrival at the laboratory were stored at -80 °C until processed, within 18 to 38 months of collection. To release collected particles, filters were immersed in 5-8 mL ultrapure water (Cat. Number W4502, Sigma-Aldrich®, St. Louis, MO, USA) and shaken for 20 minutes just prior to measurement. Eight samples were additionally diluted 100-fold to measure INP concentrations at lower freezing temperatures (Fig. S5).

INP concentrations were measured using the SIO-Automated Ice Spectrometer (SIO-AIS), an immersion freezing droplet assay instrument that is described in detail in Beall et al. (2017). Briefly, the aerosol sample suspensions and SSW samples were distributed in 30 x 50-µL aliquots into clean 96-well polypropylene sample trays (OPTIMUM® ULTRA Brand, Life Science Products). An equal number and volume of aliquots of ultrapure water accompanied each sample in the tray as a control. Trays were then inserted into an aluminum block that was cooled at -0.87 °C min^{-1} until the samples are frozen. Cumulative INP number concentrations per temperature per volume liquid are calculated using the fraction (*f*) of unfrozen wells per given temperature interval:

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$$n_{INP,L} = \frac{-ln(f)}{V_d} \qquad \text{Eq. (1)}$$

where V_d is the volume of the sample in each well. For aerosol filter samples, cumulative INP number concentrations are calculated using the ratio of the volume used for resuspension of the particles (V_{re}) to the volume of air sampled (V_A):

$$n_{INP} = \frac{-ln(f) \cdot V_{re}}{V_d \cdot V_A} \qquad \text{Eq. (2)}$$

Prior to calculating $\boldsymbol{n_{INP}}$, the fraction of unfrozen wells (*f*) was adjusted for contamination in the water used for suspension by subtracting the number of frozen ultrapure water wells per temperature interval from both the total number of unfrozen wells and total wells of the sample. The $\boldsymbol{n_{INP}}$ was additionally adjusted for background INPs from filters and sampling handling processes. Background INP concentrations were estimated using measured INP concentrations in aerosol sample field blanks, which had been momentarily placed in the sampling apparatus before removal and unloading and storage of the filter. Seven field blank samples were collected, one every ~ 7 days of the cruise (Fig. S6). INP concentrations were measured in field blanks as described above, and the $\boldsymbol{n_{INP}}$ simulated using the mean air volume sampled (6680 L). Figure S6 shows the estimated $\boldsymbol{n_{INP}}$ across the 7 field blanks, which ranged between 3.0×10^{-4} and 3.0×10^{-2} L^{-1} at -20 °C. The freezing onset temperatures detected in the field blanks ranged between -6 and -27 °C. To correct $\boldsymbol{n_{INP}}$ measured in aerosol samples for background INPs from sample handling, a linear regression of the average INP concentration measured in field blank suspensions (mL^{-1} water) was used to estimate background concentrations of INPs in samples at all temperatures between -14.5 °C and -27 °C. The estimated background INP concentration was then subtracted from the INP concentration measured in filter sample suspension volumes in this temperature range prior to calculating $\boldsymbol{n_{INP}}$. The $\boldsymbol{n_{INP}}$ measured in one aerosol sample (f033) fell within the estimated INP background levels.

For this study, the detection limit was 0.68 INPs mL^{-1} liquid or 0.001-0.0024 INPs L^{-1} air for the maximum and minimum air volume sampled, respectively. To extend the upper limit of detection

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(i.e., the point at which all droplets have frozen) dilutions of 1:10 and 1:100 were performed on 8 samples (Fig. S5).

The ice-active surface site density, n_s, is a metric used to define the ice-nucleating capabilities of an aerosol species (i.e., an aerosol sample of all the same particle type) (Kanji et al., 2017) as follows:

$$n_s = \frac{N_{ice}}{N_{tot} \times A\ (cm^2)} \qquad \text{Eq. (3)}$$

where N_{ice} is the number of frozen droplets, N_{tot} is the total number of particles in a monodisperse aerosol population, and A is the surface area per particle.

The value of n_s can also be approximated for polydisperse aerosol samples containing multiple aerosol types:

$$n_s = \frac{N_{ice}}{A_{tot}\ (cm^2)} \qquad \text{Eq. (4)}$$

where A_{tot} is the total surface area of the polydisperse aerosol sample. The difference between the n_s approximation (Eq. 4) and n_s (Eq. 3) is that many particle types are typically included in the n_s approximation, and in an ambient aerosol measurement most of these are not ice nucleating. Furthermore, the subset of INPs in the sample are likely also of different types, which likely have different n_s in the strict sense (Eq. 3). Nevertheless, the n_s approximation is a useful metric for comparing the ice-nucleating ability of different air masses and source regions and is often used for comparing data across studies of INPs measured in ambient air. It is extremely challenging to separate measurements of INPs and surface area by each particle type, and requires, for example, combining online measurements of single particle chemistry, size distributions and INPs (Cornwell et al., 2019). All n_{INP} and n_s are reported normalized to a standard temperature of 273.15 °K and pressure of 1013 hPa.

Heat and hydrogen peroxide treatments were applied to a subset of samples (12 of 26) to test for heat-labile biological (e.g., proteinaceous) and organic INP composition, respectively (McCluskey et al., 2018b; Suski et al., 2018). For each heat-treated sample, a 2 mL aliquot of the original ultrapure water suspension was heated to 95 °C for 20 min in a water bath and re-tested to assess the reduction in INP concentrations. For peroxide treatments, 1.6 mL of the original suspension was combined with 0.8 mL of 30% H_2O_2 (Sigma Aldrich®, St. Louis, Missouri, USA) to achieve

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a final concentration of 10%, then the mixture was immersed in water, and heated to 95 °C for 20 min while being illuminated with two 26-W UVB fluorescent bulbs to generate hydroxyl radicals. To remove residual H_2O_2, to prevent otherwise significant freezing point depression, the solution was cooled and catalase (Cat. number IC10042910, MP Biomedicals, Santa Ana, California, USA) was added. Since catalase is itself decomposed by H_2O_2, while simultaneously catalyzing peroxide's disproportionation into water and oxygen, the enzyme was added in several 20 µL aliquots, allowing several minutes between each, until no effervescence resulted upon its addition. Fisher's Exact Test was applied to frozen and unfrozen well fractions between each untreated sample and its corresponding treated sample to test for significant differences ($p < 0.05$). Note that significant difference in frozen well fraction is insufficient as a sole indicator of sensitivity in peroxide treated samples because samples are diluted 2:3 (by 33%) compared to untreated samples. As INP concentrations can be corrected for the dilution by scaling (as opposed to frozen well fractions), the overlap in 95% binomial sampling confidence intervals (Agresti and Coull, 1998) between the untreated and peroxide-treated sample is an additional indicator of sensitivity for a given data point in the peroxide-treated sample spectrum within ± 0.2 °C, the uncertainty in the SIO-AIS temperature measurement (Beall et al., 2021). A lack of overlap in the 95% binomial sampling confidence interval within ± 0.2 °C equates to a significance threshold of $p < 0.005$ (Krzywinski and Altman, 2013).

