Field Intercomparison of Ice Nucleation Measurements: The Fifth International Workshop on Ice Nucleation Phase 3 (FIN-03)

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Abstract

The third phase of the Fifth International Ice Nucleation Workshop (FIN-03) was conducted at Storm Peak Laboratory in Steamboat Springs, Colorado in September 2015 to facilitate the intercomparison of instruments measuring ice nucleating particles (INPs) in the field. Instruments included two online and four offline measurement systems for INPs, a subset of those utilized in the laboratory study that comprised the second phase of FIN (FIN-02). Composition of the total aerosols was characterized using the Particle Ablation by Laser Mass Spectrometry (PALMS) and Wideband Integrated Bioaerosol Sensor (WIBS) instruments, and aerosol size distributions were measured by a Laser Aerosol Spectrometer (LAS). The dominant total particle compositions present during FIN-03 were composed of sulfates, organic compounds, and nitrates, as well as particles derived from biomass burning. Mineral dust containing particles were ubiquitous throughout and represented 67% of supermicron particles. Total WIBS fluorescing particle concentrations for particles with diameters > 0.5 μm were 0.04±0.02 cm^{-3} (0.1 cm^{-3} highest, 0.02 cm^{-3} lowest), typical for the warm season in this region and representing ≈9% of all particles in this size range as a campaign average.

The primary focus of FIN-03 was the measurement of INP concentration via immersion freezing at temperatures > –33 °C. Additionally, some measurements were made in the deposition nucleation regime at these same temperatures, representing one of the first efforts to include both mechanisms within a field campaign. INP concentrations via immersion freezing agreed within factors ranging from nearly 1 to 5 times on average between matched (time and temperature) measurements and disagreements only rarely exceeded one order of magnitude for sampling times coordinated to within three hours. Comparisons were restricted to temperatures lower than –15 °C due to limits of detection related to sample volumes and very low INP concentrations. Outliers of

77   up to two orders of magnitude occurred between –25 °C and –18 °C; better agreement was seen at

78   higher and lower temperatures. Although the 5-10 factor agreement of INP measurements found

79   in FIN-03 aligned with the results of the FIN-02 laboratory comparison phase, giving confidence

80   in progress of this measurement field, this level of agreement still equates to temperature

81   uncertainties of 3.5 to 5 °C that may not be sufficient for numerical cloud modeling applications

82   that utilize INP information.

83   INP activity in the immersion freezing mode was generally found to be an order of

84   magnitude or more, more efficient than in the deposition regime at 95-99% water relative

85   humidity, although this limited data set should be augmented in future efforts.

86   To contextualize the study results an assessment was made of the composition of INPs

87   during the late Summer to early Fall period of this study, inferred through comparison to existing

88   ice nucleation parameterizations and through measurement of the influence of thermal and organic

89   carbon digestion treatments on immersion freezing ice nucleation activity. Consistent with other

90   studies in continental regions, biological INPs dominated at temperatures > –20 °C and sometimes

91   colder, while arable dust-like or other organic-influenced INPs were inferred to dominate at most

92   times below –20 °C.

1 Introduction

Atmospheric ice nucleation is one of the least certain aerosol-cloud interactions influencing climate (Kanji et al., 2017). Particles that physically catalyze freezing, known as ice-nucleating particles (INPs) (Vali et al., 2015), are found in the atmosphere in concentrations that span many orders of magnitude, ranging from 10^{-3} L^{-1} or fewer at -5 °C to 1000 L^{-1} or greater at -35 °C (Petters and Wright, 2015). INP number concentrations typically increase exponentially with degree of supercooling below 0 °C. However, chemical composition plays an important role in determining if, and at what temperature, individual particles may serve as INPs (Murray et al., 2012). INPs initiate the formation of ice in cold and mixed-phase clouds and in turn influence their physical and optical properties. An increase in INP concentration over a geographic area may increase the frequency of glaciated clouds at constant temperature, which in turn increases precipitation and decreases cloud lifetime (Lohmann and Feichter, 2005). Nevertheless, INP impacts on clouds simulated in global climate models are highly sensitive to how aerosol's ability to nucleate ice is parameterized (Boucher et al., 2013). Parameterizations can only be as accurate as the measurements on which they are based (e.g., Knopf et al., 2021).

Measurements of atmospheric INPs remain challenging due to the difficulty representing the physical processes involved in ice nucleation instruments. At temperatures below $\approx$ -38 °C, micrometer-sized, dilute water droplets spontaneously freeze due to homogeneous freezing nucleation. Homogeneous freezing nucleation is well understood and included in most cloud formation models. However, at temperatures between 0 and -38 °C, freezing requires INPs to facilitate nucleation through a heterogeneous nucleation mechanism (Kanji et al., 2017; Murray et al., 2012; Vali, 1985). Nucleation is hypothesized to proceed through (1) immersion freezing, which occurs when an INP embedded within a water droplet enters a cooler environment, and nucleates an ice crystal, (2) condensation freezing, which occurs when freezing ensues as an

119   aqueous droplet condenses on the surface of an aerosol particle, (3) contact freezing, which occurs

120   when an aerosol in contact with a water droplet surface initiates freezing (Durant and Shaw, 2005;

121   Fornea et al., 2009), and (4) deposition nucleation, which is thought to occur through the direct

122   deposition of water vapor on an INP surface. Of these mechanisms, immersion freezing nucleation

123   is thought to be the most active heterogeneous nucleation process in the atmosphere, though there

124   is considerable disagreement in the literature about the relative importance of other mechanisms

125   (Kanji et al., 2017; Ullrich et al., 2017). When the ambient humidity is below water saturation,

126   nucleation can occur via deposition of water from the vapor phase. In some cases, this behavior

127   may be ascribable instead to water condensation in pores and cavities in aerosols facilitating

128   freezing through a non-deposition mechanism (Marcolli, 2014; Wagner et al., 2016). However,

129   this process is unlikely to be of importance at temperatures > -38 °C (David et al., 2020), which

130   are the focus of this study. We will thus refer to ice nucleation at > –38 °C and below water

131   saturation as happening within the "deposition regime". Study of the efficiency of the deposition

132   nucleation process in comparison to immersion freezing has been limited for natural INPs.

133   Ice nucleation measurements have been made with instruments designed and built by

134   individual scientists, and more recently with commercial instruments. The ice nucleation

135   community has a history of collaborating to address instrument performance and inconsistencies

136   through participating in instrument intercomparisons, in which the custom-built instruments were

137   operated side-by-side to evaluate instrument response to the same aerosol populations. Ice

138   nucleation workshops have a history to 1967, with repetitions occurring in 1970, 1976, and 2007

139   (DeMott et al., 2011). These exercises were repeated not due to a difference in goals but due to the

140   development and improvement of new ice nucleation instrumentation and a focus on better

141   characterization of heterogeneous ice nucleation processes. An additional factor that has motivated

143   formal and informal instrument intercomparisons is growing recognition of the importance of

144   having coordinated detailed aerosol characterizations and better instruments to provide that

145   information (e.g., Coluzza et al., 2017; DeMott et al., 2011, DeMott et. al, 2018; Knopf et al, 2021;

146   Brasseur et al., 2022; Lacher et al., 2024). To compare concentrations and compositions of INPs,

147   a three-part workshop series, the Fifth International Ice Nucleation Workshop, or "FIN" was held

148   in 2014-2015. The first two phases were held at the Karlsruhe Institute of Technology's Aerosol

149   Interactions and Dynamics in the Atmosphere (AIDA) facility. FIN-01 focused on determination

150   of composition of INPs by mass spectroscopy (Shen et al., 2024), while FIN-02 entailed a

151   laboratory ice nucleation instrument comparison (DeMott et al., 2018). FIN-03, the mountaintop

152   field intercomparison of ice nucleation instruments is the focus of this manuscript. While

153   laboratory experiments can easily provide broad concentration ranges of particles of specific types

154   for testing, measurements in the ambient atmosphere are the ultimate application of INP measuring

155   systems, and the ambient atmosphere presents the most challenging measurement scenario due to

156   sometimes very low INP concentrations and a host of potential INP source compositions.

157   Ice nucleation measurements have experienced a renaissance in the past decade, resulting

158   in a proliferation in both the number of custom-built instruments and a diversification of

159   measurement techniques employed (Zenker, 2017; DeMott, 2018; Möhler, 2021). Participation in

160   FIN-02 was twice that of the previous formal international workshop intercomparison in 2007 (the

161   International Workshop on Comparing Ice Nucleation Measuring Systems, or ICS-2007 held at

162   the (AIDA) facility (Jones et al., 2011; Kanji et al., 2011). During FIN-02, online and offline

163   instruments sampling the same population of aerosolized particles reported INP concentrations

164   that generally agreed within one order of magnitude across a broad temperature range. Agreement

165   was best in tests of immersion freezing on soils, dusts and bacteria but spanned up to 2 orders of

167 magnitude (or 3 °C in temperature for the same active site density) for illite NX and K-feldspar

168 (DeMott et al., 2018). While relatively good agreement in the laboratory between different

169 measurement methods during FIN-02 represented significant progress for the atmospheric ice

170 nucleation community, intercomparisons in ambient atmospheric settings are more difficult due to

171 lower typical INP concentrations (Lacher et al., 2018) and variations in the chemistry and size of

172 source aerosol and INPs (DeMott et al., 2017; Knopf, 2021; Lacher et al., 2024; Brasseur et al.,

173 2022).

174   To evaluate how a suite of instruments operating collectively perform under the greater

175 measurement challenges of the field setting, FIN-03 was conducted from September 12 to 28, 2015

176 at Storm Peak Laboratory (SPL) in Steamboat Springs, CO, USA (Elevation: 3220 m MSL).

177 Unlike the closure studies of Knopf et al. (2021) and the similar comparative sampling studies of

178 Lacher et al. (2024), both of which occurred in regions surrounded by agricultural activities and

179 possible nearby urban influences, or the study of Brasseur et al. (2022) that was focused within a

180 boreal forest, this remote continental mountaintop site at an elevation of 3220 m provided the

181 opportunity to sample both regional and long-range INP sources within both the boundary layer

182 and free troposphere. The site is typically in the free troposphere during the nighttime and early

183 morning, and in the boundary layer from the late morning to early evening, although topography

184 and wind direction influence this timing (Collaud Coen et al., 2018). When in the free troposphere,

185 the site is more likely to reflect influences by regional or long-range transport of aerosols. For

186 example, during FIN-03, the variety of air masses that were sampled and sensed by aerosol

187 instruments included ones passing over phosphate mines in Idaho (on September 18 and 20) and

188 mined deposits of rare earth metals at Mountain Pass, CA (on September 27) (Zawadowicz et al.,

189 2017). When the convective boundary layer height reaches the elevation of the laboratory, the site

192   is likely more impacted by local/regional aerosol sources. Additionally, meteorological transitions

193   can occur (e.g., frontal boundary passage, wind direction shifts), driving changes in aerosol sources

194   that may indirectly occur in response to those changes (e.g., biological aerosols, carbonaceous

195   particles from biomass burning, and mineral/soil dust). While the constantly fluctuating

196   environmental conditions during FIN-03 added an additional challenge to the intercomparison,

197   they also provided a realistic setting for atmospheric INP measurements. In addition to adding

198   challenges, conducting the intercomparison in the presence of complex aerosols in the field

199   provided the opportunity to survey instrument response to varied particle sources.

200   Participation in FIN-03 included online continuous flow diffusion chambers (CFDCs) and

201   aerosol collections for offline INP measurements, representing a subset of the instruments that

202   operated in FIN-02 (DeMott et al., 2018). Online instruments have the advantage that the aerosol

203   being evaluated as INPs remain free-floating and unaltered, never touching a substrate nor

204   requiring shipment of samples to a laboratory. Online techniques can also monitor INP

205   concentration changes occurring over short time scales. Nevertheless, they are limited in the

206   thermodynamic conditions that can be represented over a given time frame, and they are limited

207   by volume sampling rates in assessing the low concentrations of INPs at modest supercooling.

208   Offline techniques, i.e., those in which samples are collected in the field and subsequently

209   processed in laboratory, provide the opportunity to capture large sample volumes (albeit over

210   longer time scales) and consequently assess a wider temperature range of INP activation

211   properties.

212   Since aerosol physical and chemical properties strongly influence their ability to activate

213   as INPs (Hoose and Möhler, 2012; Kanji et al., 2017; Murray et al., 2012), measurements of

214   particle sizes and composition (see Section 2) were included to lend context to the variable

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218 composition of aerosols and evaluate their potential role in ice nucleation activity. Rather than use

219 these data for attempting closure, FIN-03 focused on using data to constrain existing

220 parameterizations to diagnose INP compositions during the study period. Also, in contrast to other

221 recent studies, special effort was made to characterize deposition nucleation activity in addition to

222 immersion freezing.

2 Methods

2.1 Aerosol property measurements

225 Measurements of aerosol physical, chemical, and biological particle properties were made

226 during FIN-03 to provide context to INP measurements. Sampling manifolds, which draw air into

227 SPL from outdoors at high flow, are as follows: Inlets were located in each of the two wings of

228 SPL that frame the living area, referred to as the "instrument" laboratory (facing north) and the

229 "chemistry" laboratory (facing south). The "original" inlet system in the instrument laboratory

230 (Hallar et al. 2011; Petersen et al. 2019) feeds a nephelometer (see below) and a standard suite of

231 aerosol instruments (not operational for FIN-03). This 15 cm diameter aluminum inlet rises 4 m

232 above the roofline. At $\approx$1 m inside the laboratory, it transitions to a 15 cm horizontal manifold.

233 With a flow of $\approx$500 L min^{-1}, aerosol transmission calculations have characterized the system to

234 have a 50% upper particle size cut-off at an aerodynamic diameter of 5 μm (Hallar et al., 2011).

235 The "new" inlet system consists of two identical stainless steel, turbulent-flow, ground-based inlets

236 described by Petersen et al. (2019), which are straight and enter the laboratory vertically. One is

237 in the SPL instrument laboratory, and one is in the chemistry laboratory. These inlets that extend

238 10 m above the laboratory roof have been demonstrated to have 50% upper particle size cut-offs

239 at an aerodynamic diameter of approximately 13μm for a wind speed of 0.5 m s^{-1}. Additional

242   computational fluid dynamics simulations suggest that this size cut-off remains above 5 μm even

243   for exterior wind speeds up to 15 m s^{-1} (Petersen et al., 2019), higher than achieved at any time

244   during FIN-03 sampling. Little bias was seen in ambient aerosol sampling between the original

245   inlet system and the new, turbulent flow-based inlets based on the metric of total aerosol scattering

246   (Petersen et al., 2019). Flow rates and transfer lines to individual instruments are described after

247   the aerosol property measurements are introduced, at the conclusion of this section.

248   A Laser Aerosol Spectrometer (LAS, model 3340, TSI Inc., St. Paul, Minnesota, USA)

249   was used to measure the aerosol size distribution over the diameter range 0.089-10 μm. Aerosols

250   were assumed dry based on relative humidity always remaining below 30% when measured from

251   its sample line. Sample was drawn at 0.1 L min^{-1} and sampling was done from the turbulent flow

252   inlet system located in the SPL chemistry laboratory, as described further below. Size calibrations

253   were performed using polystyrene latex spheres (PSL, Duke Scientific). PSL diameters were

254   converted to ammonium sulfate equivalent diameters using Mie theory (Froyd et al., 2019).

255   Particle concentrations are reported as a function of equivalent ammonium sulfate diameter.

256   Volume and surface area distributions are derived assuming spherical particles. Number

257   concentrations and surface areas, further informed by aerosol composition measurements, allows

258   for connection to INP concentration predictions, and this information is used herein to

259   diagnostically infer mineral and soil dust influences on INPs during the study. We will particularly

260   reference the parameterizations of Niemand et al. (2012) that links mineral surface area to INP

261   concentrations and DeMott et al. (2015) that links dust number concentrations at sizes larger than

262   0.5 μm to INP concentrations.

263   Measurements using a three-wavelength integrating nephelometer (TSI Model-3563,

264   Shoreview, MN) also provided information on aerosol distributions via their optical properties.

266   This nephelometer is part of the National Oceanic and Atmospheric Administration Federated

267   Aerosol Network (Andrews et al., 2019). The nephelometer splits scattered light into red (700 nm),

268   green (550 nm), and blue (450 nm) wavelengths. Impactors to cut aerosols at aerodynamic sizes

269   below 1 and 10 μm are alternately used upstream of air flowing into the instrument. The

270   nephelometer sampled within the original inlet in the SPL instrument laboratory. A blunt tap from

271   this original SPL inlet manifold provided air samples to the nephelometer system via 1" i.d.

272   conductive tubing.

273   The Particle Analysis by Laser Mass Spectrometry (PALMS) instrument performed

274   measurements of the composition of 0.2 to 3.0 μm aerosol particles. The PALMS was designed

275   and operated by the National Oceanic and Atmospheric Administration (NOAA) as described in

276   Thomson et al. (2000). Particles are sampled, focused, and accelerated via an aerodynamic lens

277   inlet (Schreiner et al., 2002) before passing into a vacuum chamber where they successively pass

278   through two continuous-wave detection laser beams (532 nm Nd:YAG) and scatter light. Vacuum

279   aerodynamic diameter is determined based on the transit time. The detection signal triggers an ArF

280   excimer laser that emits a 193 nm pulse to simultaneously ablate and ionize single particles. The

281   resulting ions are analyzed with a unipolar time-of-flight mass spectrometer, which allows polarity

282   switching during the particle flight, thereby producing positive or negative ion mass spectra

283   for individual particles. PALMS spectra are classified into compositional categories, and

284   fractions are averaged over 5 min sample periods. Number, surface area, and mass concentration

285   products for the different particle types are generated by combining PALMS size-dependent

286   fractional composition data with absolute particle concentrations measured by the LAS instrument

287   (Froyd, et al. 2019; Froyd et al., 2022). When PALMS compositional concentrations are referenced

289   in the results of FIN-03 aerosol compositions in Section 3.2, they have been determined by these

290   methods.