INP concentrations were additionally measured in 10 SSW samples. For seawater sampling, a water intake vertical steel pipe was positioned on the starboard of the ship approximately 2 m below the seasurface level. The seawater was pumped into a 200 L stainless steel tank and continuously exchanged at a rate of 3000 L h^{-1}. SSW samples for INP analysis were collected in 15 mL sterile centrifuge tubes (Falcon™, ThermoFisher Scientific, Waltham, Massachusetts, USA) and stored frozen at -20 °C until they could be shipped in a dry shipper via Cryoport® (-180 °C) and ultimately stored at -80 °C as for aerosol samples. Heat and hydrogen peroxide treatments as described above were applied to five of these. To assess the contribution of submicron INPs to

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total measured INPs, 2 mL of SSW was filtered through a 0.2 µm sterile syringe-filter (Acrodisc® Pall®, Port Washington, New York, USA) and re-tested.

2.5 FLEXPART Back Trajectories

Air mass 72-hour back-trajectories for each sample were simulated using the FLEXible PARTicle dispersion model (FLEXPART) in backward mode (Stohl et al., 1998). NOAA Climate Forecast System (CFS) short-duration (t < 6 h) forecasts (Saha et al., 2014) were used as three-dimensional forcing datasets. Particle releases from 35 m above sea level (ASL) followed the vessel track using vessel position information from the European Common Automatic Weather Station (EUCAWS; http://eumetnet.eu/; last access Sept. 2021).

3 Results and Discussion

3.1 Characteristics of INPs Observed During AQABA

A total of 26 aerosol samples were collected July – August 2017 during AQABA for offline measurements of INPs. The INP concentrations (n_{INP}) measured in samples collected in the Mediterranean Sea, the Red Sea, the Gulf of Aden, Arabian Sea, Gulf of Oman, and Arabian Gulf spanned up to 3 orders of magnitude at -15 °C (Fig. 1). Average ambient dust concentrations during each sampling period ranged from 2-490 µg m^{-3} (PM_{10} Table 1). There is no agreed-upon standard for definition of extreme dust events in the literature, though the 24-hr average WHO or US EPA health standards for average PM_{10} are commonly used (Gandham et al., 2020; Khaniabadi et al., 2017). Using the US EPA health standard for PM_{10} as a threshold for extreme events (150 µg m^{-3}), 14 of the 26 samples were collected during dust events. This is conservative given the equivalent WHO guideline for PM_{10} is 50 µg m^{-3} (WHO, 2005), in which case 22 of the 26 sampling periods would be classified as dust events. Prior studies have reported comparable PM_{10} levels during dust events in the region (Gandham et al., 2020; Krasnov et al., 2016; Shahsavani et al., 2012).

FLEXPART 72-hour air mass back trajectories show that many of samples collected during extreme dust events (f013, f014, f016, f018, and f020) were influenced by emissions from North Africa and the Arabian Peninsula (Figs S8-S9). Other source regions included the Mediterranean,

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Nile Delta, Sinai Peninsula (f006-f008), Northeast Egypt (f009-f010), Iran (f024), and Southern and Eastern Europe (f040, f042, f044).

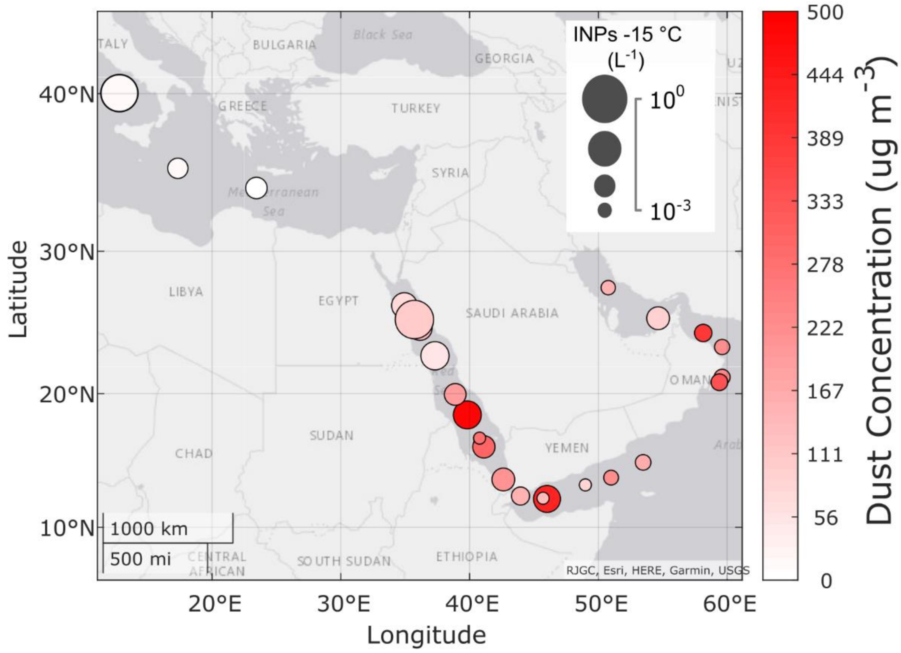

Figure 1. Map of the sample locations for 26 aerosol samples collected on the RV *Kommandor Iona* during Air Quality and climate change in the Arabian BAsin (AQABA). Measured INP concentrations spanned three orders of magnitude at -15 °C, from 10^{-3} to 0.5 L^{-1}. Marker sizes indicate abundance of INPs. Marker colors indicate the average ambient dust mass concentration during the sampling period from hourly MERRA-2 reanalysis data.

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Table 1. Summary of aerosol samples collected during AQABA. "—" indicates where data are missing.

Sample ID	Start datetime (UTC)	Stop datetime (UTC)	Latitude	Longitude	Sample Volume (L air)	Aerosol Surface Area (μm^2 cm^{-3})	Average Dust Concentration (μg m^{-3})	Average Seasalt Concentration (μg m^{-3})
f006unt	05-Jul 05:46	05-Jul 11:37	26.224	35.025	3370	290	170	-
f007unt	05-Jul 16:40	05-Jul 19:51	26.291	34.933	2588	260	70	-
f008unt	06-Jul 07:09	06-Jul 14:08	25.225	35.775	5225	180	100	-
f009unt	07-Jul 05:50	07-Jul 15:07	25.011	35.947	6940	350	110	-
f010unt	08-Jul 16:33	09-Jul 05:59	23.623	36.931	8073	220	50	-
f013unt	14-Jul 12:26	14-Jul 16:13	18.687	39.672	2283	260	490	10
f014unt	15-Jul 05:10	15-Jul 11:49	16.552	40.834	4000	270	300	5
f016unt	18-Jul 07:04	18-Jul 14:52	11.939	45.334	4690	260	430	-
f018unt	22-Jul 10:20	22-Jul 18:44	20.941	59.474	5025	210	340	-
f019unt	23-Jul 04:48	23-Jul 13:34	21.410	59.691	5270	220	230	-
f020unt	25-Jul 17:15	26-Jul 04:02	23.976	58.809	6511	-	390	5
f023unt	04-Aug 04:05	04-Aug 11:56	28.084	50.284	4720	830	150	4
f024unt	05-Aug 05:57	05-Aug 13:53	25.432	53.853	5221	360	90	-
f025unt	07-Aug 09:26	07-Aug 16:46	23.814	59.186	4410	50	220	12
f030unt	13-Aug 07:08	14-Aug 11:06	15.970	54.705	15111	30	160	-
f031unt	14-Aug 15:03	15-Aug 09:03	14.003	52.357	12972	30	220	-
f032unt	15-Aug 09:42	15-Aug 15:07	13.354	49.432	3260	100	80	6
f033unt	16-Aug 09:30	16-Aug 13:17	12.208	45.706	2280	90	130	2
f034unt	16-Aug 13:27	17-Aug 07:04	12.177	45.429	8464	170	150	1
f035unt	17-Aug 07:30	17-Aug 14:55	13.308	42.974	4460	340	210	2
f036unt	18-Aug 06:36	18-Aug 15:03	16.290	41.038	6634	210	280	2
f037unt	19-Aug 07:05	20-Aug 07:04	18.699	39.609	18806	240	190	7

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f038unt	21-Aug 07:22	21-Aug 16:01	24.112	36.554	6700	260	140	-
f040unt	26-Aug 16:02	27-Aug 07:04	33.803	24.814	9030	90	<10	3
f042unt	28-Aug 07:51	28-Aug 16:02	35.310	17.965	6396	160	10	2
f044unt	31-Aug 08:30	31-Aug 20:16	39.569	13.380	11296	210	10	-

In Figure 2, approximated ice nucleation site densities (n_s) are compared with multiple population-specific observations and parameterizations for dust and marine INPs. The AQABA measurements are also compared with observations from dust-laden air over the Tropical Atlantic (Price et al., 2018). Overall, observations nearly bridge the full regime between M18, the parameterization for marine INPs (McCluskey et al., 2018b), and multiple dust INP parameterizations based on laboratory studies of surface dust. At higher temperatures, between -5 and -12 °C, most observations show agreement with the composite spectrum of n_s observed in a range of marine and coastal environments from DeMott et al. (2016) and Yang et al. (2019), and/or the Atkinson et al. (2013) K-feldspar parameterization. Between -10 and -20 °C, several samples agree with M18 marine INP parameterization within an order of magnitude, whereas two to three n_s spectra approach the U17 and N12 laboratory-derived dust INP parameterizations within an order of magnitude (Niemand et al., 2012; Ullrich et al., 2017), depending on temperature. Multiple samples (~8) additionally agreed with Price et al.'s (2018) observations of INPs between 30-3500 m above the dusty Tropical Atlantic, and most agree with the Gong et al. (2020) surface-level observations in the same region (Cabo Verde).

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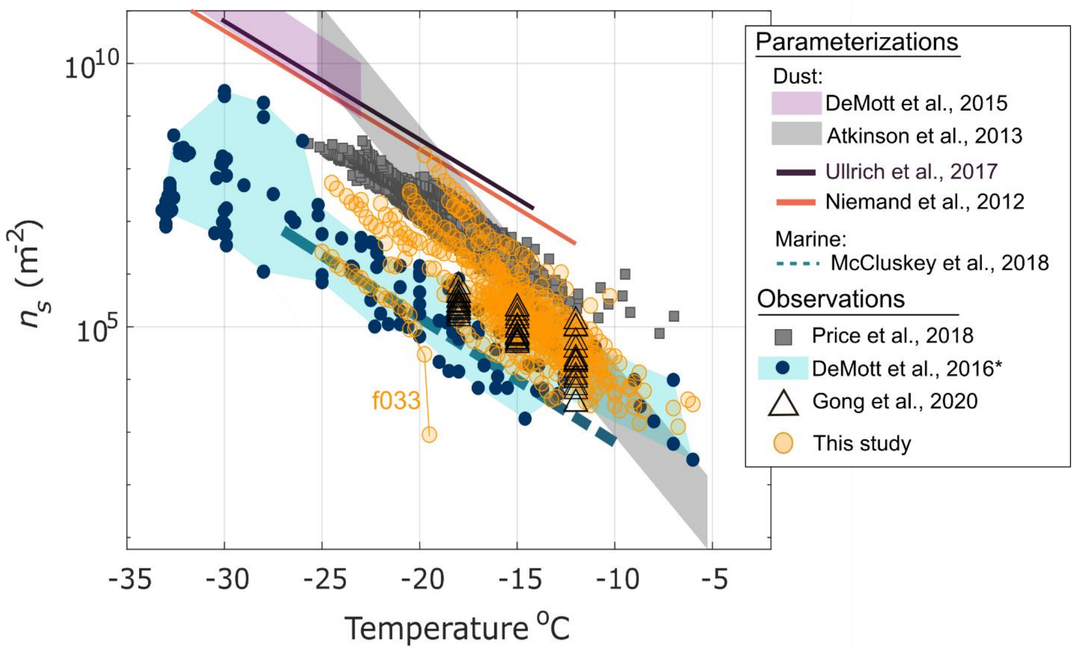

Figure 2. Ice nucleation site densities (n_s) as a function of temperature for 25 of 26 aerosol samples collected during AQABA. Gong et al. (2020) and Price et al. (2018) measured INPs in dust-dominant air masses in the tropical east Atlantic, with minor contributions from SSA, while DeMott et al. (2016) measurements were collected across a range of locations and conditions within the marine boundary layer comprising air masses mostly dominated by relatively pristine marine SSA. INP concentrations measured in sample f033 were below the detection limits imposed by field blanks (see Sect. 2.4, Fig. S6). Sample f020 is not shown due to missing aerosol surface area data during the sampling period. *DeMott et al. (2016) data shown have been updated with additional data from Yang et al. (2020).

Considering the frequency of dust events encountered (Table 1), and the high probability that dust was the dominant aerosol source during most sampling periods, it is striking that most n_s spectra observed are 1-3 orders of magnitude lower than the values predicted by dust parameterizations. As noted in Gong et al. (2020), some deviations could be expected due to the difference between approximated n_s based on total particle surface area in ambient measurements and true n_s based on

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surface area of a homogeneous aerosol population (see Methods Sec. 2.4). The FLEXPART back trajectories show that air masses for multiple samples originated from densely populated regions such as Southern and Eastern Europe (f040, f042, f044, Fig S9). The back trajectories also show that for samples f006-f008, f010, and f038, air masses were influenced by the populous region around the Nile River Delta. Agricultural soil dusts represent a potential constituent of the INPs observed from these regions. A range of n_s has been reported in studies of agricultural soil dusts, the lower end of which agrees with the n_s observed in the present study between -8 and -25 °C (Steinke et al., 2016; Tobo et al., 2014; O'Sullivan et al., 2014).