291   The NOAA Wideband Integrated Bioaerosol Sensor, Model 4A (WIBS-4A; Droplet

292   Measurement Technologies, Longmont, CO) was used to detect fluorescent properties of

293   individual particles and assess the presence of biological particles. Measurements are presumed

294   to characterize dry particles. The WIBS-4A is described in detail elsewhere (Gabey et al., 2010;

295   Kaye et al., 2005; Perring et al., 2015) and is only briefly summarized here. As described in

296   Zawadowicz et al. (2019), the gain for the WIBS-4A used at SPL was set to detect and classify

297   particles between 0.4 and 10 μm. First, the optical diameter of particles entering the detection

298   cavity is determined by light scattered during transit through a 635 nm laser beam. This signal

299   triggers the sequential firing of two xenon flash lamps filtered to produce narrow excitation

300   wavebands centered at 280 and 370 nm. The resulting fluorescence is detected by two wideband

301   photomultiplier detectors observing 310-400 nm and 420-650 nm. Fluorescing particles were

302   categorized according to the intensity of the signal in each of three channels (channel A excitation

303   280 nm/emission 310-400 nm, channel B excitation 280 nm/emission 420-650 nm, channel C

304   excitation 370 nm/emission 420-650 nm). Particles for which the measured emission intensity in

305   only one channel met the threshold (such that the signal intensity exceeded the value equal to three

306   standard deviations above the mean) were assigned Type A, B, or C, and particles for which the

307   measured emission intensity in two or more channels met the threshold were assigned Type AB,

308   BC, BC, or ABC (Perring et al., 2015). The interpretation of particle composition according to

309   the seven WIBS-4A channels is not straightforward, as many fluorophores are active in each

310   channel, including non-biological components (Perring et al., 2015; Pöhlker et al., 2012). Channel

311   A fluorophores include biological components such as tryptophan, phenylalanine as well as

312   nonbiological components which interfere with the determination of biological content, including

313   polycyclic aromatic hydrocarbons (PAHs) (pyrene, naphthalene, phenanthrene). Biological

314   fluorophores, which produce a signal in channel C, include the reduced form of nicotinamide

315   adenine dinucleotide (NADH), nicotinamide adenine dinucleotide phosphate (NADPH), and

316   riboflavin, and potential non-biological interference in channel C may result from the presence of

317   humic acid in aerosol particles. Channel B fluorophores are not generally considered to be

318   biological in nature, though riboflavin and dry cellulose both produce signals in this channel.

319   We report WIBS-4A channel data herein under these noted caveats and further utilize these

320   data to explore links to immersion freezing biological INP concentrations, as has been done in

321   some previous efforts. Tobo (2013) previously reported relations of biological INPs acting in the

322   immersion freezing mode (measured by the Colorado State University (CSU) CFDC) to

323   fluorescent biological aerosol particles (FBAP) at sizes > 0.5 μm measured in the understory of a

324   Ponderosa pine forest in Colorado. In that work, an ultraviolet aerodynamic particle sizer (UV-

325   APS) with excitation wavelength at 355 nm and emission wavelengths 420-575 nm was used as a

326   reference for FBAP concentrations. Due to differences between the excitation and emission

327   wavelengths, UV-APS measurements correspond most closely with Type C particles detected by

328   the WIBS-4A (Healy et al., 2014). Consequently, a conservative or "low" estimate of FBAP for

329   use in the parameterization of Tobo et al. (2013) we employ herein uses the sum of C, AC, BC and

330   ABC particles. A "high" FBAP for this parameterization has also been used by Twohy et al.

331   (2016), considering all non-B-only particles (A, AB, ABC, AC, BC, C). We will use both

332   definitions in our presented results and partly justify the higher estimate because the CSU CFDC

333   assuredly does not capture all biological INPs due to the use of the upstream impactor (see below).

334   A final class of particles defined by WIBS-4A data for relation to immersion freezing INPs are

337 denoted as FP3 particles (Wright et al., 2014). FP3 particles are particles that show strong emission

338 in the 310 to 400 nm spectral band when excited by 280 nm light (A type) but are only weakly

339 represented as B and C types. A threshold of 1900 arbitrary fluorescence units in the 310 to 400

340 nm band is used to denote FP3 particles (Wright et al., 2014). FP3 particles have been connected

341 to immersion freezing INP concentrations in multiple environments (Wright et al., 2014; Suski et

342 al., 2018; Cornwell et al., 2023).

343   Flow rates and transfer lines to each instrument are summarized as follows. The PALMS,

344 LAS, and WIBS-4A sampled from the SPL turbulent flow inlet stack at 0.75, 0.1, and 0.3 vlpm,

345 respectively, via a common 0.19" i.d. aluminum tube. The total flow was held at 1.2 vlpm using a

346 variable dump flow, and the line was split into multiple 0.115" o.d. stainless steel tubing sections

347 connecting to each instrument. All tubing junctions employed Y-splitters, and all reducing fittings

348 were internally beveled to prevent impaction losses. Sample lines were not actively dried, but

349 relative humidity was < 30% in LAS and WIBS-4A. For the LAS instrument, the theoretical

350 transmission of the inlet system was 98%, 84%, and 57% for 1, 3, and 5 μm aerodynamic diameter

351 particles, respectively, with gravitational settling being the dominant loss process. Transmission

352 to WIBS-4A for the same sizes was 99%, 90%, and 76%. Size distributions were not corrected for

353 transmission losses. The nephelometer sampled from the original inlet in the SPL instrument

354 laboratory via a blunt tap manifold and 1" i.d. conductive tubing.

355 **2.2 INP measurement methods**

356   All specific instruments used in FIN-03 were also used in the FIN-02 laboratory campaign.

357 A summary listing of all ice nucleation instruments utilized in FIN-03 is provided in Table 1.

358 Detailed operating principles, siting of samplers (rooftop versus within SPL), and experimental

359 methods for each instrument follow below. In this work, we will refer to the FIN-03

366 "intercomparison period" to define the times that all INP instruments co-sampled air with

367 substantial temporal overlap for direct comparison. This means that on a given day a sample was

368 fully collected within the comparison time unit of 3 hours (informed by aerosol data, as discussed

369 later) or overlapped the comparison period if the collection time was somewhat longer. Other times

370 of sampling by the different instrument groups were devoted to special science investigations only

371 partly covered herein.

372

373 **Table 1** Descriptions of INP instruments.

	Instrument	Type	Institute	References
Online/direct	Continuous flow diffusion chamber (CSU-CFDC)	Continuous flow diffusion chamber (cylindrical)	Colorado State University	(Eidhammer et al., 2010; Rogers, 1988; Rogers et al., 2001)
	Spectrometer for ice nuclei (MIT-SPIN)	Continuous flow diffusion chamber (parallel)	Massachusetts Institute of Technology	(Garimella et al., 2016; Garimella et al., 2017; Kulkarni & Kok, 2012)
Offline/post-processing	Frankfurt Ice Nuclei Deposition Freezing Experiment deposition mode (FRIDGE-DC)	Low pressure diffusion chamber (on wafers)	Goethe University Frankfurt	(Schrod et al., 2016)
	Frankfurt Ice Nuclei Deposition Freezing Experiment immersion freezing mode (FRIDGE-CS)	Cold stage droplet freezing array (on wafers)	Goethe University Frankfurt	(Schrod et al. 2020; DeMott et al. 2018)
	Ice spectrometer (CSU-IS)	Aliquot freezing array	Colorado State University	(Hill et al., 2016; Hiranuma et al., 2015)
	Cold stage (NCSU-CS)	Cold stage droplet freezing array (on hydrophobic glass slides)	North Carolina State University	(Wright & Petters, 2013; Yadav et al., 2019)

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2.2.1 Online INP measurements

Two online instruments participated in intercomparison experiments in FIN-03. We describe the basic design and operating principles and procedures, sampling inlets, measurement uncertainties and correction for false counting issues, and any special studies reported herein for the CFDC instruments from CSU and the Massachusetts Institute of Technology (MIT). A third CFDC from Texas A&M University was used primarily for special studies to develop depolarization detection of ice, already reported (Zenker et al., 2017).

The CSU-CFDC

This online INP instrument has the most established history as an online technique for activating and counting INPs. The CSU-CFDC operating principles are described in prior works (Rogers, 1988; Rogers et al., 2001; Eidhammer et al., 2010). Application and considerations for interpreting data have been described by DeMott et al. (2018). The CSU-CFDC is composed of nested cylindrical copper walls that are chemically ebonized to be hydrophilic so they can be evenly coated with ice. The chamber is divided into two sections vertically. For FIN-03, the CSU-CFDC was operated to establish a temperature gradient between the colder (inner) and warmer (outer) ice walls in the upper $\approx$50 cm "growth" section to produce either water subsaturated or water supersaturated conditions at various temperatures within a central lamina. Aerosol particles were directed into that central lamina. For the flow rates used (10 vlpm total flow, 1.5 vlpm sample flow) the residence time was $\approx$5 s in the growth region. Ice crystals forming on INPs in the growth section continued to grow for $\approx$2 s in the lower $\approx$35 cm "evaporation" section of the chamber where the outer wall temperature was adjusted to be at an equivalent temperature to the inner (cold) wall

Moved up [3]: Online instruments have the advantage in that the aerosol being evaluated as INPs remain free-floating and unaltered, never touching a substrate nor requiring shipment of samples to a laboratory. Online techniques can also monitor INP concentration changes occurring over short time scales. Nevertheless, they are limited in the thermodynamic conditions that can be represented over a given time frame, and they are limited by volume sampling rates in assessing the low concentrations of INPs at modest supercooling. Offline techniques, i.e., those in which samples are collected in the field and subsequently processed in laboratory, provide the opportunity to capture large sample volumes (albeit over longer time scales) and consequently assess a wider temperature range of INP activation properties.

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A combination of direct-processing (online) and post-processing (offline) ice nucleation instruments were employed during the FIN-03 field campaign. All these instruments were also used in the FIN-02 laboratory campaign. Online instruments have the advantage in that the aerosol being evaluated as INPs remain free-floating and unaltered, never touching a substrate nor requiring shipment of samples to a laboratory. Online techniques can also monitor INP concentration changes occurring over short time scales. Nevertheless, they are limited in the thermodynamic conditions that can be represented over a given time frame, and they are limited by volume sampling rates in assessing the low concentrations of INPs at modest supercooling. Offline techniques, i.e., those in which samples are collected in the field and subsequently processed in laboratory, provide the opportunity to capture large sample volumes (albeit over longer time scales) and consequently assess a wider temperature range of INP activation properties. A summary listing of all ice nucleation instruments is provided in Table 1. Detailed operating principles, siting of samplers (rooftop versus within SPL), and experimental method[... [1]]

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Moved up [1]: Aerosol particles were directed into that central lamina. Ice crystals forming on INPs in the growth

477   to promote evaporation of liquid drops. When operating in the water supersaturated regime, water

478   relative humidity was controlled to be nominally at 105% during FIN-03 to stimulate droplet

479   growth and subsequent freezing, for best comparison to offline immersion freezing methods. For

480   probing ice nucleation in the deposition nucleation regime, relative humidity (RH) was controlled

481   to ≈95%. Temperature uncertainty is ± 0.5 °C at the reported CSU-CFDC lamina processing

482   temperature and relative humidity uncertainty depends inversely on temperature, as discussed by

483   DeMott et al., (2018), estimated for example as 2.4 % for a lamina RH of 105% at –25 °C.

484   Processing temperatures spanned –15 to –32 °C during FIN-03.

485   The CSU-CFDC sampled from one of the turbulent aerosol inlet ports, located in the SPL

486   instrument laboratory. Connection was via ¼" o.d (0.19" inner diameter conductive tubing. Prior

487   to entering the CFDC, aerosol was further dried using two inline diffusion driers and then size-

488   limited using dual single-jet impactors that achieve a 50% upper particle size cut-off at an

489   aerodynamic diameter of 2.5 μm. This limitation on aerosol sizes helps to remove ambiguity when

490   distinguishing ice crystals at ≈4 μm sizes from aerosol particles using an optical particle counter

491   at the CSU-CFDC outlet. Counts greater than this size divided by sample volume define INP

492   concentrations.

493   Uncertainty in calculation of INP concentrations must account for background counts that

494   can occur due to ejection of frost emanating from interior surfaces of the CSU-CFDC over

495   operational periods. We follow Levin et al. (2019) in this regard. Frost corrections are defined

496   through via use of time intervals of sampling ambient air through a HEPA filter. A typical daily

497   cycle at each temperature point was to bookend 10-min ambient air sampling with 5-min filter

498   periods. Sample data were background corrected by subtracting the interpolated filter period

499   concentration before and after each sampling period. Background corrected data were then

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510 averaged to ≈5-min sampling times to increase statistical confidence. Poisson counting errors

511 during filtered and ambient sampling periods were added in quadrature, and INP concentrations

512 were judged statistically significant at the 95% confidence level if they were greater than 1.64

513 times this combined INP error (one-tailed z test). Interior inlet tubing losses are not considered in

514 the reported INP data because they have been estimated at 10% or less in the past. INP

515 concentration correction underestimates inferred (by a factor of 3) 3 time) to be due to aerosols

516 spreading outside of the lamina during measurements specifically of mineral dust INPs (DeMott

517 et al., 2015) are not generally applied to the data herein, though this is discussed regarding the

518 intercomparison results and INP parameterizations in this paper.

519   An aerosol concentrator (MSP Model 4240) was used at selected times during FIN-03 to

520 improve sampling statistics, in the same manner as in previously published studies (Tobo et al.

521 2013; Suski et al., 2018; Cornwell et al., 2019). The aerosol concentrator was positioned open to

522 the air on the roof of the instrument laboratory room (covered and not used during rainfall), with

523 a short ¼0.19" o.d. inner diameter copper line containing the concentrated aerosol entering the

524 laboratory vertically from about 3 m above the CFDC. Concentration factors for INPs can vary

525 depending on the ambient INPs present in a given environment. These were evaluated in the same

526 manner as Tobo et al. (2013), leading to an average increase of INPs by 90 times (±45) during

527 operation of the aerosol concentrator compared to ambient inlet periods during this study (not

528 shown here because analysis repeats the efforts referenced above). A three-way manual stainless-

529 steel valve was used to direct sample air to the CSU-CFDC from either the turbulent flow inlet or

530 the aerosol concentrator.

531   Supplemental studies with the CSU-CFDC reported herein used a high temperature heating

532 tube (Suski et al., 2018) placed in-line following the three-way valve for removing aerosol organics

545 prior to INP measurements. The use of a tube heater upstream of the CSU CFDC to expose single

546 particles to 300°C is intended to isolate the action of total organic versus inorganic INPs via

547 comparison of ambient versus heat-treated particle streams. Simultaneous measurements of heated

548 and unheated aerosol streams are not possible with a single CFDC, so sampling was conducted by

549 alternating the channel chosen following a flow splitter during subsequent 10-minute periods, and

550 ignoring aerosol changes that rarely occurred over such short times.

551   The MIT Spectrometer for Ice Nuclei (SPIN)

552   The MIT-SPIN (Droplet Measurement Technologies, Boulder, CO), a commercially

553 produced, parallel-plate CFDC, also sampled during FIN-03. Measurements were focused on ice

554 nucleation below water saturation for FIN-03. Operating principles are described in Garimella et

555 al. (2016) and Garimella et al. (2017). SPIN consists of two flat walls separated by 1.0 cm and

556 coated in approximately 1.0 mm of ice. Aerosol particles are fed into the chamber in a lamina flow

557 of about 1.0 liters per minute and are constrained to the centerline with a sheath flow of about 9.0

558 liters per minute. The temperature and relative humidity that the aerosol lamina experiences were

559 controlled by varying the temperature gradient between the two iced walls (Kulkarni & Kok,

560 2012). After exiting the nucleation chamber, the particles enter SPIN's optical particle counter,

561 which sizes aerosol on a particle-by-particle basis for diameters between 0.2 and 15 μm.

562 Temperature uncertainty was 0.5 ºC. For the lamina RH conditions below 100% used in FIN-03,

563 the RH uncertainties were 0.7, 1.3 and 1.7% at –20, –25, and –30 C, respectively.

564   The MIT-SPIN sampled from one of the turbulent flow inlet systems, located within the

565 SPL aerosol chemistry laboratory. It was connected to the inlet system port with a short section of

566 0.19" inner diameter conductive tubing.

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Data processing for SPIN, including definition of uncertainties, was performed following similar procedures as used for the CSU-CFDC instrument, with a few distinctions. A cut-size for potential ice particles was set to 5 μm diameter. A low-pass filter was applied next to remove all 1 Hz data that exceeded a total of three counts s^{-1}, as recommended by Richardson et al. (2007) to reduce frost background noise that equated to INP concentrations larger than about 200 L^{-1} (>2 standard deviations above mean values discussed later) for the SPIN sampling flow rate. A depolarization filter was next applied to isolate particle data specific to ice using 1 Hz averaged backscattering data from the SPIN's OPC, with instrument specific values of 3.5 and -0.25 for the $\log_{10}$(Size) and $\log_{10}$(S1/P1) measurements respectively (Garimella et al., 2016). Ice particle data was then converted from counts per second to number density per volume of sample flow (L^{-1}). Frost ejected from the plates of the SPIN chamber beyond that removed by the low-pass filter was characterized using particle-free sampling periods when the sample flow was diverted through a HEPA filter by an automated three-way valve. Linear interpolation of filter period INP concentrations was used to approximate background frost concentrations throughout the measurement period (a minimum of 4, 5-min filter periods for each set-point temperature within a 2–3-hour period) and smoothed using a five-minute moving average. Sample data was background frost corrected by subtracting this smoothed background frost density from total number density in each 5-min sample period. Finally, a SPIN specific particle concentration correction factor of 1.4 is applied to account for non-ideal instrument behavior (e.g., out of lamina particles) resulting in underestimation of INPs as described by Garimella et al. (2017). As the field measurements from this study predate the laboratory experiments performed to determine SPIN uncertainties, the minimum reported correction factor was selected to remain conservative in reported measurements.