Given the marine environment in which sampling occurred, a significant amount of sea spray aerosol (SSA) was also detected in many of the sampled airmasses (Table 1), and likely present in others for which no composition data were available. Edtbauer et al. (2020) reported the detection of high levels of dimethyl sulfide (DMS, up to 800 ppt) in the Gulf of Aden associated with a local phytoplankton bloom during AQABA (as evidenced by visible bioluminescence around the ship at night) as well as high levels of $DMSO_2$ and other marine biogenic VOCs from the Somalian upwelling region. As mentioned above, the n_s for most samples between -6 and -18 °C agree with n_s derived from observations across various locations within the marine boundary layer (Fig. 2). However, considering that SSA is associated with 1000 times fewer IN sites per unit surface area than dust (i.e. 1000× lower n_s) (McCluskey et al., 2018b) and the high relative abundance of dust compared to sea salt concentrations (Table 1), it is unlikely that the observed INPs originated from SSA. In general, detection of marine INPs in ambient aerosol is challenging due to their low relative abundance and decreased efficiency compared to dust (DeMott et al., 2016; McCluskey et al., 2018b). Thus, while SSA contributed to the measured aerosol surface area (Table 1), it is unlikely that the INPs observed in this study were marine in origin, or at least that this is indiscernible in the present study or based on present parameterizations of these populations.

Heterogeneous aerosol composition in the sampled air masses likely contributed to some of the low n_s spectra observed due to the contribution of non-INPs to the measured aerosol surface area. However, the difference between n_s observed during the most extreme dust events, i.e., when the aerosol population was likely approaching homogeneity in composition, and the n_s predicted from N12 and U17 was still greater than 2 orders of magnitude. Figures 3(a) and (b) show overlap in n_{INP} and n_s observed in samples collected in low dust and high dust conditions, indicating that the

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INP populations observed during AQABA exhibited similar efficiencies despite variation in total aerosol composition and dust loading. No correlation was found between n_{INP} and aerosol surface area (Fig. S7) or dust concentration. This result is in contrast to Price et al. (2018) who found the variability in n_{INP} to be largely determined by variability in dust loading or aerosol surface area. Yet, the aerosol surface area concentrations compare very well, indicative of comparable dustiness in the two studies. Excluding three samples Price et al. (2018) collected in an exceptionally optically thick dust layer, the average aerosol surface area was 227 ± 68 μm^2 cm^{-3} vs. 226 ± 26 μm^2 cm^{-3} for the present study. Furthermore, the sample with the highest n_s at -15 °C (f040) was collected when dust concentrations were lowest (2 μg m^{-3}) (Fig. 3, Table 1). This is also in direct contrast to Price et al. (2018), who found that the highest n_s observed corresponded to the highest dust loading.

Gong et al. (2020) also observed n_s lower by more than 2 orders of magnitude compared to N12 and U17 despite the large fraction of supermicron INPs (77-83% depending on temperature), and that the supermicron particles were mainly mineral dust. The large differences between parameterized n_s for dust, and n_s observed in both Gong et al. (2020) and the present study between -12 and -25 °C demonstrate that existing n_s-based parameterizations may not faithfully represent n_s at moderate freezing temperatures, despite proximity to major source regions. Whereas DeMott et al. (2015a) found that for temperatures < -20 °C, mineral dust particles from Saharan and Asian deserts may be parameterized as a common particle type, our findings suggest that characteristic n_s parameterizations for dust from different source regions may be needed > -20 °C, or, alternatively, that this temperature regime requires an alternative to an n_s-based parameterizations. Gong et al. (2019a) demonstrated that predicting n_{INP} from surface area size distributions alone may not be feasible in environments where the aerosol and/or INP composition are unknown and proposed a probability density function PDF-based approach to predicting INPs at a given freezing temperature.

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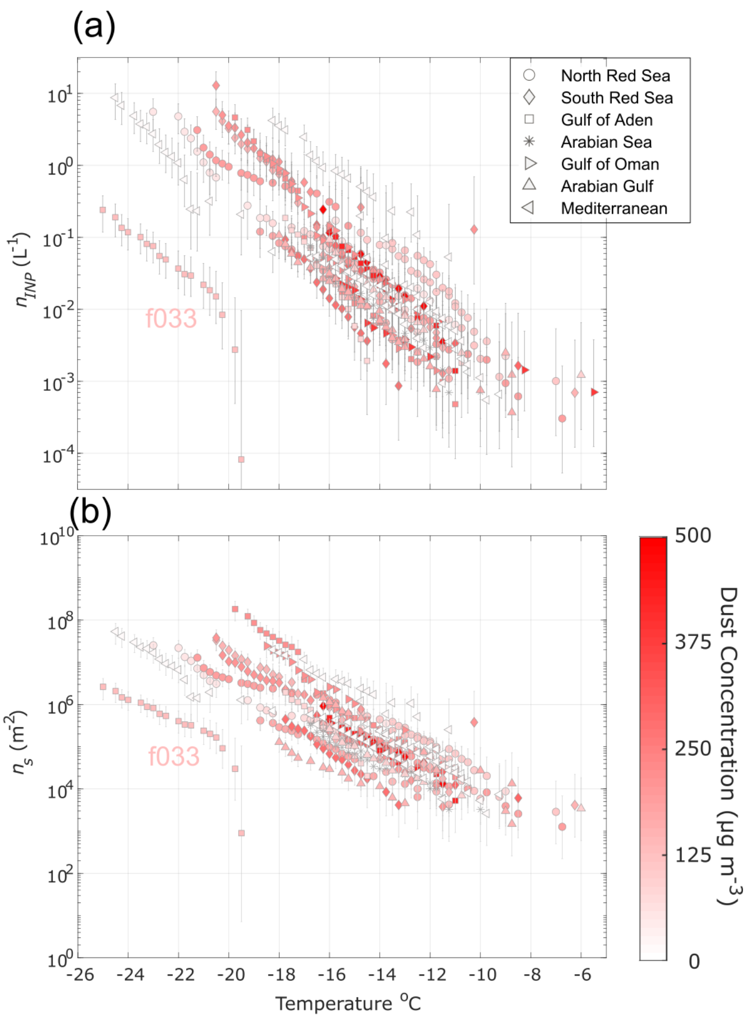

Figure 3. INP concentrations (n_{INP}) **(a)** and ice nucleation site densities (n_s) **(b)** as a function of temperature for 26 aerosol samples collected during AQABA. Markers are colored by the average ambient dust concentration for the respective sampling period. The n_s measured in samples collected during low dust conditions are equal to or greater than (up to 100×) the n_s measured during dust events between -9 and -18 °C. INP concentrations measured in sample f033 were below the detection limits imposed by field blanks (see Sect. 2.4, Fig. S6). Sample f020 is not shown in (b) due to missing aerosol surface area data during the sampling period.