As for the CSU-CFDC, estimation of INP concentration measurement error for the MIT-SPIN assumed the background corrected INP concentration follows a Poisson distribution. Then, the Poisson error for both INP and background frost concentrations were defined as the square root of the sample mean. The significance test statistic was defined by the quadrature sum of counting errors multiplied by the z-score for a one-tailed z-test at the 95% confidence interval. INP measurements were deemed statistically significant if the mean INP concentration was greater than this test statistic.

2.2.2 Offline INP measurements

Offline methods have undergone many improvements in recent years and have been successfully used in a complementary manner for comparison to online methods in other recent intercomparisons (DeMott et al., 2017; DeMott et al., 2018; Hiranuma et al., 2015; Wex et al., 2015; Knopf et al., 2021; Brasseur et al., 2022; Lacher et al., 2024). In FIN-03 particles were collected from the air using liquid impingers and filter samplers. Impinger liquid and water suspensions created from immersed filters were analyzed for immersion freezing of distributed droplet volumes using the North Carolina State University Cold Stage (Wright et al., 2013), the CSU Ice Spectrometer (Hiranuma et al., 2015; DeMott et al., 2018), and the FRankfurt Ice Nuclei Deposition FreezinG Experiment (FRIDGE) instrument (Schrod et al., 2016). All measurements were made offsite after the return of impinger liquid and filters to the participant institutions, as done in most intercomparisons of this type. The handling of samples is mentioned regarding each instrument below.

The North Carolina State University Cold Stage (NCSU-CS)

683   The North Carolina State University cold stage (NCSU-CS) has been previously described

684   by Wright and Petters (2013) and Hader et al. (2014). Procedures used for collecting immersion

685   freezing spectra are described below and by Yadav et al. (2019). During FIN-03, filter samples,

686   impinger samples and precipitation samples were collected for analysis using the NCSU-CS. For

687   the intercomparison, the filter and impinger results are considered. Filter samples were collected

688   from the roof of Storm Peak Lab for 3–4 hours twice daily using 47 mm Nuclepore polycarbonate

689   filters (0.2 µm pore size) housed in an open-faced stainless-steel filter holder operated at 14 L

690   min^{-1} (at altitude) or ≈9 L min^{-1} at standard temperature and pressure conditions (STP) of 1013 mb

691   and 0 °C. Filter holders were directed downward and sheltered from precipitation by a large,

692   inverted metal bowl. Images are shown in supplemental Section S1. Each filter was resuspended

693   in 6 ml prefiltered HPLC grade ultrapure water. Impinger samples were collected directly into

694   ultrapure water using a glass bioaerosol impinger (SKC, Inc.) as described by Hader et al. (2014)

695   and DeMott et al. (2018). The impinger jets air at 10.6 L min^{-1} (≈7 L min^{-1} STP) into a 20 mL

696   water reservoir, impacting 80% of particles ≥ 200 nm in diameter and ≈100% of particles ≥ 1 µm

697   (Willeke et al., 1998). Impinger samples were collected in the same manner as was done for all

698   shared liquid samples for the FIN-02 intercomparison (DeMott et al., 2018) except that Teflon tape

699   replaced stopcock grease to seal the impinger glass lid to prevent jamming. Water evaporating

700   from the reservoir was replaced hourly; the impinger was in a rooftop shelter with its inlet

701   extending horizontally through a hole in the shelter wall, into the open air at a height of ≈6 feet

702   below the position of filter sampling units that were mounted on an outside railing. Water used

703   onsite was filtered (0.2 μm) Milli-Q water. All samples were stored at –20 °C onsite, shipped on

704   dry ice, then stored at –80 °C until analysis at NCSU..

714   Freezing statistics for each liquid sample were acquired by pipetting an array of

715 approximately 256 droplets of 1 μL ± 0.88% volume on four hydrophobic glass slides under dry

716 N$_2$ gas. Temperature was ramped at a rate of –2 °C min^{-1} and freezing was detected at a temperature

717 resolution of 0.17 °C (every 5 s) using CCD camera images collected from an optical microscope.

718 Temperature uncertainty based on repeatability of homogeneous freezing tests is 0.1 °C (Hiranuma

719 et al., 2015). Except for pure dust samples, the dependence of the population median freezing

720 temperature on cooling temperature is less than 1 °C per decade in cooling rate, including

721 measurements of ambient INPs (Wright et al., 2013). A decade in cooling rate is much larger than

722 the variations in cooling rate used by instruments in FIN-03 (-0.33 to 2 °C min^{-1}). The expected

723 shift in INP spectra due to variability in cooling rate is much less than the total uncertainty and

724 thus corrections for cooling rate are not further considered here. The concentration of ice nuclei at

725 temperature T per unit volume of liquid is given by Vali (1971):

$$c_{IN}(T) = \frac{-ln\left(f_{unfrozen}(T)\right)}{V_{drop}\Delta T} \qquad (1)$$

727 where $f_{unfrozen}$ is the fraction of unfrozen droplets at T and V_{drop} is the population-median droplet

728 volume. The concentration of ice nucleating particles (INP) in the atmosphere is given by:

$$c_{INP}(T) = \frac{c_{IN}(T) \cdot f \cdot V_{liquid}}{V_{air}} \qquad (2)$$

730 where f accounts for any serial sample dilutions with pure water used to focus measurements within

731 different temperature ranges, V_{liquid} is the liquid suspension sample volume, and V_{air} is the volume

732 of air sampled (flow rate at STP × duration). The high temperature resolution freezing data were

733 collected 3× per sample and $f_{unfrozen}$ was binned into 1 °C intervals for spectral calculations.

734 Confidence intervals reported in archived data were given as ±2 standard deviations of the mean

735 temperature uncertainty of measurements (typically 0.5 to 1 ºC). We will refer to the processed

filter samples as NCSU-CS (F) and processed impinger samples as NCSU-CS (I). Note that the filter samples were more concentrated by a factor ≈5due to the greater V_{liquid} used in the impinger for the stated air collection volumes. Thus, the filter technique is more sensitive and has a lower limit of detection (LOD). The precise ratio for a specific experimental period depended on the exact sampling times of filter and impinger, and the exact number of droplets analyzed for the filter, impinger sampling, averaging across repeats, and binning into 1-degree intervals. For this reason, the ratio of LOD for the averaged samples may differ slightly from this estimate.

As for all INP samples in FIN-03, "blanks" were collected for each of the NCSU-CS sample types. The normal procedure for most blank filter assessments in the field is to momentarily expose a clean filter to flow in a collection unit. In the spirit of procedural testing that typifies workshops like FIN-03, a different method was trialed by the NCSU group., Ten filter "blanks" were specially collected on days during FIN-03 by placing a 0.2 µm pore size filter as a backing filter to an 0.05 µm pore size filter in a secondary filter unit that was exposed to the same total ambient flow conditions as the primary INP filter unit. This 0.2 µm filter was processed the same as the primary INP filter (rinsed in 6 ml ultrapure water) and freezing results were presumed to provide a quite conservative estimate of filter background INPs. It was indeed found that the number of INPs per blank filter in these collections were much higher than for standard blank filter method used by the other groups. The results from the 10 blank filters were averaged across the processed temperature range, and an upper confidence limit of 1 °C was defined. All INP concentration results for each ambient filter were rejected if in any given temperature bin they fell below this upper confidence bound. In sum, 20% of the original measurement points based on filter collections were removed from measurement intercomparisons by this blanking operation. Impinger blanks were collected via separation of some water from the pure water storage container

767 each time the impinger unit was filled with pure water to begin an air sampling period. Thus,

768 blanks were specific to each ambient sample. The same 1 ºC upper confidence bound that

769 characterizes NCSU-CS measurements was applied in each case to identify sample temperature

770 points where the liquid suspension INPs fell below the upper confidence limit of the impinger

771 blanks. These were removed from intercomparisons.

772   CSU Ice Spectrometer (CSU-IS)

773   The CSU-IS also post-processed particles sampled onto filters during FIN-03. This

774 instrument has been described in Hiranuma et al. (2015) and Suski et al. (2018). Samples were

775 collected for approximate periods of 4 hours for intercomparison periods (longer for overnight

776 samples – not part of the intercomparison) using pre-cleaned 0.2 μm pore diameter, 47 mm

777 polycarbonate Nuclepore filters (Suski et al., 2018) mounted in disposable, sterile open-faced and

778 face-up holders (Nalgene), with a typical sample flow rate of 14.9 L min^{-1} (ambient) and 9.5 L

779 min^{-1} (STP). Filters were collected on the same exterior laboratory roof railing as the NCSU filters,

780 approximately 2 m distant. All filter samples were frozen following collection and stored at -20 ºC

781 before transit on dry ice and storage again at -20 ºC until processing at the CSU laboratory. Pre-

782 sterilization procedures and overall clean protocols for preparation and handling of filters are

783 detailed in Suski et al. (2018) and Barry et al. (2021b). Particle re-suspension was done through

784 20 minutes of shaking filters in sterile 50 mL Falcon polypropylene tubes (Corning Life Sciences)

785 with 6-10 mL of 0.02 μm pore diameter filtered deionized water. Further 20-fold dilutions using

786 filtered water were made as needed to permit measurement of freezing spectra to the low

787 temperature limit of operation of the CSU-IS.

788   Immersion freezing INP temperature spectra were obtained by distributing 24 - 32 aliquots

789 of 50 μL particle suspensions into the sterile 96-well PCR trays that mount in the CSU-IS. Other

wells were filled with serial dilution samples and pure water. The cooling rate was –0.33 °C min⁻¹. Frozen wells were counted at 0.2 - 1 °C degree intervals to a limit of about –28 °C, and cumulative numbers of INP mL⁻¹ of suspension estimated using Eq. 1. Conversion to ambient air concentrations std L⁻¹ were made based on distributed suspension volume and the total air volumes collected (Eq. 2). Several filter blanks were collected during FIN-03, and one was tested and used to obtain background INP numbers per filter. Blank INPs were found to account for <5% of INPs at –20 and –25 °C, and thus corrections were ignored. Binomial sampling confidence intervals (95%) were derived for INP concentrations following Agresti & Coull (1998). The temperature uncertainty of INP measurements is ±0.2 °C (Hiranuma et al., 2015).

As a supplemental contribution to FIN-03, portions of IS aerosol suspensions were set aside (e.g., suspensions of 6 to 8 ml can serve up to three or more IS aliquot fills) for treatments to proximally isolate total biological, other organic and inorganic contributions to measured immersion freezing INP concentrations. To assess removal of heat labile INP entities, a 2 mL aliquot of suspension was re-tested in the IS after heating to 95 °C for 20 min (McCluskey et al. 2018). To attempt to remove all organic INPs, 1 mL of 30% H_2O_2 was added to a 2 mL aliquot of suspension and the mixture heated to 95 °C for 20 min while illuminated with UVB fluorescent bulbs to generate hydroxyl radicals (residual H_2O_2 is then removed using catalase) (Suski et al. 2018), and the INPs were again assessed for freezing spectra in the IS. Herein we describe a subset of samples collected on September 15, September 23, and September 25 that were subjected as IS suspensions to the two treatments. The interpretation of data from exposure of particle suspensions to 95 °C is that the reduction of INP concentrations under thermal treatment is a proxy for the concentration of biological (proteinaceous and microbial) INPs which have been eliminated or deactivated through treatment. A strong reduction in INP activity observed after peroxide

treatment indicates dominant organic INP populations, whereas a lack of response to this treatment is assumed to indicate that inorganic INPs such as mineral dusts dominate non-heat labile INPs. This assessment for bulk suspensions of particles could be directly compared to measurements of 300 ºC heat treated single particles in the online CSU CFDC measurements on these same days, providing a more insightful investigation of INP compositions.

Taken together, such treatment studies show general utility for estimating biological contributions to INP, overall organic contributions and the importance of inorganic contributions, as done for a variety of locations (McCluskey et al., 2018; Suski et al., 2018; Barry et al., 2021a; Knopf et al., 2021; Testa et al., 2021). However, we note that not all biological materials may be completely denatured or removed by heat (Testa et al., 2021; Daily et al., 2022; Alsante et al., 2023) and not all organics may be removed by peroxide. For example, denaturation is the disruption of higher order (secondary, tertiary, and quaternary structure) in a protein which leads to a loss or lessening of function. Simpler proteins or peptides, such as glutathione, have no higher order structure, and thus cannot be denatured (Alsante et al., 2023). Consequently, estimates of biological contributions to INP based on these treatments may be considered as lower limits for the FIN-03 samples analyzed.

FRIDGE Cold Stage and Deposition Nucleation Measurements

The FRIDGE instrument can operate as a low temperature cold chamber or low temperature and pressure diffusion chamber device for measuring the concentration of INPs by two independent methods: a) a droplet freezing assay on a cold stage, hereafter FRIDGE-CS (Schrod et al., 2020; DeMott et al. 2018; Hiranuma et al. 2015) which addresses immersion freezing similarly to the NCSU-CS and the CSU-IS and b) the diffusion chamber method, hereafter FRIDGE-DC, that addresses the deposition nucleation and condensation freezing modes

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864 introduced in Schrod et al. (2016) and is the standard method for operating the FRIDGE device

865 (e.g., DeMott et al, 2018). The ice nucleation analysis is performed inside the FRIDGE instrument

866 for both methods, yet the sampling process, addressed nucleation modes and the specific analytical

867 procedures differ, as described below.

868   For the FRIDGE-CS method, aerosol particles were sampled via a short ¼" conductive

869 tube from the shared turbulent flow aerosol inlet in the SPL instrument laboratory on Teflon

870 membrane filters (Fluoropore PTFE, 47 mm, 0.2 μm, Merck Millipore Ltd.). The sampling

871 duration ranged from 50 to 240 minutes, resulting in air volumes between 250 and 1000 std. L.

872 The particles were extracted in 10 ml deionized water by shaking. Approximately 150, 0.5 μL

873 droplets from that solution were pipetted onto a clean, silanized silicon wafer on the cold stage of

874 the FRIDGE instrument and cooled by $-1°C$ min^{-1} at ambient pressure. A CCD camera detects

875 freezing events and counts the number of frozen droplets as a function of temperature. This process

876 is repeated with fresh droplets and fresh substrates until approx. 1000 droplets are attained. The

877 INP number concentration is derived using Eqs. 1 and 2, as for the NCSU-CS and CSU-IS. An

878 upper bound on temperature uncertainty is estimated as +/-0.5 ºC. Binomial sampling confidence

879 intervals (95%) were derived for INP concentrations as done for the CSU-IS, following Agresti &

880 Coull (1998). Pure water and suspensions of blank filters in pure water showed no freezing at

881 temperatures > -20 ºC and a contribution of no more than 15% toward total INPs at -29 ºC, the

882 lowest temperature for which data are reported herein. Consequently, corrections were ignored for

883 this intercomparison.

884   For the FRIDGE-DC measurements, particles were collected using an electrostatic aerosol

885 collector (EAC) (Schrod et al., 2016) was connected to the same aerosol flow inlet via a short ¼"

886 conductive tube. Within the EAC aerosol particles are electrostatically precipitated onto silicon

wafers, which are used as sample substrates. After sampling is completed, the analysis at select pairs of temperature and relative humidity set points follows in a separate step. For that, the wafer is placed on the cold stage inside a diffusion chamber. After the chamber is evacuated, the temperature is set to the first analysis temperature. In a second, much larger volume, pure water vapor is regulated by pressure control to the desired supersaturation. Once the water vapor diffuses into the chamber, ice forms on the activated INPs and a CCD camera is used to record and count the emerging ice crystals, which appear as bright objects. It is assumed that one ice crystal represents one INP. The water vapor atmosphere and thus the growth of ice crystals is maintained for up to 100 seconds until the valve to the water vapor source is closed and the chamber is evacuated again. The process is repeated at increasing humidity first, and then at progressively lower temperatures. At SPL samples were taken with the EAC for 50, 75 and 120 minutes, resulting in volumes of approximately 64-150 sL. The samples were analyzed by default at –20 °C, –25 °C and –30°C and 95 %, 99% and 102% water saturation. In addition, a few samples were analyzed at –15 °C. This was a supplemental contribution by the FRIDGE group for comprehensive analysis of INP activation in the deposition regime, and for comparison to online data in this regime collected for some days. Temperature uncertainty is the same as for the FRIDGE-CS method. RH uncertainty is +/-2% on the basis of observing visible condensation on particles at 100% RH. INP concentration uncertainties are given as binomial confidence limits, the same as for the CSU-IS.

2.3 INP processing and sampling strategies

As a campaign strategy, samples were collected over different time periods in the day to reflect both varied weather conditions and aerosol populations arriving at the mountain laboratory. For intercomparison, a select number of 3 to 4-hour sampling periods were allocated in which

928 online instruments nominally operated at a few predesignated temperature and relative humidity

929 ranges, while samples were collected continuously for off-line analysis. While aerosol conditions

930 can change within a 4-hour time frame, this was agreed upon as a minimal reasonable period for

931 comparability to obtain statistically reliable results. Similar sampling strategies have been

932 employed in the past intercomparisons (DeMott et al., 2017; Knopf et al., 2021). Overall,

933 measurements were conducted over a wide range of temperatures (–7 to –34 °C) in the

934 heterogeneous ice nucleation regime.

935 **3 Results and discussion**

936 **3.1 Meteorological context**

937   Weather conditions during FIN-03 were characterized using auxiliary measurements.

938 Weather data (temperature, humidity, winds and pressure) were obtained for Storm Peak

939 Laboratory through the MesoWest (https://mesowest.utah.edu/cgi-

940 bin/droman/meso_base_dyn.cgi?stn=STORM) mesonet (STORM site), supplemented with

941 measurements from instruments operated at SPL through the Western Regional Climate Center

942 (WRCC) (https://wrcc.dri.edu/weather/strm.html) for the two days that were absent in the

943 MesoWest record. Air temperature, relative humidity, and barometric pressure time series are

944 shown in Figure 1(a), 1(b) and 1(c), respectively. Precipitation was measured via a rain gauge at

945 Storm Peak Laboratory provided by NCSU. Precipitation rate was calculated from the quotient of

946 precipitation (in mm) and time collected (in hours), as shown in Figure 1(d). Back trajectories for

947 all the sampling days in FIN-03 are reported by Zawadowicz et al. (2017), showing 72-hr air mass

948 transits from regions that included Southern California, Washington State and Eastern Nebraska.

949   Relatively warm, dry conditions were observed initially at the Storm Peak Laboratory.

950 Clear skies on September 11 and 12, 2015 gave way to clouds and haze on September 13. Cooler

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960 temperatures, lower barometric pressure, and higher relative humidity (generally above > 70%)

961 accompanied rainfall on September 14. This was followed by continued rain on September 15,

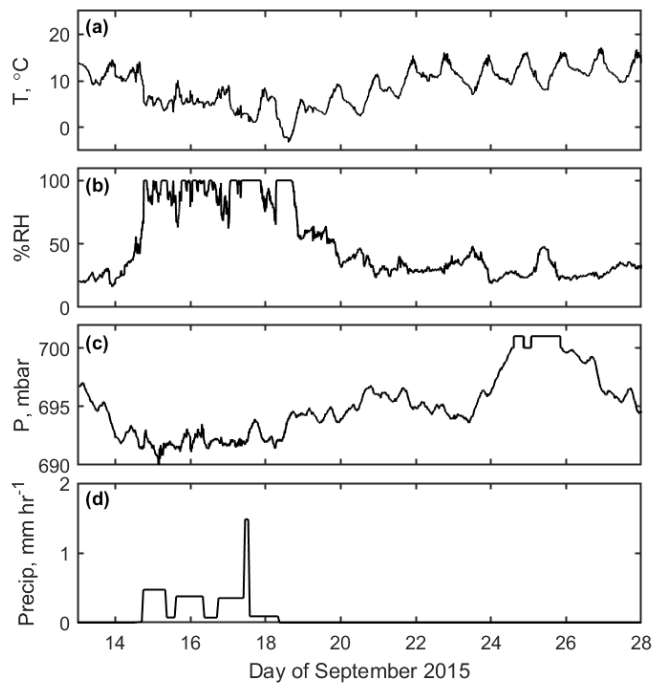
962 intermittent rain and short periods of hail on September 16, a mixture of rain, snow, and sleet on

963 September 17, and snow on September 18. The next and longest period in the study, September

964 19 to 28, was marked by an increase in temperature, an increase in barometric pressure, lower

965 relative humidity, and a lack of precipitation. More detailed weather records including daily

966 photographs and a summary of human-produced daily observations are summarized in

967 supplemental Section S1. Daily wind rose plots are provided in Figure S1.

968

969 **Figure 1.** Weather conditions over the course of FIN-03, including (a) air temperature, (b) relative

970 humidity, (c) barometric pressure, and (d) precipitation rate.

971

3.2 Aerosol context

3.2.1 *Aerosol size distribution and surface area*

The time series of aerosol size distribution measured by the LAS (in three hour means) is shown in Figure 2a. The maximum and minimum total LAS concentrations were 706 cm^{-3} and 74 cm^{-3} respectively, and the mean and standard deviation of the total LAS concentration throughout FIN-03 were 410 cm^{-3} and 138 cm^{-3}, respectively. The highest total LAS concentration recorded during FIN-03 (706 cm^{-3}) occurred in the early hours on September 25. Elevated aerosol concentration (at least one standard deviation above the mean) was also observed during midday on September 13, before and during midday on September 14, before midday on September 25, in the afternoon on September 26, and around midday on September 27.

The timeline of LAS aerosol surface area in Figure 2b emphasizes that surface area was predominately submicron throughout the study, with a mode at about 0.16 μm. This is important to note, in combination with chemical composition information discussed in the next section because it is relevant to understanding the likely sizes and surface areas of INPs. We will revisit the surface area of INPs for use in parameterizations in a later section. Quantitative timelines of

1005

1006 **Figure 2.** Time series of dry particle number concentration distribution (ambient conditions, not STP)

1007 measured by the laser aerosol spectrometer (LAS) in a), shown as three-hour means at ambient pressure.

1008 Time series of particle surface area distribution is in b). c) Timeline of nephelometer scattering (1-hr data)

1009 in the red channel for < 1 μm and 1 - 10 μm size ranges, 3-hr LAS number concentration > 0.5 μm, 3-hr

1010 LAS surface area at sizes below and above 0.5 μm, and Angström exponent (dashed, right axis).

1011

1013 LAS surface area above and below 0.5 µm are shown in Figure 2c. Surface area at above 0.5 µm

1014 is about a factor of 30 lower than at below this size over most of the study period. Also shown in

1015 Figure 2c is nephelometer scattering (b_{sp}) in the red channel (700 nm) showing a dominant

1016 contribution when the upstream impactor was set to 1 µm (aerodynamic) and a much lower level

1017 of 1 – 10 µm scattering. This scattering from coarse mode particles is consistent with and trends

1018 with the LAS surface area in the supermicron regime, while the Angström exponent (calculated

1019 using red and blue channels) being close to 2 (small particle dominance) throughout the study is

1020 consistent with the dominance of submicron contributions to total surface area. Figure 2 also

1021 emphasizes that the lowest aerosol concentrations and surface areas occurred during varied time

1022 in the wet period of the study from midday on the 14th through the 17th of September. Finally,

1023 adjacent 3-hr periods rarely represented surface area changes of more than a factor of 2 in the size

1024 range > 0.5 um and was usually within 10-20%. Large differences across 3-hour periods were less

1025 frequent for surface area at smaller sizes. These factors confirm the validity of the selected

1026 intercomparison time periods.

1027 **3.2.2 *Aerosol composition***

1028   The number concentration of aerosol particles from 0.2 to 3 µm with characteristic spectra

1029 belonging to eight composition categories (sulfate/organic/nitrate, biomass burning, elemental

1030 carbon, sea salt, mineral dust, meteoric, alkali salt, and fuel oil combustion), and the number

1031 concentration of unclassified aerosol particles by the PALMS, were assessed for three-hour

1032 averages through the FIN-03 period. For simplicity, four of these categories (elemental carbon,

1033 meteoric, alkali salt, and fuel oil combustion) were combined into a category called "other" due to

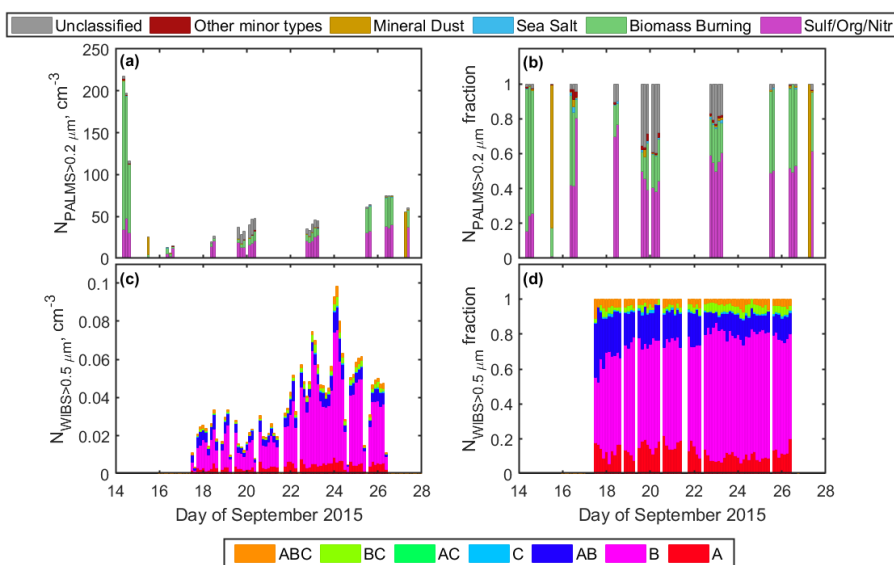
1034 the low concentration of particles in each of these categories resulting in 6 total classifications

1035 (SulfOrgNit = sulfates/organics/nitrates, BioBurn = products of biomass burning, Sea salt, Mineral

1037   dust, and Unclassified), as shown in Figure 3a. The three-hour averages of the number fractions of

1038   each particle type were also calculated as the fraction of the total aerosol number concentration

1039   measured by the PALMS in each of the six classifications, as shown in Figure 3b. The dominant

1040   categories throughout the FIN-03

1041

1042   **Figure 3**. Subplots (a) and (b) show the aerosol particle number (ambient conditions, not STP) and relative

1043   fractions (by cumulative count at all sizes) of each of the six PALMS compositional particle types for the

1044   three-hour periods during which the PALMS was used to sample ambient air. Subplots (c) and (d) show

1045   the aerosol particle number concentration and relative fractions (by count) of particles with diameter > 0.5

1046 μm in each of the channels (A, B, AB, C, AC, BC, and ABC, which are described in Perring et al., 2015)

1047   over the course of the FIN-03 field campaign.

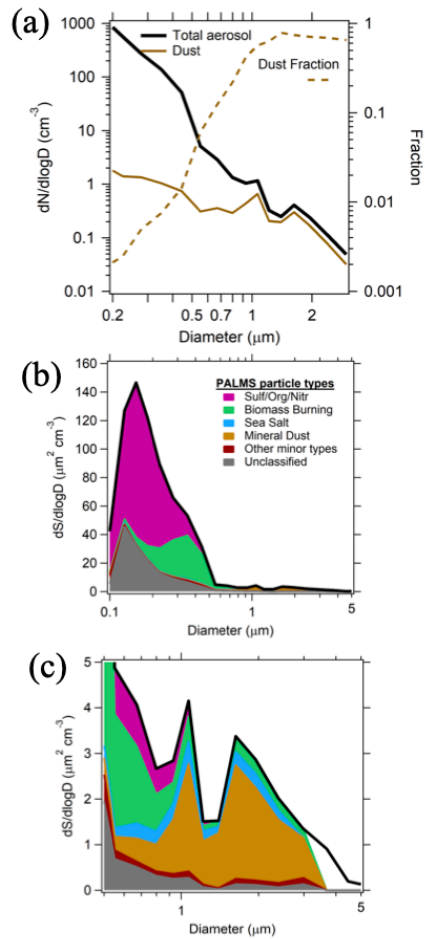
1048

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Figure 4. a) Total aerosol versus mineral/soil dust (ambient) number size distribution and dust fraction

interpreted from PALMS and LAS data for all times that the PALMS was sampling during FIN-03. b)

Surface area distribution differentiated for PALMS compositional types during the same sampling times.

c) Expanded plot from b) for the coarse mode size range to emphasize progressive dominance of dust

components at diameters > 0.5 µm.

campaign were BioBurn (mean 26 ± 43 cm^{-3}, maximum 177 cm^{-3}), SulfOrgNit (mean 22 ± 13 cm^{-3}, maximum 48 cm^{-3}), and mineral dust (mean 3 ± 11 cm^{-3}, maximum 55 cm^{-3}). The mineral dust type also includes soil particles (crustal species mixed with organic material) (Zawadowicz et al., 2019). The highest total particle number concentration measured by the PALMS (218 cm^{-3}) occurred on September 14 (of which 177 cm^{-3} consisted of biomass burning and 34 cm^{-3} consisted of sulfates/organics/nitrates). This biomass burning plume impacted the site for several hours. Mineral/soil dust particles were ubiquitous throughout the study, with a concentration of 0.128 ± 0.446 cm^{-3} (median and interquartile range). Anomalous concentrations >10 cm^{-3} observed for a few 5-min sample periods are likely due to road dust emitted from site. Dust concentrations were <1 cm^{-3} for 90% of the PALMS samples. Mineral/soil dust represented a median of 0.3% of particles in the >0.2 µm size range, increasing to 23% and 67% for >0.5 and >1.0 µm particles (Figure 4a). Similarly, mineral dust contributions to total surface area are inconsequential for total aerosol (Figure 4b) but dominate in the coarse mode regime for the study (Figure 4c). We revisit this result in discussions of parameterization of INPs in Section 3.5.

The daily average number concentration of fluorescing aerosol particles corresponding with each of the seven WIBS-4A types with diameter > 0.5 µm is shown in Figure 3(c), and the daily average number fraction of each WIBS-4A type is shown in Figure 3(d). The dominant types of fluorescent aerosol particles throughout the FIN-03 field campaign were types B, AB, and A, which on average accounted for 63.2%±8.7%, 16.0%±6.3%, and 12.5%±3.9% of the particles detected by the WIBS respectively.

In contrast with the daily average number fraction in each PALMS category, the relative contributions of each of the seven WIBS-4A particle types did not vary much over the course of the study when the WIBS-4A was operational, with perhaps the exception that Type AB decreased

1079   in prevalence from September 18 (42.9%) to September 21 (10.1%). A modest trend from lower

1080   total fluorescing particle concentrations (0.02 to 0.04 cm^{-3} at STP) from September 17 through the

1081   21st to higher concentrations (0.07 to 0.15 cm^{-3} at STP) from September 22 through the 26th. WIBS-

1082   4A data was not collected on September 13-16, nor on September 27. The first period was

1083   somewhat critical to evaluating INP relations to bioaerosols, so we note here in advance this

1084   caveat. Time-resolved size distributions for each WIBS-4A channel, as well as the total particle

1085   concentration measured across these seven channels, are shown in supplemental Figure S2. FBAP

1086   assignments related to INP predictions will be discussed in Section 3.5.

1087   **3.3   Immersion freezing measurements**

1088   A summary of the number concentrations of immersion freezing INPs (N_{INP}) over the

1089   course of the field campaign, for all measurements averaged at one degree temperature intervals

1090   for each instrument, is shown in Figure 5. The concentration of INPs detected over this range

1091   ranged over five orders of magnitude (0.01 to 160 L^{-1}). Only two sets of instruments were able to

1092   explore the temperature regimes of –30 °C and colder due their design to permit operation there,

1093   or warmer than –15 °C due to detection limits (controlled by sample volume and drop size used

1094   for immersion freezing). At any one temperature, differences up to a little more than one order of

1095   magnitude are apparent in comparing average data from individual methods, mirroring results

1096   presented in previous laboratory and field studies (Hiranuma et al., 2015; DeMott et al., 2017,

1097   2018; Knopf et al., 2021; Brasseur et al., 2022; Lacher et al., 2024).

1098   As expected, a trend of increasing N_{INP} with decreasing temperature was observed for the

1099   FRIDGE-CS, CSU-IS, NCSU-CS (I and F), and CSU-CFDC. Incremental changes in N_{INP} with

1100   decreasing temperature was similar for all measurements that spanned a broad temperature

1101   range. The dependence of N_{INP} on temperature is nearly log-linear from –10 to –27 °C, excepting

1109 perhaps a steepening of slope from –20 to –25 °C and some lowering of slope below this

1110 temperature. This comparability of dN$_{INP}$/dT contrasts with an apparent increasing high bias of

1111 drop suspension freezing measurements versus CFDC measurements

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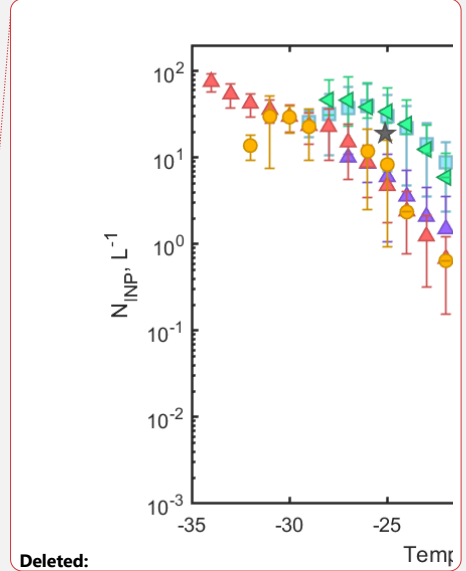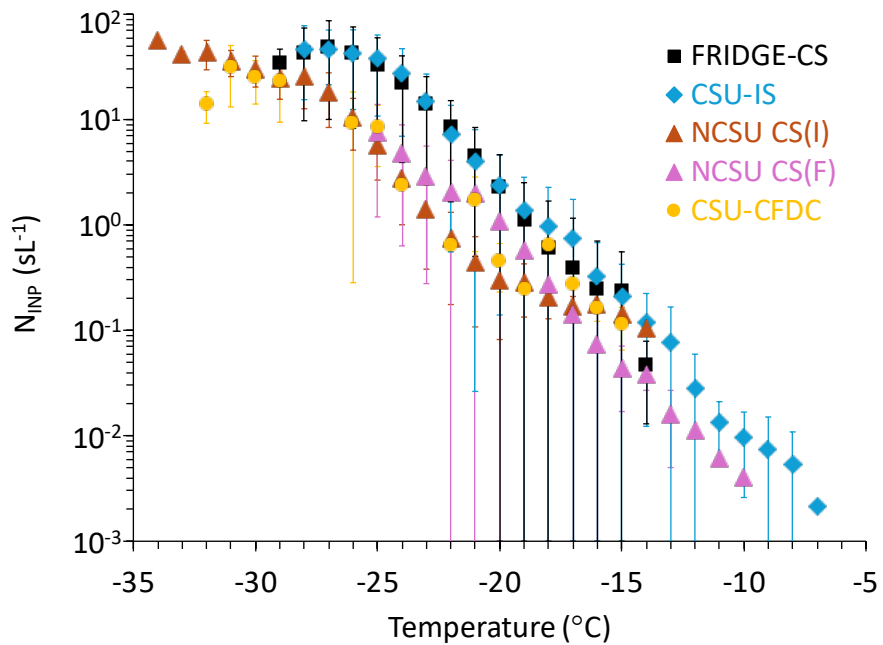
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Figure 5. Campaign average immersion freezing INP concentrations (sL^{-1}) in 1 °C bins for instruments

1114 participating in intercomparison studies. Error bars represent one standard deviation in the measurement

1115 means collected at the specified temperature and not measurement uncertainties. The error bars strike the

1116 lower axis when the standard deviation exceeded the mean values. The times over which the INP

1117 concentration has been averaged for each instrument is explained in the text.