Offline treatments for testing heat lability and organic composition of INPs were performed on 12 samples via heat and H_2O_2 treatments, respectively (Fig. 4). Prior studies have shown that the IN-

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active component of various types of mineral dusts are insensitive to heat treatments (Conen et al., 2011; Hara et al., 2016; Hill et al., 2016; O'Sullivan et al., 2014). The IN activity of K-feldspar, the dominant IN component of mineral dust, was additionally found to be insensitive to digestion with peroxide (O'Sullivan et al., 2014). A small number of studies reported degradation of IN activity with peroxide treatment and/or heat treatment in Arizona Test Dust (ATD), that they attributed to organic material (Perkins et al., 2020; Yadav et al., 2019). Thus, we assume here that any degradation of IN activity due to heat and peroxide treatment are due to loss of heat-labile (e.g. proteinaceous) and heat-stable organic INPs, respectively.

Fisher's Exact Test was applied to frozen and unfrozen well fractions for each untreated sample and its corresponding treated sample to test for significant differences ($p < 0.05$). Sensitivity to peroxide in most samples demonstrate the consistent presence of stable organic INPs at temperatures ≥ -15 °C. The lack of peroxide sensitivity at temperatures below -15 °C indicates dominance by mineral dust INPs at lower temperatures. Heat sensitivity in five samples suggests that biological INPs contributed to their warmest freezing INPs. Gong et al. (2020) similarly found heat-sensitivity in INPs at temperatures > -10 °C. Four of the 12 samples exhibited heat sensitivity at relatively moderate temperatures -11 to -18 °C, including the two samples collected in the Mediterranean Sea. One sample (f010) exhibited increased INP concentrations in freezing temperatures below -18 C after heat and peroxide treatments. That the response to both heat and peroxide were nearly identical (Fig. 4) suggests that compounds may have been released from the surface during heating, uncovering a more IN active surface underneath (heating was common to both procedures). The increased n_{INP} post heat and peroxide treatment is an unexpected result given previous studies on treated soil dust measurements (Conen et al., 2011; Hill et al., 2016; O'Sullivan

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et al., 2014; Tobo et al., 2014), though an increase in IN activity after peroxide treatment has also been reported in a Himalayan dust sample (Paramonov et al., 2018).

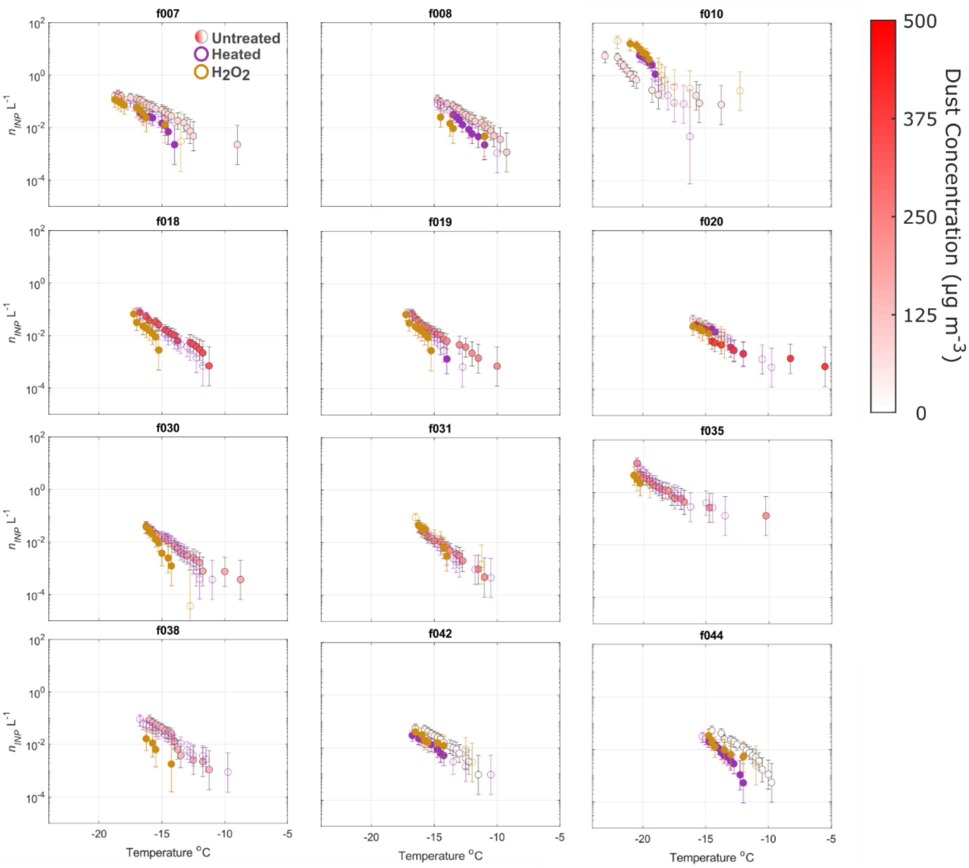

Figure 4. INPs in aerosol samples treated with heat and H_2O_2 (Methods Sec. 2.4) to test for INP heat-lability and organic composition. Markers of untreated spectra are colored by the average dust concentration during the sampling period. Markers of heat-treated and H_2O_2-treated samples are filled to indicate significant INP concentration difference from untreated samples according to Fisher's Exact Test ($p < 0.05$). Sensitivity to H_2O_2 is evident for all samples ≥ -15 °C, indicative of stable organic INPs. Heat-lability is also evident at high to moderate temperatures in multiple samples, demonstrating that biological (e.g., proteinaceous) INPs also contributed to INPs observed during AQABA.

Given the frequency of dust storms and generally high concentrations of dust during most sampling periods, it is surprising that most samples exhibit peroxide sensitivity. Aridisols and entisols are

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the dominant soil types in North Africa and the Arabian Peninsula (Nortcliff, 2012). Both types are associated with the lowest levels of organic carbon, commonly used as a proxy for total soil organic matter, compared to other soil types (3 and 9 g kg^{-1}, respectively) (Yost and Hartemink, 2019).

INP measurements of soil dusts in this region are scarce and have only been reported for a single surface dust soil sample, sample "SD", collected 50 km north of Cairo (Niemand et al., 2012). For comparison with this study, we measured INPs in untreated, heat-treated, and peroxide-treated subsamples of an archived aliquot of the SD sample described in Niemand et al. (2012). Sample SD exhibits sensitivity to both heat and peroxide at temperatures > -16 °C, indicating biological composition of INPs at high freezing temperatures. Multiple AQABA samples influenced by desert air mass sources show similar sensitivities at higher temperatures: f006, f007, f019, and f020. Several others exhibit only peroxide-sensitivity in this temperature range. Overall, the heat and peroxide sensitivities in the SD sample indicate that desert dusts may contribute biological and/or organic INPs at moderate to high-freezing temperatures, such as those observed in AQABA samples. Gong et al.'s (2020) results showing heat-sensitivity in INPs at temperatures > -10 °C further demonstrate the contribution of biological INPs at high temperatures in dusty air masses near N. Africa.