1118

1119 during comparable sampling at various surface sites (non-mountaintop or free troposphere)

1120 found in DeMott et al. (2017) but agrees with FIN-02 laboratory studies (DeMott et al., 2018)

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1126   and recent atmospheric studies at Puy de Dome (Lacher et al., 2024). INP concentration

1127   variability at single temperatures, reflected in Figure 5 as a standard deviation of bin means, is

1128   likely due to variations in aerosol properties affecting INPs in response to production and

1129   scavenging processes upstream of the site. Nevertheless, generally higher N_{INP} measurements

1130   were obtained with the FRIDGE-CS and the CSU-IS than the CSU-CFDC and NCSU-CS (F)

1131   and NCSU-CS (I) analyses. Such biases in other studies have been attributed to different

1132   efficiencies in sampling of largest particles (e.g., Lacher et al., 2024; Cornwell et al., 2023), but

1133   the collection methods for offline measurements in this study were substantially similar, as

1134   discussed further below. Hence, we cannot attribute measurement differences to a systematic

1135   source. Comparability of impinger versus filter sampling methods for immersion freezing

1136   measurements via the NCSU-CS mirrors the findings in DeMott et al. (2017), suggesting that

1137   particle removal from filters can be highly effective for immersion freezing measurements of

1138   ambient particles.

1139   To view the data in a more complete manner over the entire project, we explore direct

1140   comparisons of different instrument data as scatterplots and measurement ratios on temporal

1141   bases. First, in Figure 6, we show a commonly used representation of large INP project data as

1142   INP concentrations for four instruments versus one other and segregate the data into broad 4-

1143   degree temperature ranges. The data used for normalization were from the CSU-IS, though we

1144   might have used any other. Linear regressions were plotted in Figure 6 to show the overall

1145   average differences between measurements that are already evident in Figure 5. Figure 6a

1146   thereby demonstrates the generally good correspondence between the NCSU-CS data of both

1147   types and the CSU-CFDC data that measure factors of 5 to 8 lower INP concentrations on

1148   average compared to the CSU-IS, as well as the closer correspondence of the FRIDGE-CS (31%

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lower) and CSU-IS data. Greatest variations in INP concentrations over the course of the project

were focused in the –20 to –25 °C temperature regime (Figure 6b), where variations reached

nearly two orders of magnitude. This is not an uncommon observation, also seen in Lacher et al.

(2024). Surprising, but not easily understood yet, is the fact that all measurement methods could

at times measure equivalently to or more than the CSU-IS.
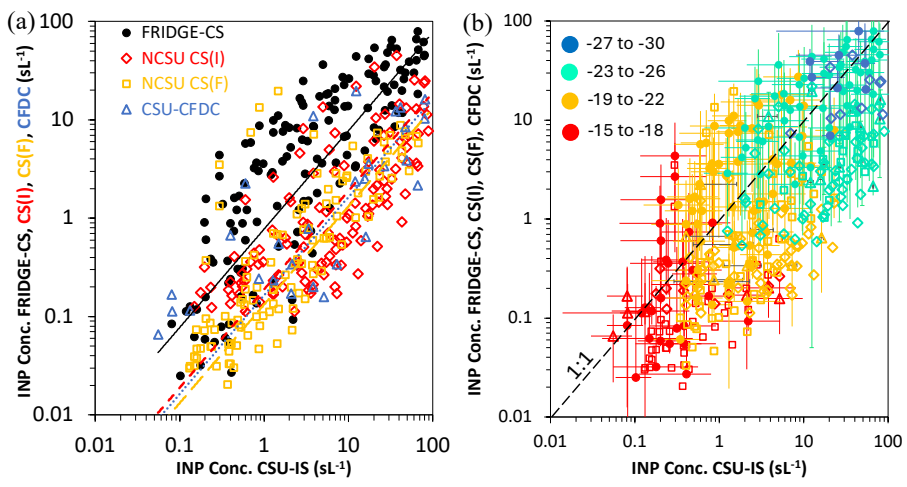
Figure 6. (a) INP concentrations for all intercomparison measurement points of FIN-03 from the FRIDGE-

CS, NCSU-CS (I), NCSU-CS (F) and CSU-CFDC compared to the INP concentrations from the CSU-IS

measurements. Linear regressions with zero intercepts are color coded for each, having slopes of 0.78, 0.19,

0.13 and 0.16 for the FRIDGE-CS, NCSU-CS (I) and CSU-CFDC, respectively. (b) The same data are

color coded for different temperature ranges in °C and the 1:1 relation is shown. Errors are confidence

intervals for FRIDGE-CS, CSU-CFDC, and CSU-IS data. These are not shown for the NCSU-CS data since

these are given as temperature errors and would need interpolation to plot as N_{INP} errors.
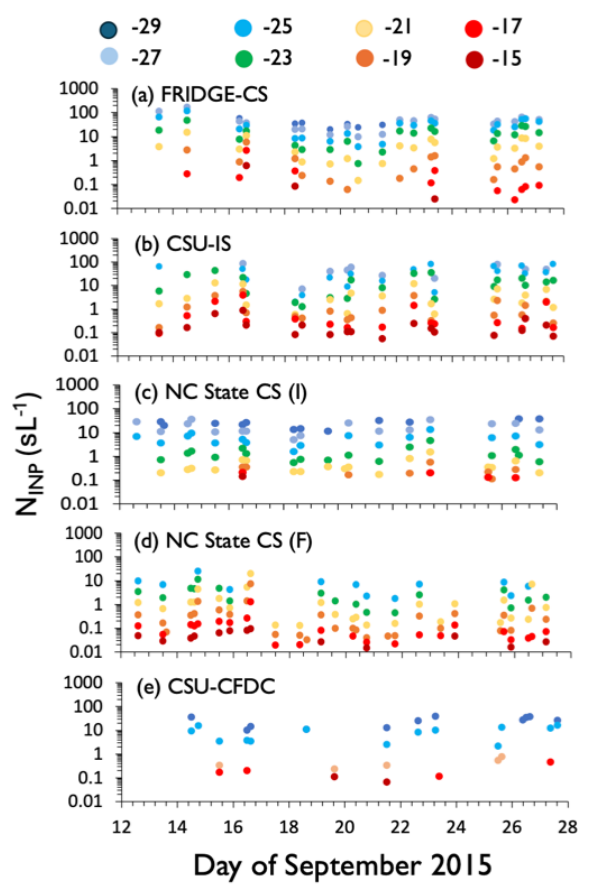
Figure 7. Time series of immersion-freezing mode INP concentrations (sL^{-1}) measured during intercomparison periods by (a) the FRIDGE-CS, (b) the CSU-IS, (c) the NC State CS (I), (d) the NC State CS (F), and (e) the CSU-CFDC. An additional data point from the MIT-SPIN is shown as a square data point in the CSU-CFDC panel. Note that data for the CFDC is plotted only for the most common temperatures of -30. -25, -20 and -15 °C. INP concentrations shown in this figure are those measured within three-hour blocks of time but may capture longer or shorter time periods depending on the specific instrument sampling time that overlapped these periods.

1178

1179   Temporal data provided further descriptions of instrument comparability. Immersion

1180 freezing N_{INP} in 1 °C bins were compared for periods of the day broken into three-hour intervals

1181 in the time series of Figure 7. While absolute INP concentration magnitudes differ, it is not

1182 difficult to see comparability of general trends amongst the data sets, albeit with episodic

1183 discrepancies that will be discussed further below. For example, all methods measure higher INP

1184 concentrations early in the study, a low point around the 18th of September and a build up again

1185 toward the end of the study. For example, INP concentrations at temperatures > –20 ºC were at a

1186 maximum during the precipitation period, as might be expected for rainfall production of

1187 biological INPs (Huffman et al., 2013; Mignani et al., 2021; Testa et al., 2021; Cornwell et al.,

1188 2023), while the strongest differences between the concentrations of INPs active at higher and

1189 lower temperatures occurred for all instrumental measurements during the period of warming

1190 under high pressure later in the study. The latter observation might be expected for a strong

1191 contribution of dust-like INPs, with a steeper dN_{INP}/dT. These positive points suggesting that the

1192 instruments were measuring the same INP cycles was also seen in the study of Lacher et al.

1193 (2024), c.f., their Figure 4.

1194   Periods of agreement and discrepancy are clearer in examining the ratios of time-matched

1195 and temperature-matched three-hour immersion N_{INP} values that were calculated for each pair of

1196 instruments, as shown in Figure 8. Numbers of overlapping measurement periods, their

1197 geometric means, standard deviations and normal 95% confidence intervals of all ratios (all

1198 times and temperatures) plotted in each panel of Figure 8 are documented in Table 2. Reiterating

1199 what is apparent from campaign-wide results in Figure 5 and 6, Figure 8 indicates the best

1200 agreement for short-term periods throughout the study was observed between the FRIDGE-CS

1209 and the CSU-IS, in which only 4 out of 146 3-hour, time- and temperature-matched N_{INP} (3%)

1210 did not agree within an order of magnitude. Nevertheless, discrepancies of a few to several times

1211 did occur from September 16th onward, focused most often at >–22°C. These biases flipped in

1212 both directions, with the CSU-IS measuring higher from the 19th to the 22nd and the FRIDGE-CS

1213 higher at some other times, notably the 16th, 23rd and 26th of September. None of these periods

1214 were distinguished in any discernible manner by weather or aerosol properties. For example,

1215 LAS and PALMS concentrations were no more than 20% different from the FIN-03 campaign

1216 means during any of these periods. Aerosol surface areas were about a factor of two lower

1217 overall during the 19th to 22nd period than for the period after the 23rd (Figure 2), which does not

1218 imply a special sampling bias for larger particles for the IS filter that was open to the air, a point

1219 we will discuss further below.

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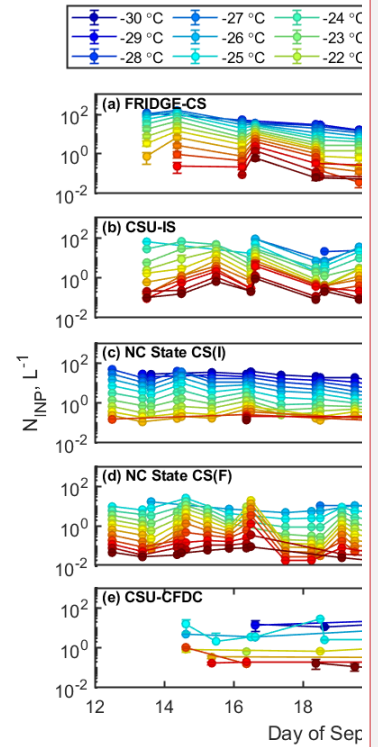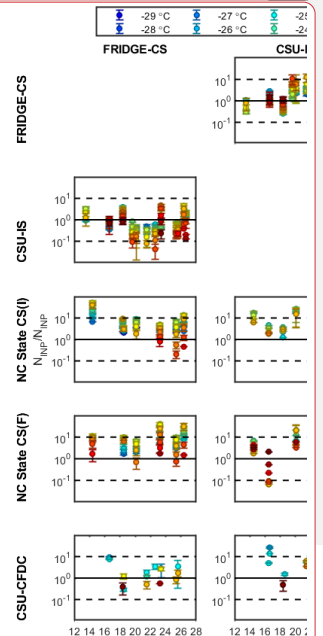
Figure 8. Ratios of the immersion freezing INP concentrations measured by each instrument, to the immersion INP concentrations measured by each other instrument (three-hour averages). Each instrument (FRIDGE, CSU-IS, NC State-CS (I), NC State-CS (F), and CSU-CFDC) is represented by one of the five columns as well as one of the five rows.

Both the FRIDGE-CS and CSU-IS showed high bias from a few to more than 10 times versus NCSU-CS(I) or CS(F), primarily at processing temperatures below –20 °C, whereas ratios closer to 1 indicated much better agreement at >–20 °C later in the study. The poorest agreement overall was observed for the CSU-IS compared to the NCSU-CS(I), a combination for which 26 out of 128 (20%) immersion N_{INP} means did not agree within an order of magnitude. Agreement between the FRIDGE-CS and the NCSU-CS(I) was only slightly better, as 15 out of 108 (14%) time-matched N_{INP} means did not agree within an order of magnitude. Higher than order of magnitude such discrepancies at lower temperatures were markedly present on September 13, 14, 23 and 26. Based on PALMS data, the 14th was richer in compounds from biomass burning, poorer in sulfates, organics, and nitrates, and slightly poorer in mineral dust than average, as discussed in Section 3.2. The concentration of > 0.5 μm particles measured by the LAS during this time was also relatively high (2.5 cm^{-3} compared to the campaign mean 0.45±0.62 cm^{-3}). However, the 14th is not markedly distinguished overall in the timeline of all INP measurements in Figure 7, so perturbations to composition and concentrations of all particle sizes due to the biomass burning event did not appear to specially perturb the INP populations. We have already noted that the 23rd and 26th of September had aerosol populations that were not much different than the project mean on those days.

The CSU-CFDC INP measurements generally agreed with the other measurements within an order of magnitude for data collected on the same day and temperature, excepting a particularly low bias versus the CSU-IS at higher temperatures on the 16th of September (rain and hail day) and at lower temperatures on the 25th of the month. Nevertheless, its measurements of INP concentration were in best agreement with all methods overall for temperatures > –20 °C, albeit for the most limited number of matches (19 to 29). CSU-CFDC INP concentrations also tended to be lower than those from the FRIDGE-CS and CSU-IS at temperatures below –20 °C. A similar divergence in online versus offline N_{INP} measurements in this temperature range was reported by DeMott et al (2017) for ground-based sampling, with online measurements tending to measure progressively lower INPs than offline integrated filter or impinger collections at below –20 °C, approaching one order of magnitude below –25 °C. At the Puy de Dome Mountain station (Lacher et al. 2024), only modest and insignificant underestimates were made by the CSU-CFDC (also using a 2.5 μm upstream impactor) versus offline INP concentrations when all were measured from a PM10 inlet. CSU-CFDC INP measurements were comparable on average with measurements from the NCSU-CS(I) and NCSU-CS(F), consistent with the mean results shown in Figure 5.

Comparing the timeline of ratios of NCSU-CS(I) to NCSU-CS(F), only 3 out of 87 (3%) of the INP concentrations obtained through analysis by the identical off-line apparatus differed by more than an order of magnitude.

Despite the discrepancies noted in the time- and temperature-matched data, a more positive message from the intercomparison is that the mean N_{INP} reported by different instruments for all temperature conditions taken together generally fell well within a span of one order of magnitude. Figure S3 (values provided in Table 3) shows the percent of immersion INP

1400 measurements in which all instrument pairs agreed within one order of magnitude. This is also

1401 consistent with the representation shown in Figure 6 for which linear regressions imply that the

1402 CSU-IS measured N_{INP} a factor of 1.4 to 8 times higher than other methods. Similarly, and

1403 importantly, the geometric mean ratios for Figure 8 listed in Table 2 were below a factor of about

1404 5 in all cases. This level of agreement compares well with the findings from FIN-02, for which

1405 the immersion N_{INP} measured by several online and offline instruments agreed within an order of

1406 magnitude. This is encouraging given that FIN-02 was a laboratory intercomparison on single

1407 composition aerosol samples consisting of particles with diameter < 2 μm whereas FIN-03 was a

1408 field campaign in which temporal changes in the concentration, size distribution, and

1409 composition of INPs at Storm Peak Laboratory were all potential factors. This level of

1410 correspondence shows that field data can be collected with nearly the same level of accuracy as

1411 laboratory experiments. While also mimicking the results of DeMott et al. (2017) for a smaller

1412 instrument comparison exercise, agreement was quite similar to that found in another recent

1413 intercomparison where INP concentrations measured by multiple systems were found to match

1414 within a factor of 5 (Lacher et al., 2024).

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Table 2. Count number (N), geometric mean, standard deviation (St. dev.), and 95% normal confidence intervals (CI) for the N_{INP} ratio data of Figure 8 in the main manuscript, including all temperature points. As for that figure, numerator instrument is on the upper horizontal scale and denominator instrument is listed on the vertical scale.

		FRIDGE-CS	CSU-IS	NCSU-CS(I)	NCSU-CS(F)	CSU-CFDC
FRIDGE-CS	**N** **Mean** **St. dev.** **CI**		146 0.93 2.86 0.46	107 0.20 0.57 0.10	90 0.26 0.43 0.09	20 0.52 1.12 0.49
CSU-IS	**N** **Mean** **St. dev.** **CI**	146 1.07 2.41 0.39		128 0.19 0.52 0.09	112 0.21 2.39 0.44	29 0.26 0.92 0.34
NCSU-CS(I)	**N** **Mean** **St. dev.** **CI**	107 4.99 9.85 1.87	128 5.40 9.41 1.63		83 1.49 5.03 1.08	28 0.97 1.11 0.41
NCSU-CS(F)	**N** **Mean** **St. dev.** **CI**	94 3.81 7.78 1.60	112 4.80 5.47 1.01	83 0.66 1.51 0.32		18 1.37 2.88 1.33
CSU-CFDC	**N** **Mean** **St. dev.** **CI**	20 1.91 3.54 1.55	29 3.79 8.98 3.26	28 1.02 1.91 0.71	18 0.73 1.61 0.74	

1433 **Table 3.** Percent agreement within one order of magnitude for all times and temperatures

	N_{INP} (FRIDGE-CS)	N_{INP} (CSU-IS)	N_{INP} (NCSU CS(I))	N_{INP} (NCSU CS(F))	N_{INP} (CSU-CFDC)
N_{INP} (FRIDGE-CS)	100.0				
N_{INP} (CSU-IS)	97.3	100.0			
N_{INP} (NCSU CS(I))	85.9	68.6	100.0		
N_{INP} (NCSU CS(F))	75.0	59.2	96.2	100.0	
N_{INP} (CSU-CFDC)	100.0	87.5	100.0	84.6	100.0

1434

1435   A possible explanation for N_{INP} measurement discrepancies that has been tendered in

1436 other intercomparison campaigns sampling ambient air is that INPs are highly sensitive to the

1437 size range of collected aerosol, and systematic size-dependent differences in collection

1438 efficiencies vary for different collection types (DeMott et al., 2017; Knopf et al., 2021; Lacher et

1439 al., 2024). For example, Lacher et al. (2024) found significant underestimates of INPs by both

1440 online and offline methods measuring from the PM10 inlet versus offline measurements from

1441 filter collections made on the laboratory rooftop. In this study, as we have noted above, a

1442 similarly consistent difference between rooftop versus laboratory or between online and offline

1443 measurements is not found. FRIDGE-CS INP concentration measurements from the turbulent-

1444 flow inlet and CSU-IS INP concentration measurements from the rooftop filter agreed within an

1445 average of about 30% over the course of the study. The CSU-CFDC INP measurements that

1446 were limited and thus biased by its upstream total particle impactor (at 2.5 µm) agreed well on

1447 average with the NCSU (F) and (I) measurements, although we may note that if the CSU-CFDC

1448 data had been corrected for instrumental loss of particles "out-of-lamina" as found for

measurements on mineral dust (DeMott et al., 2015), INP concentration results would have been within a factor of two of the CSU-IS and FRIDGE-CS data. Larger particles do tend to have higher likelihood of containing ice nucleation sites, so biases in their collection can lead to sometimes large differences in assessed INP concentrations (Mason et al., 2016). Disaggregation of the very largest collected particles when placed in water suspensions has also been implicated for discrepancies between different substrate collections (DeMott et al., 2017; Lacher et al., 2024). For example, if very large aggregates that are preferentially collected by one substrate versus another, disaggregation in water could lead to a high bias in ice nucleation sites effective at lower temperatures. There may have been additional line losses for the online instruments sampling from an inlet and using tubing to transfer particles, though these tend to be of minor influence at below the impactor size cut (Knopf et al., 2021). The impinger is known to be less efficient for small (<200 nm) and large (>10 μm) particle capture, but unless the relatively light to moderate wind conditions at the inlet during FIN-03 conferred some special bias, Hader et al. (2014) predict a 50% capture efficiency at near 10 μm. The filter samplers on the rooftop should have been equivalent, with the only difference being the orientation of filters for the NCSU samples (mounted face-down). The size bias in this configuration is unknown. The FRIDGE filter should have captured particles with the same efficiency as the turbulent flow inlet, since only a very short line connected the filter to the interior inlet structure in the laboratory. Only if very large INPs > 13 μm were dominant by number amongst total INPs, which is unexpected, would the FRIDGE filter collection have been expected to differ from the rooftop CSU-IS filter collections.

Besides size-dependent sampling biases, the fact that measurements of immersion freezing INP concentrations from ambient air are generally uncertain by up to one order of

1484 magnitude may result from unquantifiable random or non-random factors, or more likely from

1485 quantifiable factors that were not fully controlled in this field study nor easily controlled across

1486 investigating teams in general. Examples of known issues that were only documented after FIN-

1487 03 relate to inconsistency in sample materials or sample handling and storage (e.g., Barry et al.,

1488 2021b; Beall et al., 2021).

1489 **3.4 Relation of immersion freezing INPs to aerosol properties**

1490   While establishing correlations between INPs and aerosol properties were not a focus of

1491 the intercomparison, the ancillary aerosol data did allow for inspecting some simple linear

1492 correlation analysis. This provides insight into the size range of greatest relevance for the INP

1493 intercomparison period. Throughout the campaign, a positive and significant trend between total

1494 LAS particle concentration (i.e., > 0.1 μm) and N_{INP} was observed for FRIDGE-CS (R = 0.55-

1495 0.74 and $p < 0.05$ for measurements at $-28\ °C < T < -15\ °C$), but no clear statistically significant

1496 trend was observed between total LAS particle concentration and N_{INP} for the other four

1497 instruments (Figure S4a). A greater number of significant positive trends were found between the

1498 concentration of particles with diameter > 0.5 μm and N_{INP}. This was the case for the FRIDGE-

1499 CS (R = 0.54-0.94 and $p < 0.05$ for measurements at $-28\ °C < T < -19\ °C$), CSU IS (R = 0.46-

1500 0.72 and $p < 0.05$ for measurements at -21 to $-25\ °C$), NCSU CS(I) (R = 0.46-0.61 and $p < 0.05$

1501 for measurements at $-29°C < T < -24\ °C$), and the NCSU CS(F) (R = 0.51-0.64 and $p < 0.05$ for

1502 measurements at $-26\ °C < T < -22\ °C$).

1503   No consistent, significant ($p < 0.05$) correlation was found between changes in

1504 composition (from the PALMS categories and WIBS-4A types) and immersion freezing N_{INP}

1505 across the range of setpoint temperatures employed during FIN-03 (Figure S4b).

1506 **3.5 Inferences to INP compositions during FIN-03**

1518   To provide context for the discussed intercomparisons and because this study provides

1519 data needed for testing the relevance of existing parameterizations of ice nucleation in regional

1520 and global climate models (Andreae & Rosenfeld, 2008; Morris et al., 2011; Seifert et al., 2011),

1521 we utilize some previously-developed ice nucleation parameterizations for specific compositions

1522 to diagnose consistency or not with INP compositions in the high altitude environment of FIN-

1523 03. We examine parameterizations for mineral dust INPs that have different links to larger size

1524 particle concentrations (DeMott et al., 2015) versus mineral dust surface area (Niemand et al.,

1525 2012), and biological INPs as linked to fluorescent particle concentrations (Tobo et al., 2013;

1526 Twohy et al., 2016). Hereafter we will refer to these parameterizations as DeMott 2015,

1527 Niemand 2012, and Tobo 2013. We also utilize a more direct method of probing INP

1528 compositions using the CSU-IS sample treatments discussed in Section 2.2.2 and the CSU-

1529 CFDC heat treatments of single particles discussed in Section 2.2.1. In relation to these latter

1530 investigations, we also introduce diagnostic tests of the arable soil dust INP parameterizations of

1531 Tobo et al. (2014).

1532   Each of the above-noted deterministic parameterizations was used to predict N_{INP} at –30

1533 °C, –25 °C, –20 °C, and –15 °C using the equations and inputs described in Table 4 and

1534 summarized below. We do not attempt an analysis using stochastic parameterizations.

1535   1) DeMott 2015 is based on CSU-CFDC laboratory measurements of ice nucleation on

1536   mineral dust soil samples as well as field data from situations dominated by mineral dusts

1537   (i.e., dust plumes from major deserts), collected for CFDC operational conditions

1538   essentially the same as for this study (i.e., simulated immersion freezing conditions at

1539   105% RH) (DeMott et al., 2015). For FIN-03, aerosol concentrations measured by the

1540   LAS (> 0.5 μm dry diameter) and converted to STP concentrations were used as the input

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1543   for this parameterization for comparison to INP data that is also reported at STP

1544   concentrations. Predictions also depend on temperature (Table 4). Since PALMS data

1545   indicates that dust particles dominated the coarse mode only at sizes above 1 µm in

1546   diameter (Figure 4), we first adjust LAS data accordingly for the percentage of dust

1547   particles with diameters > 0.5 µm as input to this parameterization, which we have

1548   already stated is 23%. A correction factor (CF) of 3 was also applied (as indicated in

1549   Table 4) according to the results in DeMott et al. (2015) which showed that when

1550   applying the parameterization to represent immersion freezing dust INP concentrations in

1551   a model or in comparison to other immersion freezing methods, this CF is needed to

1552   account for CFDC underestimates of immersion freezing INPs (see Methods). The CF is

1553   applied in this case because calculations will be compared to the average N_{INP} from all

1554   measurements.

1555   2) The Niemand 2012 parameterization (Table 4) for mineral dust INPs is based entirely

1556   from laboratory measurements and incorporates measurements of temperature and

1557   particle surface area as the basis for prediction of INPs. It is especially important to limit

1558   the size range of aerosols for which this parameterization is applied, because total surface

1559   area was dominated by small particles in FIN-03. Therefore, with reference to Figure 4,

1560   we will assume that all dust surface area occurs at sizes larger than 0.5 µm and represents

1561   50% of that surface area.

1563   **Table 4.** Summary of INP parameterizations.

Param.	Equation	Constants
Mineral dust INPs: Niemand et al. (2012)	$N_{INP}(T_C) \approx n_s(T_C)S_{tot} = (a\ exp\ (b(T_C)+c))(S_{tot})$ N_{INP} (T_C) = INP concentration (sL^{-1}) at T (Celcius) S_{tot} in units µm^{2}cm^{-3} and n_s in units m^{-2}	a = 1x10^{-9} b = -0.517 c = 8.934

Mineral dust: DeMott et al. (2015)	$$N_{INP}(T_K) = (cf)(n_{a>0.5\mu m})^{(\alpha(273.16-T_K)+\beta)} exp\,(\gamma(273.16-T_K)+\delta)$$ $N_{INP}(T_K)$ = INP concentration (sL⁻¹) at T (Kelvin) $n_{a>0.5\mu m}$ = mineral particle number concentration > 0.5 μm (scm⁻³) cf = 1 (CFDC data comparison) or 3 (other immersion freezing)	$\alpha = -0.074$ $\beta = 3.8$ $\gamma = 0.414$ $\delta = -9.671$
Fluorescing biological aerosol particle INPs: Tobo et al. (2013)	$$N_{INP}(T_k) = (N_{FBAP>0.5\mu m})^{(\alpha'(273.16-T_k)+\beta')} exp\,(\gamma'(273.16-T_k)+\delta')$$ N_{INP} = INP concentration (sL⁻¹) N_{FBAP} = FBAP concentration (scm⁻³)	$\alpha' = -0.108$ $\beta' = 3.8$ $\gamma' = 0$ $\delta' = 4.605$
Fluorescing biological aerosol particle INPs: Cornwell et al. (2023)	$$N_{INP}(T_C) = f(T_C)1000N_{FBAP>0.5\mu m}$$ f(T_C = -20 °C) = 0.318 f(T_C = -15 °C) = 0.016	N/A
Arable soil dust INPs: Tobo et al. (2014)	$$N_{INP}(T_C) \approx n_s(T_C)S_{tot} = (a\,exp\,(b(T_C)+c))(S_{tot})$$ $N_{INP}(T_C)$ = INP concentration (sL⁻¹) at T (Celcius) S_{tot} in units μm²cm⁻³ and n_s in units cm⁻²	Total soil: a = 1x10⁻⁵ b = -0.4736 c = 0.3644 Inorganics: a = 1x10⁻⁵ b = -0.6773 c = 7.8436

1569

1570   3) As discussed earlier, we use two definitions of FBAP at sizes larger than 0.5 µm to and

1571   temperature to predict biological INP concentrations based on Tobo 2013 as defined in

1572   Section 2.1, presuming to bracket low and high estimates of their links to INPs. We also

1573   explore links of higher temperature freezing data (> –20 °C) to FP3 particles, using the

1574   same scalings of the relation between FP3 concentrations and INP concentrations as a

1575   function of temperature that were established by Cornwell et al. (2023) for a coastal

1576   California environment. While we have no reason to expect that these scaling factors

1577   listed in Table 4 are valid for the high altitude, continental environment of FIN-03, they

1578   are starting points to explore this additional link of certain FBAP particles to INPs.

1580   To compare these parameterized values with observations, an overall mean observed

1581   immersion freezing N_{INP} was calculated for each three-hour period based on all the available data

1582   from all the instruments. This was considered as a reasonable approach since it factors in the

1583   inherent variability found between methods. Immersion freezing N_{INP} was predicted for each

1584   parameterization using mean WIBS-4A, and LAS data, both at STP concentrations, collected in

1585   the coincident 3-hour periods of time as the INP data. The observed and predicted immersion

1586   freezing N_{INP} are plotted against each other in Figure 9. Four temperatures of comparison (–15, –

1587   20, –25 and –30 °C) are presented in Figure 9 for DeMott 2015, Niemand 2012, and Tobo 2013,

1588   while two temperatures of comparison (–15, –20 °C) are used for links to FP3-based prediction

1589   of biological INPs. Temperatures are indicated via levels of shading of the data points.

1590   Using the constraint on mineral particles from the combination of PALMS and LAS data for the

1591   campaign average, predictions underestimate the mean N_{INP} at all temperatures (Figure 9a). The

1592   Niemand 2012 surface-area-based INP estimates come modestly closer to observations,

1593   averaging 25% of the total INP concentrations for all times and all temperatures, while the

1594   DeMott 2015 predictions average 4% of INP concentrations, with large variability apparent.

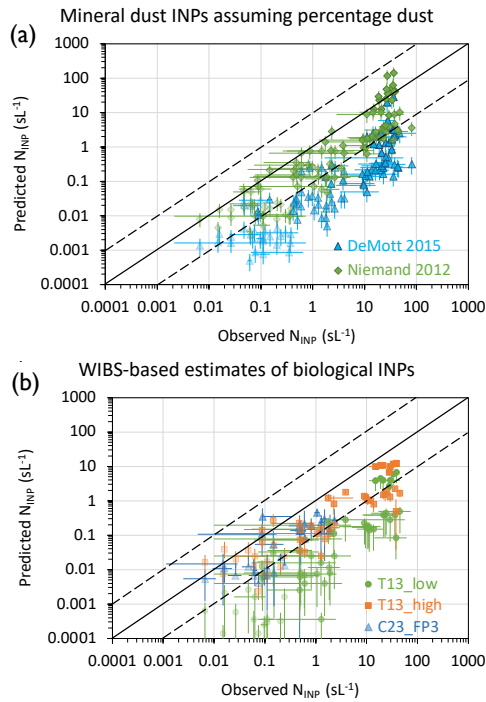
1595   These results can be expected to be highly sensitive to the assessed average

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Figure 9. a) Comparison of mean observed N_{INP} (all instrument average) and predicted N_{INP} calculated

from DeMott et al. (2015) (DeMott 2015) and Niemand et al. (2012) (Niemand 2012) mineral dust INP

parameterizations at temperatures -30 °C, -25 °C, -20 °C, and -15 °C (gradations in shading from dark to

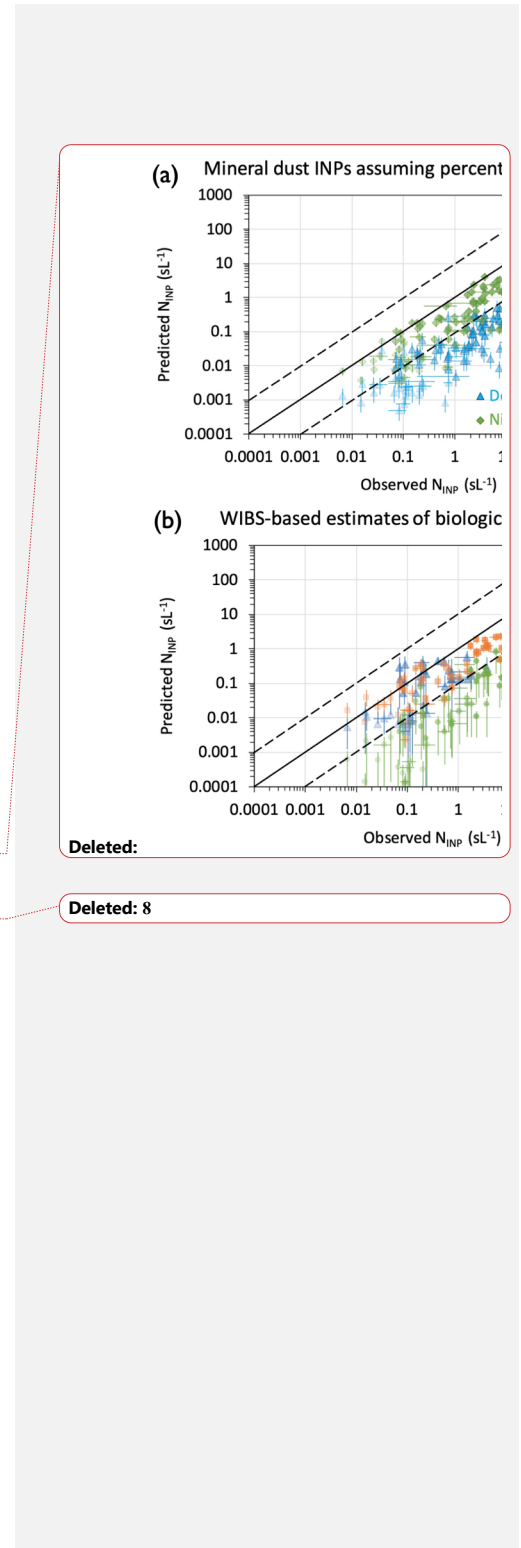
light) for the PALMS estimated percentages of dust particle number and surface area at sizes above 0.5

μm. Mean N_{INP} are averaged over three-hour periods and plotted uncertainties are standard deviations.