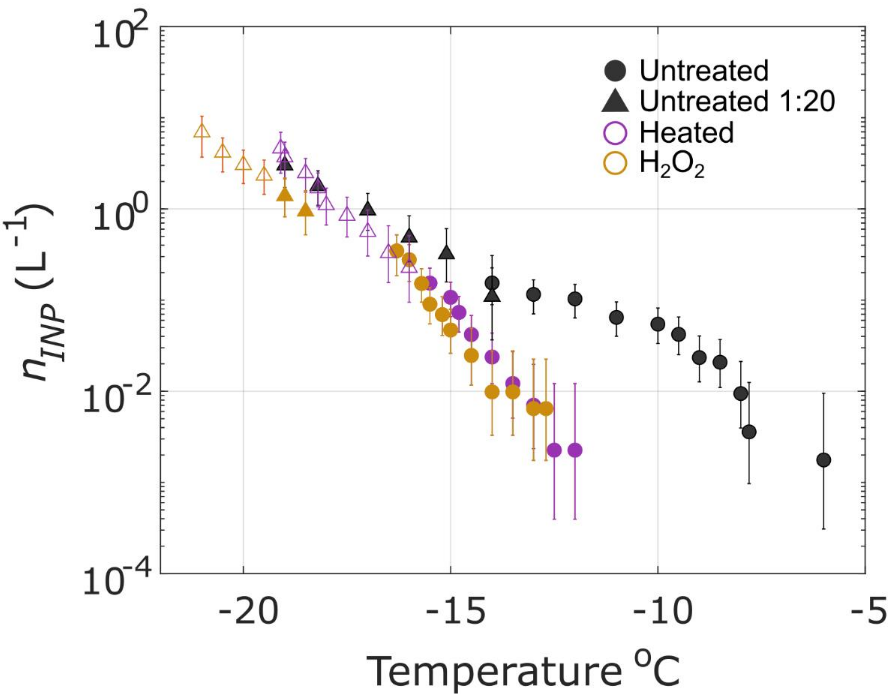

Figure 5. Measured concentrations of INPs in an aerosolized soil dust sample collected 50 km north of Cairo, Egypt, that was treated with heat and peroxide to test for INP heat-lability and organic composition, same as in Fig. 4 above (Methods Sec. 2.4). A 1:20 dilution of the sample is shown (triangles) and markers of heat-treated and H_2O_2-treated samples are filled to indicate significant INP concentration differences from untreated samples according to Fisher's Exact Test ($p < 0.05$). Sensitivity to peroxide and heat treatments indicates biological INPs between -6 and -16 °C.

Considering the high freezing temperatures observed, evidence of organic composition, and FLEXPART back trajectories showing that aerosol sources included populous regions and at least one agriculturally active region (the Nile River Delta), it is possible that agricultural soil dusts contributed to some of the relatively higher n_s, n_{INP}, and heat and peroxide sensitivity observed during AQABA. Samples from air masses influenced by the Nile River Delta or Southern Europe

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(f007-8, f010, f038, f042, f044) show a higher fraction of heat-sensitive INPs (Fig. 4). Heat-sensitivity is indicative of biological INPs, which have been associated with agricultural soil dusts in prior studies (Hill et al., 2016; O'Sullivan et al., 2014). Hill et al. (2016) and O'Sullivan et al. (2014) showed peroxide sensitivity in agricultural soil dusts at temperatures > -18 to -15 °C, respectively, a range which aligns with the peroxide sensitivity exhibited in the present study. Agricultural soil dusts are relatively rich in organic and biological material (Conen et al., 2011, 2016; Ellerbrock et al., 2005; Kögel-Knabner et al., 2008; O'Sullivan et al., 2014) and contribute up to 20-25% of the global dust load (Ginoux et al., 2012). Furthermore, they are associated with IN activities higher than that of mineral dust (Conen et al., 2011; Fornea et al., 2009; Isono and Ikebe, 1960; O'Sullivan et al., 2014; Steinke et al., 2016; Tobo et al., 2014). High onset temperatures, up to -6 °C, are the norm (Conen et al., 2011; Garcia et al., 2012; Hill et al., 2016; O'Sullivan et al., 2014), and the high activity of agricultural soil particles has been attributed to internally mixed organic matter (O'Sullivan et al., 2014; Tobo et al., 2014).

Organic material can condense or adsorb onto aerosols during photochemical and oxidative processes, representing another potential source of organic INPs during AQABA (Dall'Osto et al., 2010; Hinz et al., 2005; Krueger et al., 2004). Could aging explain the organics and decreased n_s observed? Though dust aerosol was collected within 1 day's transport from source regions throughout this study, we cannot rule out the possibility of aging impacts, lacking single particle chemistry measurements (e.g., Sullivan et al., 2007). In addition to field observations of INP concentrations demonstrating that aging increased the IN efficiency of desert dust INPs (see Introduction; Boose et al., 2016; Conen et al. 2015), prior studies of the effects of aging on mineral dust INPs have yielded mixed and sometimes contradictory results, indicating that the impact of aging on IN properties likely depends on multiple factors including the ice nucleation pathway, the type of aging process, surface morphology, and mineralogy (Perkins et al., 2020). Multiple studies have investigated the effects of various aging processes on Arizona Test Dust (ATD) as a proxy for diverse natural dust samples. These included exposure to sulfuric acid, nitric acid vapor, and solution-phase processes (Cziczo et al., 2009; Eastwood et al., 2009; Knopf and Koop, 2006; Salam et al., 2007; Sullivan et al., 2010b, 2010a). Perkins et al. (2020) demonstrated the INP lability in ATD through multiple solution-phase aging processes (e.g., incubation in water, exposure to acid or salt), with up to 1000-fold reductions in INP abundance at freezing temperatures > 10 °C. This result contrasts with the increase in IN activity attributed to aging reported in Boose et al. (2016) and Conen et al. (2015). Perkins et al. (2020) additionally reported that the lability of IN activity in ATD is temperature dependent, with large reductions evident at freezing temperature > 10 °C, yet little to no change at temperatures below -15 °C. By contrast, most of the n_s spectra in AQABA samples were 10 – 1000× lower than established dust parameterizations even at temperatures below -15 °C. In summary, it has proven difficult to determine any consistent impact of atmospheric processing on the IN activity of dust in model systems such as ATD, and few studies have investigated impacts of aging on ambient desert dust, especially at modest supercooling. Furthermore, the use of ATD as a proxy for natural dust in INP studies has been questioned due to the complex IN-properties of natural dust, including mineral composition and defect sites at the particle surface, the latter of which is likely affected by the mechanical processing and milling involved in ATD production (e.g., Perkins et al., 2020 and references therein).