Predicted N_{INP} uncertainties are propagated based on 25 % uncertainty in aerosol number and surface area

concentrations. b) Comparison of mean observed N_{INP} and predicted N_{INP} calculated from

parameterizations linking to FBAP concentrations from *Tobo et al.* (2013) (T13_low and T13_high; see

text for description) and from *Cornwell et al.* (2023) (C23_FP3) following the FP3 particle definition of

1613   *Wright et al.* (2014). Only -15 and -20 °C comparisons are shown for the FP3 prediction. The solid line in

1614   each plot is the 1:1 line and the dashed lines represent an order of magnitude in both directions.

1615

1616   mineral particle fraction at sizes above 0.5 μm (varied over the study) and on whether particles

1617   that have a source from regional soils will be represented only by those with mineral content.

1618   Therefore, for comparison, parameterization results in Figure S5 use the assumption that all

1619   particles at diameters exceeding 0.5 μm were dust particles. In this case, a somewhat unrealistic

1620   maximum assumption on soil dust numbers and surface area that considers all particles and

1621   compositions in this size range as emanating from dust, Niemand 2012 estimates a dust source

1622   for 50% and DeMott 2015 estimates 25% of observed INPs on average. Thus, the predictions of

1623   the two parameterizations become more closely aligned for assumption of more overall mineral

1624   dust particles in the size range larger than 0.5 μm. Discrepancy has been noted previously in

1625   applying these parameterizations to link to the aerosol model in an Earth System model for the

1626   Southern Ocean region (McCluskey et al., 2023). In that case, calculations were based on aerosol

1627   model derived dust distributions and occurred under very low dust loading scenarios where

1628   neither parameterization has been firmly tested in the laboratory or field. Under both

1629   assumptions on mineral particle number, since DeMott 2015 was developed based on CFDC

1630   measurements for particles < 2.5 μm in the field and laboratory, a low bias compared to

1631   Niemand 2012 might be expected in comparison to average immersion freezing data that

1632   includes larger particles.

1633   The timeline of predicted N_{INP} for the two dust parameterizations in comparison to mean

1634   observed N_{INP} is shown in Figure 10 for the same temperatures used in Figure 9. These analyses

1635   emphasize that 1) INP observations do not show a special enhancement during the biomass

1636   burning event at the start of FIN-03, and hence closer agreement of the dust parameterizations

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1644   with observations at that time is likely an artifact of attributing dust-like INP activation

1645   properties to the dominant biomass burning compositions at that time; 2) the structure of the

1646   timeline of predicted N_{INP} resembles that of the observed N_{INP} only below –20 °C, as expected

1647   for a dominance of dust-like INPs; and 3) the predictions fare less well in describing the

1648   observed INP populations at > –20 °C where biological INPs may be expected to have greater

1649   influence. Thus, these analyses overall suggest the presence of a dust-like immersion freezing

1650   INP type active at lower temperatures during FIN-03, but that the typical INP efficiency (INP as

1651   a function of dust concentration and temperature) attributed to mineral dust underestimates the

1652   freezing behavior of INPs overall during the period of study.

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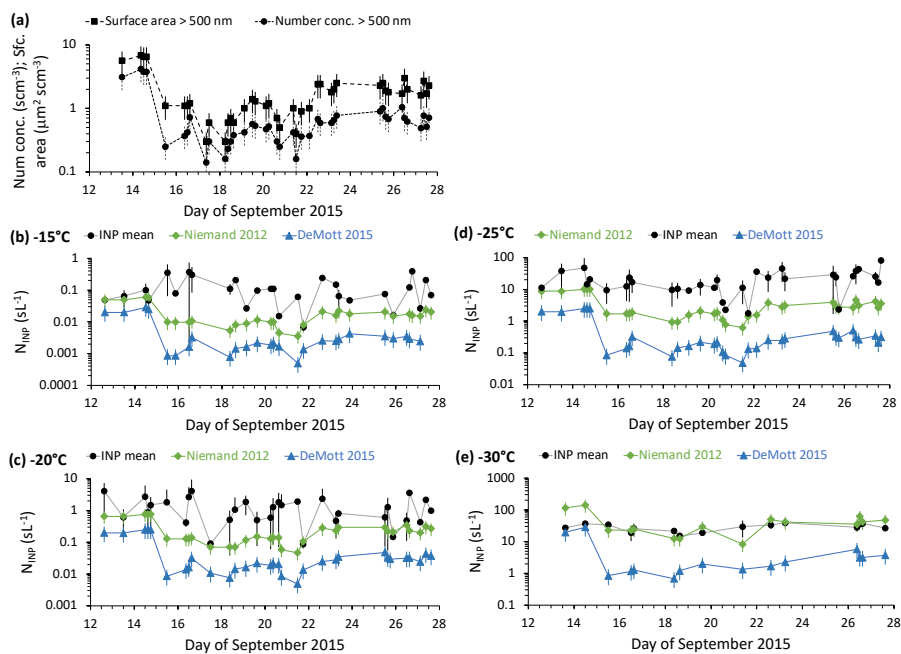
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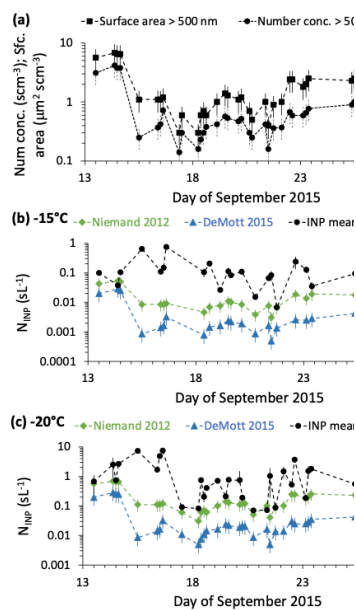

1658   **Figure 10.** Time series of aerosol number concentration and surface area (3-h averages at STP) in a), and

1659   observed mean measured immersion freezing N_{INP} (INP mean) plotted with predicted N_{INP} from the

1660   mineral dust parameterizations of Niemand 2012 and DeMott 2015 as described in the main text (all

1661   three-hour averages at STP) at temperatures of -15, -20, -25, and -30 °C in b) to e), respectively. Lines are

1662   intended only to connect data points and do not imply knowledge of intermediate values. Uncertainties

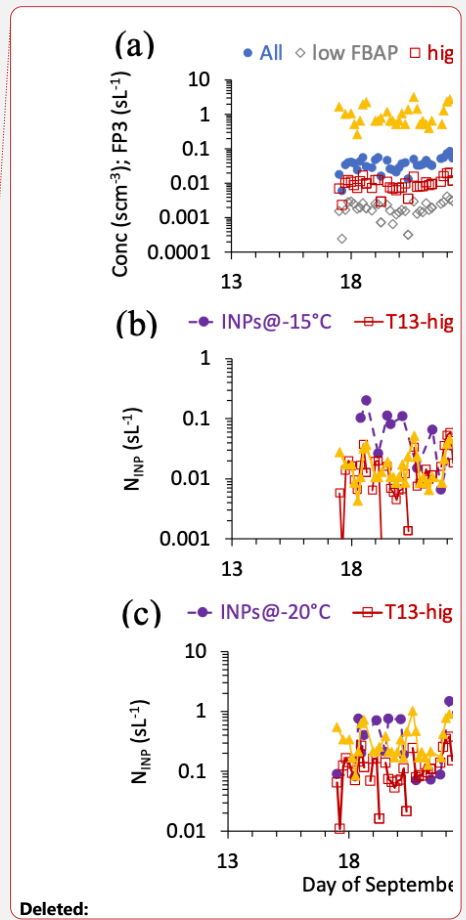
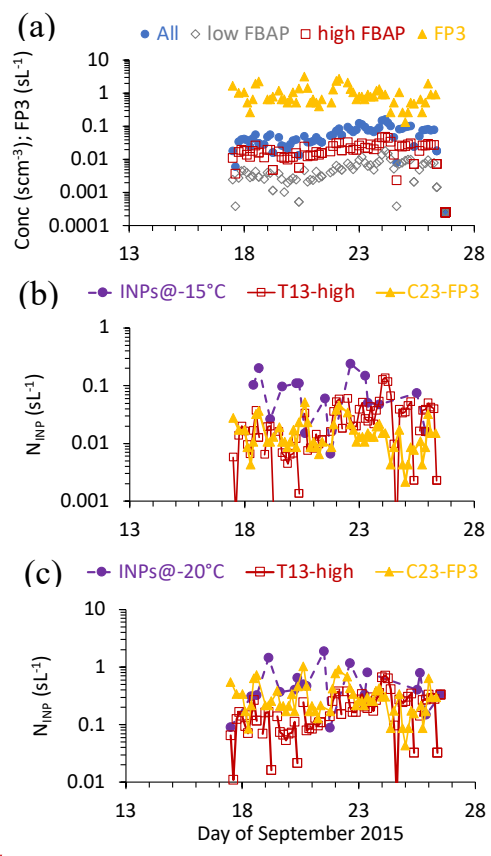
1663   mark one standard deviation above and below the mean values of all parameters.

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Figure 11. a) Timelines of WIBS-based fluorescent particles assignments (all fluorescing in any channel, low and high FBAP, and FP3 particles), as defined in the text, during FIN-03. b) INP observed mean concentrations and biological INP parameterization predictions linked to high FBAP following Tobo et al. (2013) (T13-high) and FP3 particles following Cornwell et al. (2023) at -15 °C in b) and -20 °C in c).

For FIN-03, the Tobo 2013 parameterization of biological INPs consistently underpredicted N_{INP}, independent of the WIBS FBAP definition used, denoted as T13_low and T13 high in the scatterplot comparison of measured versus predicted values (at all times and temperatures) in Figure 9b and the timeline comparisons at –15 and –20 °C shown in Figure 11. Figure 11 also shows the timeline of WIBS total fluorescent particle concentrations, the high and low FBAP concentrations, and FP3 concentrations. The higher FBAP prediction of INPs falls much closer to the observations than the low FBAP prediction in Figure 9b and shares some proximal equivalence to observations at –15 to –20 °C at times. This result is like that found by Twohy et al. (2016) for air over the site where Tobo et al. (2013) collected their data, with the higher FBAP estimate bounding the upper end of measured immersion freezing INP concentrations at temperatures > –20 °C. Also notable in Figure 9b and Figure 11 is that the C13-FP3 INP concentration predictions filled a similar space as the T13_high estimates, coming closest together at –20 °C. While these results suggest that biological INP parameterizations can explain the higher temperature INP concentrations observed during FIN-03, with caveats on the large and likely not fully quantifiable uncertainty in such predictions, the temporal analysis indicates that there is no consistent temporal agreement between predicted and measured INPs, even if different scaling factors were applied to the predictions. Predictions at –20 °C show better

1703 overall agreement, while those at –15 °C suggest that the Cornwell et al. (2023) scaling factor

1704 should be higher for the SPL site at the time of FIN-03 to better describe mean values of

1705 biological INP concentrations using the FP3 particle signal.

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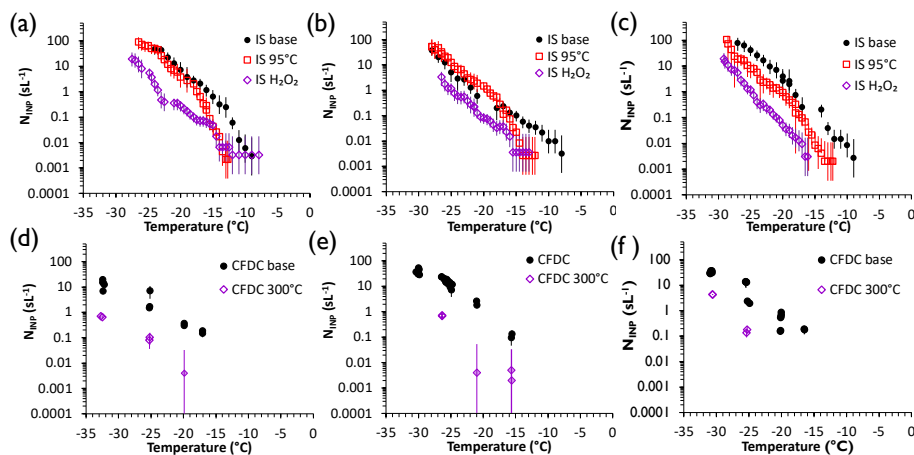
Figure 12. Summary of treated IS filter suspensions using heat and peroxide (a, b, c) and dry heat-treated CSU CFDC single particle data (d, e, f), for September 15, 23 and 25 (a-c, d-f, respectively). Error bars represent 95% confidence intervals for individual experimental spectra for the CSU-IS and for individual CSU CFDC measurements.

1712 The results of CSU-IS and CSU-CFDC treatments on INP concentrations measured for

1713 three (of 21 overall) intercomparison time periods are shown in Figure 12, for examination of

1714 consistency with the results of the diagnostic parameterization analysis just discussed. In Figure

1715 12a-c, it is seen that thermal treatments indicated the strong contribution of inferred biological

1716 INPs primarily at temperatures higher than about –20 °C, but that peroxide digestion of organic

1717 compounds lowered INP activity at all tested temperatures by an order of magnitude on average.

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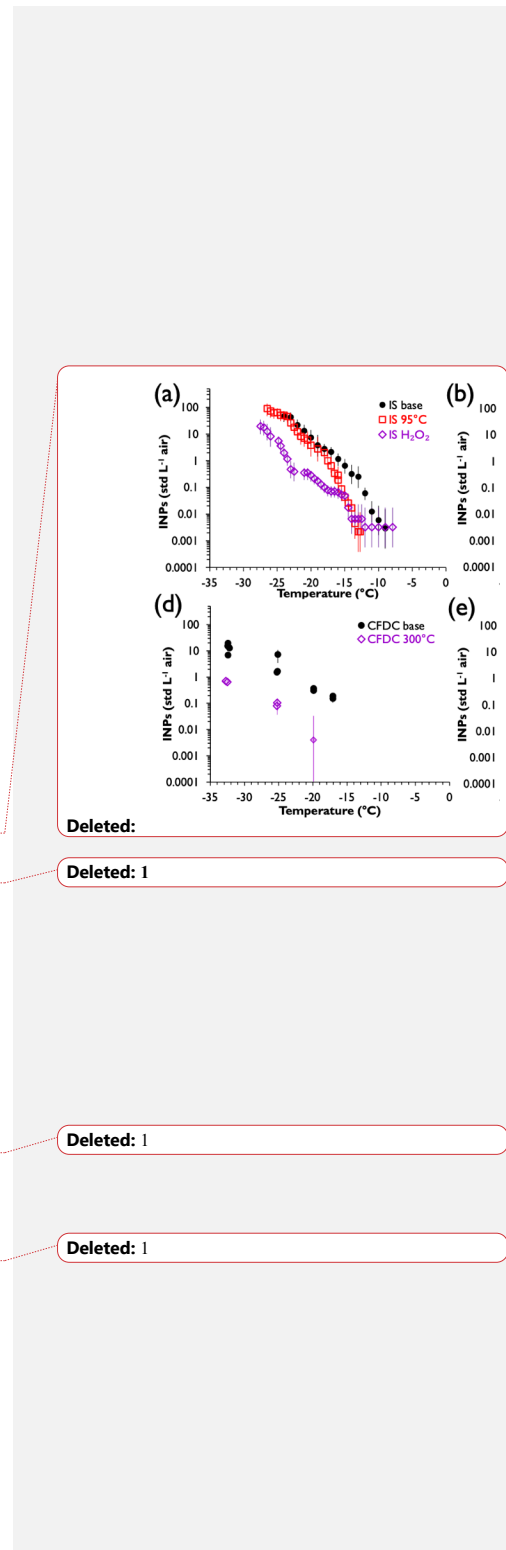
1722   Similar reductions of INPs measured for single particles by the CSU-CFDC following dry

1723   heating (Figure 12d-e) demonstrate strong consistency with the IS results for bulk immersion

1724   freezing on the dominance of organic INP compositions, even though CSU-CFDC measured

1725   unamended INP concentrations were always lower. The CSU-IS heat treatment results (Figure

1726   12a-c) suggest that biological INPs may have been ubiquitous during FIN-03 at temperatures

1727   above –20 °C, and extended to lower temperatures at times, as indicated by the results from

1728   September 25. This is broadly consistent with the parameterization results based on FBAP

1729   measurements, although the Tobo 2013 and FP3 parameterizations did not capture all the

1730   influence of apparent biological INPs during the study. Whether for size-limited (< 2.5 µm) as in

1731   CSU-CFDC measurements, or bulk aerosol collected for CSU-IS immersion freezing

1732   measurements, the inferred INP compositions that were typically dominated by organics at

1733   temperatures < –20 °C could reflect origins from arable soil dusts (Testa et al., 2021) that

1734   surround the region of study. Biomass burning aerosols also have influence as organic INPs

1735   (Schill et al, 2020; Barry et al., 2021a). However, while biomass burning type particles were

1736   noted as a prevalent composition in FIN-03, these types of potential INPs likely cannot explain

1737   INP concentrations in FIN-03 because Barry et al. (2021a) showed that Western U.S. biomass

1738   burning INPs have active site densities about 3 orders of magnitude lower than those attributed

1739   to dust particles that also were ubiquitous at modest number concentrations during FIN-03.

1740   Furthermore, the strong biomass burning event noted on September 14 had only modest, if any,

1741   apparent impacts on INP concentrations despite greatly elevated aerosol concentrations and

1742   surface areas, as already mentioned above (Figure 10).