The cause of the decreased n_s observed here and in Gong et al. (2020) compared to dust n_s parameterizations remains elusive. Both studies were conducted in air masses dominated by dust near major sources. In contrast, Price et al. (2018) found agreement near the region of the Gong et al. (2020) study. One obvious difference is that Price et al. (2018) conducted measurements at higher altitudes, between 30 and 3500 m. A prior study that compared n_{INP} in dusty air masses at the surface with n_{INP} collected between 0.5 and 3 km above sea level found that median n_{INP} increased by up to 10× above the surface and correlated to dust loading (Schrod et al., 2017). The decreased n_s compared to Price et al. (2018) is also unlikely to be related to differences in INP measurement. In all three studies, cold stage or droplet assay measurements of immersion mode INPs were used in resuspensions of aerosol collected on filter samples. Recent studies that intercompared instruments designed for measurement of immersion mode INPs showed excellent agreement (i.e., within measurement uncertainty) in measurements of standardized dust and biological samples (DeMott et al., 2018) and when co-sampling ambient aerosol (DeMott et al., 2017). Moreover, the DeMott et al. (2018) intercomparison study demonstrated good agreement in multiple natural dust samples between the various measurement methods used to derive D15,

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N12 and U17 and the droplet assay methods applied in Gong et al. (2020), Price et al. (2018)),) and the present study.

In light of the evidence from this study that INPs were primarily influenced by organics associated with dust, especially at higher temperatures, and the lack of relationship between dust loading, n_s, and n_{INP}, we offer the following points for consideration. Prior studies of aerosolized dust demonstrated that it is frequently enriched in organic matter (6-20×)× and that wind erosion selectively removes the chemically-enriched, fine portion of the soil higher of plant nutrients, organic matter and metals(Aryal et al., 2012; Delany and Zenchelsky, 1976; Van Pelt and Zobeck, 2007). Furthermore, a recent study that measured airborne concentrations of prokaryotic cells over the Red Sea characterized the region as a "global hot spot" with average concentrations of 155,000 (± 65,000), 19× higher than that over the subtropical and tropical open oceans (Mayol et al., 2014; Yahya et al., 2019). Yahya et al. (2019) demonstrated that the microbial loading was very likely related to the high concentrations of dust, as 99.9% of the cells were attached to dust particles. Organic and biological species have been shown to dominate IN activity at temperatures > ~-15 °C in many studies (e.g., Kanji et al., 2017; Ladino et al., 2019; O'Sullivan et al., 2018). Thus, a faithful representation of dust INPs may require two parameterizations: one for the IN activity dominated by minerals < ~-15 °C such as D15, U17 and N12, and another for the dust-associated organics > ~-15 °C. As IN-active organics are limited compared to the IN-active mineral component of dust, we could expect an increase in n_s slope between warm and cold regimes. The apparent decreased n_s observed in this study between -18 and -12 °C could potentially be related to a plateau in n_s through the transition between the mineral and organic "modes" (see Fig. 5). This study underscores the need to characterize the IN-active organic species associated with dust from major source regions and to investigate the extent to which biological and/or organic particles contribute to INP populations in dust-laden air masses at high to moderate freezing temperatures.

3.2 Seawater Source Potential

The n_{INP} values in 10 SSW samples collected during AQABA were used to characterize the INP source potential of SSA generated by bubble bursting (Wang et al., 2017). Results from prior studies have demonstrated that jet droplets are a more efficient transfer vehicle than film drops of INPs into SSA particles (Mitts et al., 2021; Wang et al., 2017). While it is unlikely that many of

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the INPs detected in aerosol samples were marine in origin (see Sec. 3.1), we measured the INP concentrations in SSW to test whether the seawater source strength was comparable to that of prior studies, or were possibly enriched with INPs due to biological activity or even dust deposition (Cornwell et al., 2020).

Figure S10 shows how the INP concentrations measured at -19 °C in 10 seawater samples varied by the sample collection location. Concentrations ranged between 1 and 50 INPs mL^{-1} and were highest between the Gulf of Oman and the Gulf of Aden. This region exhibited relatively high chlorophyll *a* during the cruise, with levels between 1 and 30 mg m^{-3} (Fig. S11). In Fig. 6, INP concentrations were compared with SSW from the Ellen Browning Scripps Memorial Pier in coastal Southern California (SIO Pier), Cabo Verde in the Northeast Atlantic, the Southern Ocean (McCluskey et al., 2017), and the Northwest Atlantic (Schnell, 1977). AQABA INP concentrations were most comparable with Gong et al.'s (2020) observations in Cabo Verde. The

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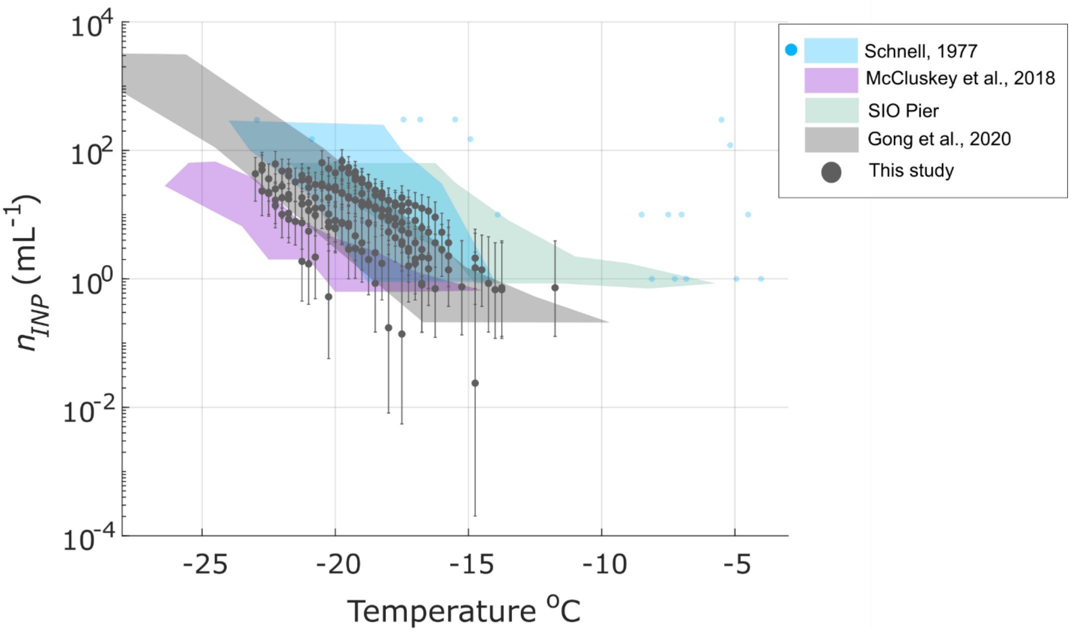
646 lack of any unusually high INP spectra suggests that INP enrichment due to dust deposition
647 (Cornwell et al. 2020) was absent or infrequent.



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649 **Figure 6.** Measured INP concentrations in 10 SSW samples collected during AQABA. Also
650 shown are the composite INP spectrum of coastal SSW samples collected on São Vincente
651 Island, Cabo Verde (Gong et al., 2020), coastal SSW samples collected at the Ellen Browning
652 Scripps Pier (green shading), and SSW samples collected in the Southern Ocean (McCluskey et
653 al., 2018c). Schnell's (1977) SSW measurements are represented as a composite spectrum of 24
654 samples (blue shaded region) and 5 additional spectra (blue markers) from samples that exhibited
655 higher freezing temperatures. All spectra presented are uncorrected for freezing point depression.

656 Offline treatments for testing heat lability, organic composition, and size were applied to 5 of the
657 10 seawater samples (Methods Sec. 2.4). Heat and 0.2 µm filtering treatments suggest that a
658 large fraction of the seawater INPs were heat sensitive and larger than 0.2 µm. These results are
659 indicative of the particulate organic carbon (POC) type of marine INP defined in McCluskey et
660 al. (2018a) (Fig. S12). Heat-resilience but peroxide-sensitivity in sample s001 additionally
661 indicates the presence of non-proteinaceous organic INPs, such as the dissolved organic carbon

(DOC) type defined in McCluskey et al. (2018a). Considering the characteristically low IN activity of the SSW, the lower n_s of SSA compared to mineral dust (McCluskey et al., 2018b), and the frequency of dust events during AQABA, our findings suggest that dust was highly likely to be the dominant INP class observed in this study.

4 Conclusions

Observations from the two-month AQABA campaign in the Mediterranean, Red Sea, Arabian Sea and Arabian Gulf are among the first INP measurements made in close proximity to the two largest dust sources globally: the Sahara and the Arabian Peninsula (Kok et al., 2021). INP concentrations spanned two or more orders of magnitude (0.002 to 0.5 L^{-1} at -15 °C).

In summary, INPs observed during AQABA were very likely dominated by mineral dust with some additional contributions possibly from densely-populated and/or agricultural regions including the Nile River Delta region and Southern or Eastern Europe. Despite proximity to major dust sources and a high frequency of dust events with mass concentrations up to 490 µg m^{-3} (PM_{10}), the observed n_s for most samples was lower by 1-3 orders of magnitude compared to n_s predicted by dust parametrizations N12 and U17 at T < -12 °C (Niemand et al., 2012; Ullrich et al., 2017). Many INPs measured in AQABA showed agreement with the A13 parameterization for K-feldspar (Atkinson et al., 2013), an ice-active component of desert dust, with observations within the marine boundary layer (DeMott et al., 2016; Yang et al., 2020), and with the Price et al.'s (2018) measurements of INP concentrations in dust-laden air masses over the Tropical Atlantic. Peroxide sensitivity was evident in all samples tested (12 of 26), at temperatures ≥ -15 °C, demonstrating a consistent contribution of organic material to warm-temperature INPs. Heat-sensitivity further suggested the presence of biological (e.g., proteinaceous) INPs in a subset of samples, particularly at high freezing temperatures. While the dominant mineral dusts in the region are associated with the lowest concentrations of soil organic carbon globally (e.g., Yost and Hartemink, 2019 and references therein), aerosolized fine dust is known to be enriched in organic matter (Aryal et al., 2012; Delany and Zenchelsky, 1976; Van Pelt and Zobeck, 2007) and is additionally associated with high microbial loading in the Red Sea (Yahya et al., 2019). A soil dust sample from North Africa exhibited heat and peroxide sensitivity between -5 and -16 °C, further demonstrating that the IN activity of mineral dust could be associated with organic and/or biological material.

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Contrary to Price et al. (2018), who measured INP in the dust-laden Tropical Atlantic, no correlation was found between dust loading and n_{INP} or n_s. Results from this study and Gong et al. (2020) indicate that the existing n_s parameterizations alone do not skillfully represent mineral dust associated INPs at modest supercooling near major dust sources.

The source strengths of Red Sea, Mediterranean, Arabian Sea, and Arabian Gulf bulk seawater were also evaluated. The maximum source potential was observed in the Arabian Sea (50 INP mL^{-1} at -19 °C).) Overall, the observed n_{INP} range agreed well with the Gong et al. (2020) SSW measurements at Cabo Verde.

Considering that desert dust parameterizations overpredicted the n_s values observed during AQABA, despite proximity to major global emissions sources, this study demonstrates the need to evaluate the fidelity of dust INP parameterizations in nascent versus aged dust populations. The discrepancies underscore the challenges of evaluating dust-specific INP parameterizations: limited observations at modest supercooling, few assured methods for distinguishing between different INP sources in ambient aerosol, a dearth of characteristic soil dust samples from major dust sources, and limited knowledge of the specific composition and characteristics of dust INPs at temperatures > -15 °C.

In addition to providing observations at high to moderate freezing temperatures, future studies could apply the methods developed in Gong et al. (2020) to estimate the contribution of marine INPs to the aerosol sampled. Furthermore, given the combination of marine, dust, and anthropogenically-influenced air masses encountered, and the evidence of organic and biological INPs at modest supercooling in this study and Gong et al. (2020), future studies could benefit from advances in on-line Light-Induced Fluorescence (LIF) measurement techniques. Whereas the interpretation of fluorescence data from most LIF-based instruments has been limited by the lack of spectroscopic information, newer instruments support real-time spectrally-resolved size and fluorescence measurement information for single particles (Fennelly et al., 2018; Huffman et al., 2020; Könemann et al., 2019). This information could be used to "tag" different classes of organics and biological aerosols, enabling investigations of relationships between n_s, n_{INP} and organic signatures in, e.g., mineral dusts and agricultural soil dusts. Finally, the decreased n_s observed in this study further motivate comprehensive aerosol-ice nucleation studies, which aim to achieve closure between measured and predicted ambient INP concentrations by simultaneously

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characterizing ambient INPs and ice nucleation relevant properties of the total aerosol population, such as composition and aerosol chemical mixing state (Sullivan et al., 2007).

Data Availability: The data set supporting this manuscript is hosted by the UCSD Library Digital Collections (**https://doi.org/10.6075/J0X0676P**) (Beall et al., 2021).

Author contributions:

CMB, TCH, PJD, MOA, CP, JL, JS, FD, BW, HH, MDS, and KAP designed the study. CMB performed the INP measurements, FLEXPART modeling and analysis with support from TCH, PJD, MOA, MDS, MP and KAP. TCH, PJD and MOA contributed significantly to the writing, preparation of figures and analysis. TK, MI, RP and HH supported the field collection of aerosol for INP analysis and TK additionally provided aerosol number concentration data. JS and MP provided aerosol water-soluble composition data. FD oversaw the aerosol sizing and AMS composition measurements and analysis. All authors contributed to the writing of the article.

Competing interests:

The authors declare they have no conflict of interest.

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