Finally, in Figure 13 we address whether the treatment results support the conclusion of the diagnostic parameterization analysis suggesting that inorganic INPs (mineral particles in particular) were of minor influence during FIN-03.
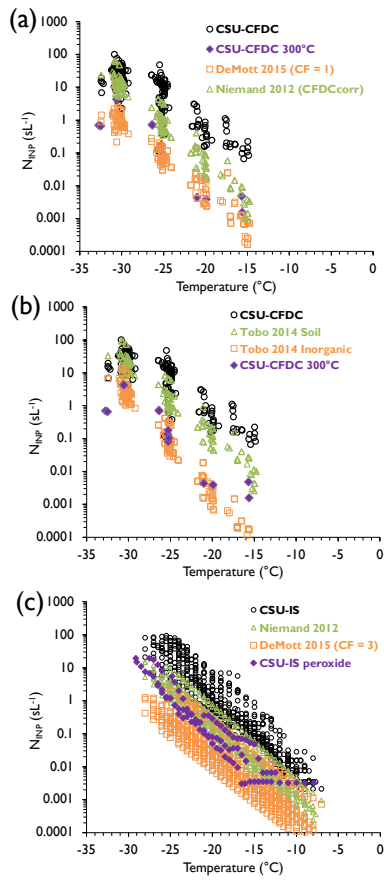
Figure 13. a) Comparison of all untreated CSU CFDC data (black circles), cases after passing through the upstream 300 °C tube heater (purple diamonds), and calculations from the DeMott 2015 dust parameterization in (orange squares) and with CF = 1 as appropriate for a direct comparison to CSU

1762 CFDC data (see text). b) The same exercise as in a) but using predictions of total soil organic INP

1763 concentrations and inorganic INP concentrations within soil INPs, both from Tobo et al. (2014). c) The

1764 same exercise but for all CSU-IS data and the cases with peroxide digestion. In this case, CF = 3 must be

1765 used in DeMott 2015 and the mineral dust INP prediction of Niemand 2012 is also shown.

1766

1767 For this purpose, we introduce results for the parameterization of Tobo et al. (2014) (hereafter,

1768 Tobo 2014) for arable soil dust INPs listed in Table 4. Tobo et al. (2014) parameterized the ice

1769 nucleation behavior of soil dusts from Wyoming, regionally proximal to the FIN-03 site at SPL,

1770 specifically using the and the CSU-CFDC dry heat method at 300 °C to indicate organic versus

1771 inorganic INP contributions from such soil particles. A caveat is that their results were for dusts

1772 generated in the laboratory and size-selected at 600 nm. This parameterization, like Niemand

1773 2012, is based on the surface area of dust particles and so we apply the same assumptions as

1774 before to restrict to the proportion of dust larger than 0.5 µm. Since the CSU-CFDC is also

1775 restricted to measuring INPs at diameters below 2.5 µm, we apply a correction factor to the

1776 surface area to account for the fact that the surface area at below this size was 90% of the project

1777 average total surface area. No significant impact of the treatments is assumed on aerosol

1778 concentrations or surface area at sizes above 0.5 µm in Figure 13.

1779   Figures 13a and 13b focus on specific comparisons to CSU-CFDC data. In Figure 13a, it

1780 is seen that INP concentrations predicted by the DeMott 2015 parameterization for sampling

1781 periods during the entire campaign show remarkable agreement with the 300 °C CSU-CFDC

1782 data on selected days when applying CF = 1 in the parameterization, as is appropriate for a direct

1783 comparison to CSU-CFDC instrument data that is uncorrected for the underestimates that led to

1784 selecting CF = 3 for atmospheric modeling studies. In Figure 13 b, it is shown that the Tobo

1785 2014 parameterizations for untreated soil dusts and the inorganic remnants also give very good

agreement with CFDC untreated and treated N_{INP} data, supporting the likely important influence

of such arable soil dusts during FIN-03. Predictions for untreated soils do not quite reach the

level of the observed INPs, but this could be explained by the additional contribution of

biological INPs that has already been discussed.

In Figure 13c, direct comparisons of the Niemand 2012 and DeMott 2015 predictions for

mineral dust INPs for the entire project are shown in comparison to the CSU-IS untreated and

H_2O_2 treated data on selected days. The DeMott 2015 prediction of INP concentrations uses CF

= 3 in this case, as appropriate. The same discrepancy between the DeMott 2015 and Niemand

2012 predictions as discussed already regarding Figure 9a appears in this comparison.

Nevertheless, it is seen that both parameterizations grossly underestimate untreated CSU-IS INP

concentrations and the treated CSU-IS results fall between the predicted values, agreeing better

with the Niemand 2012 parameterization. While one might wish to allude to the fact that the IS

filters sample particle sizes, to 10 μm and possible larger that may have higher ice nucleation

efficiencies, while the CSU-CFDC was restricted to sampling particles <2.5 μm as a source for

the lower DeMott 2015 estimate in comparison to CSU-IS data, we have already addressed that

there was no general consistency in INP concentrations for methods that sampled similar size

particles overall. The best that can be stated is that the parameterization exercises and treatment

data strongly support that inorganic INPs were of weak influence during FIN-03 and that arable

soil dusts and biological INPs accounted for the strongest influences during sampling, akin to the

findings of Testa et al. (2021).

3.6 Observations of INPs in the deposition nucleation regime

Measurements of deposition nucleation N_{INP} are summarized in Figures 14 and 15.

FRIDGE-DC nucleation substrates were collected for 1 to 5 periods on many days during FIN-03

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1822 and processed at 5-degree interval temperatures from –15 to –30 °C, and for setpoint humidity of

1823 95% and 99% RH (uncertainties to 2%). Data collected at 102% via the standard FRIDGE

1824 methods are not included herein. CSU-CFDC and MIT-SPIN deposition data were collected

1825 nominally at 95% RH with an uncertainty of about 2.5% RH, and at a range of temperatures on

1826 different days. Mean values and standard deviation error bars of the FRIDGE-DC data are

1827 shown in Figure 14a and median values of FRIDGE-DC N_{INP} (with interquartile values as error

1828 bars) are shown in Figure 14b. Standard deviations were large over the course of the study for

1829 comprehensive FRIDGE-DC data when binned at 5-degree interval temperatures. Nevertheless,

1830 average concentrations of deposition INPs measured by the FRIDGE-DC

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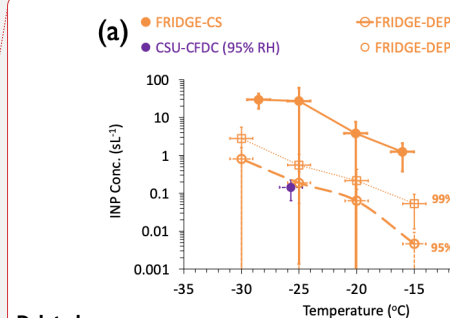
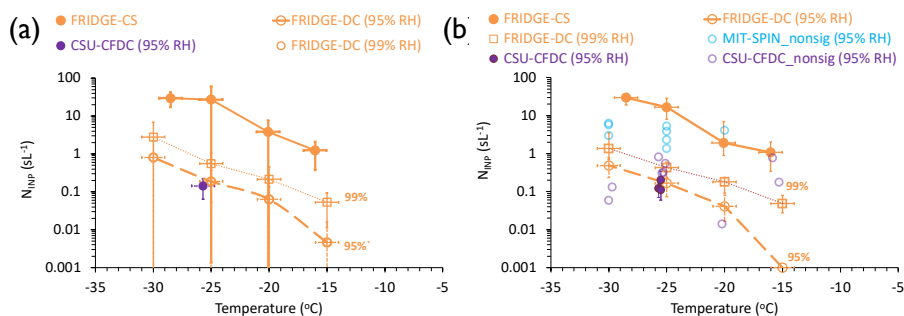

1832 **Figure 14.** Summary of deposition-mode N_{INP} (sL^{-1}) as a function of temperature. In a), mean FRIDGE-

1833 DC data at 95% (open orange circles) and 99% (open orange squares) RH are shown along with mean

1834 immersion freezing data from the FRIDGE-CS (filled orange circles) and the mean for the few cases of

1835 statistically significant CSU-CFDC data (filled purple circle) at 95% RH. Error bars are one standard

1836 deviation of the means. In b), median FRIDGE-DC data are shown and error bars for these are the 95%

1837 confidence intervals. The significant CSU-CFDC measurement points at 95% RH are also shown with

1838 their 95% confidence intervals. Data measured at 95% RH from the CSU-CFDC and MIT-SPIN that were

1849   positively valued, but failed significance testing are shown without errors as open purple and open blue

1850   circles, respectively.

1851

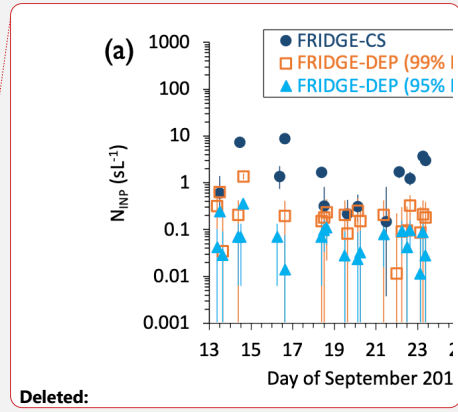
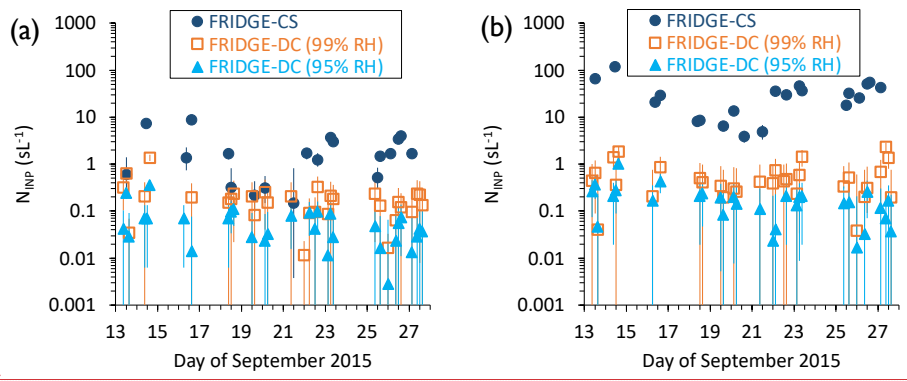
1852   indicated a consistent 3-5 factor increase between 95 and 99% RH over the range of

1853   temperatures investigated. N_{INP} differences at the two RH values were slightly smaller for

1854   median values (Figure 14b), and the median values are slightly lower than the means. Finally,

1855   FRIDGE-CS values are plotted in each panel of Figure 14, indicating that FRIDGE-DC N_{INP}

1856   concentrations averaged for 99% RH are factors 10 to 30 lower than average immersion freezing

1857 N_{INP} concentrations, depending on temperature.

1858

1859   **Figure 15.** Time series of FRIDGE-CS (immersion freezing) and FRIDGE-DC (deposition) N_{INP}

1860   measured at a) -20 °C, and b) -25 °C. Data are from individual filters or wafer collections and error bars

1861   are 95% confidence intervals.

1862   One day of significant data was obtained for the CSU-CFDC deposition measurements

1863   while using the aerosol concentrator, on September 14, containing three different time periods.

1864   These are averaged to create the only online data point represented as a mean in Figure 14a. The

1865   individual period measurements from this day, with confidence intervals as errors, are shown for

1874   the CSU-CFDC in Figure 14b. Thereby it is seen that these measurements at close to –25 °C

1875   agree very well with the mean FRIDGE deposition N_{INP} at –25 °C and 95% RH. No

1876   measurements of significance were achieved with the MIT-SPIN when operating in the

1877   deposition regime. In fact, the most common CSU-CFDC and MIT-SPIN deposition nucleation

1878 N_{INP} results were below instrument detection limits, not meeting the test for significance despite

1879   being positively valued, as shown for all periods from 6 common days of such observations

1880   represented in Figure 14b. Understanding that these data represent a failure to collect

1881   statistically-defensible data, the non-significant data generally scatter about the significant CSU-

1882   CFDC data and the FRIDGE-DC data at 95% RH, with a higher bias for the MIT-SPIN data.

1883   This indicates the difficulty for online continuous flow instruments to capture low deposition

1884 N_{INP} concentration data that fall below 1 sL^{-1} at most times, considering the FRIDGE-DC data as

1885   the standard. Higher sample volumes and limited background frost conditions are needed to

1886   sense these low atmospheric INP concentrations.

1887   Time series of the FRIDGE-DC measurements at –20 °C and –25 °C are shown in Figure

1888   15. Deposition-mode N_{INP} has been averaged over three-hour periods for this analysis. The

1889   FRIDGE immersion freezing data is included in this figure to allow for direct comparison

1890   temporally. Immersion freezing N_{INP} generally exceeded deposition-mode N_{INP} when both types

1891   of measurements were collected by the two FRIDGE operational methods within the same period

1892   (or during adjacent time periods). This difference ranged from 0 to 2 orders of magnitude, with

1893   the largest differences seen at –25 °C and a period of insignificant differences between the

1894   operational mode results seen only from the 18th to the 22nd of September at –20 °C (Figure 15a).

1895   Based on these FRIDGE-CS and FRIDGE-DC results, immersion-mode ice nucleation

1896   dominates at most times at mixed-phase cloud temperatures. Nevertheless, deposition-mode ice

1905 nucleation contributes modestly to the pool of INP at mixed-phase cloud temperatures in the

1906 atmosphere, and thus may bear consideration for parameterization in atmospheric models. The

1907 ability of online ice nucleation instruments to measure N_{INP} in the deposition mode in

1908 correspondence to offline measurements has not been confirmed due to the mentioned inability

1909 of the online instruments used in FIN-03 to capture the low deposition nucleation N_{INP}

1910 concentrations. More work should be carried out on measurements of INPs in the deposition

1911 mode to understand variabilities in time and their relation to INP size and composition, as well as

1912 to resolve if online measurements can be improved. For the time being, the substrate methods

1913 appear to be recommended for ambient atmospheric measurements in the realm below water

1914 saturation at mixed-phase cloud temperatures.

1915

1916 **4. Summary and conclusions**

1917   FIN-03 was an ice nucleation instrument intercomparison conducted in the challenging

1918 environment of the high-altitude mountaintop field setting. Two online systems (CSU-CFDC,

1919 MIT-SPIN) and three offline systems (FRIDGE, CSU-IS, NCSU-CS) were represented in FIN-

1920 03. The immersion freezing INP concentrations measured in FIN-03 by one or more instruments

1921 spanned a dynamic range of over five orders of magnitude (10^{-3} to $\approx 10^2$ L^{-1}) over the temperature

1922 range -34 °C to -7 °C. Intercomparisons for two or more measurements were made from -30 to

1923 -15°C . Agreement within one order of magnitude in immersion freezing N_{INP} was generally

1924 observed between all ice nucleation instruments measuring immersion INP concentrations at any

1925 given temperature if measurement and sampling times were matched to within 3 hours. Better

1926 than one order of magnitude agreement was found at temperatures lower than -25°C and higher

1927 than -18°C, with occasional deviations larger than an order of magnitude in the temperature

1932 range –25 °C to –18 °C. Always better than an approximate 5x factor agreement was found

1933 between average ratios of the N_{INP} measured by pairs of instruments for all times of sampling

1934 We do not have a full understanding of what controls better or worse agreement at different

1935 times or different temperatures, though some factors have been previously discussed in

1936 documenting FIN-02 laboratory studies (DeMott et al., 2018). In this study, there was some

1937 inference that the different filters and impinger used did not equally capture particles in all size

1938 ranges, which is something to improve on in future studies. A review of handling and storage

1939 protocols for consistency amongst groups could also help isolate the role of such factors. Given

1940 the constant changes in the concentration, size distribution and composition of the ambient

1941 aerosol population, inevitable with any field campaign, the level of agreement found represents

1942 state-of-the-art, at least as judged based on recent laboratory and other field comparisons using

1943 similar instrumentation that appear to show 5x factor agreements (e.g., Knopf et al., 2021;

1944 Brasseur et al., 2022; Lacher et al., 2024).

1945   Although FIN-03 was not conducted as an aerosol/INP closure study per se, ancillary

1946 data on aerosol sizes and compositions as recommended in more recent discussions of needs for

1947 true closure exercises (Knopf et al., 2021; Burrows et al., 2022) were purposefully collected for

1948 integration into analyses. This included explicit measurements of the aerosol size distribution,

1949 and single particle measurements of aerosol chemical and biological composition. These

1950 measurements allowed inferences to be made about INP compositions that provided context for

1951 the period of study and establish an example for future intercomparison and long-term

1952 measurement efforts. Through comparing INP data to some current parameterizations describing

1953 biological, mineral and soil dust INPs, and additional direct investigations of INP composition

1954 via certain pre-treatments to remove biological and organic immersion-freezing INPs, these

1956 investigations revealed ubiquitous biological and organic-influenced soil-dust-like INP

1957 influences that mimic those found over other continental regions (Knopf et al., 2021; Testa et al.,

1958 2021; Lacher et al., 2024). Biological INPs were indicated via selected immersion freezing heat

1959 treatments to be dominant at > -20 °C, although of potential influence at all mixed-phase

1960 temperatures. Prediction of these based on parameterizations that utilize single particle

1961 fluorescence data (Tobo et al., 2013; Wright et al., 2014; Cornwell et al., 2023) suggest the

1962 average utility of such parameterizations but these were unable to predict the full temporal

1963 variation of biological INPs. This suggests that local variations of these INPs, which may in fact

1964 represent multiple biological particle types, is an area that requires more effort. Based on

1965 relatively good consistency between predicted and measured mineral influences on immersion-

1966 freezing N_{INP} concentrations, strictly mineral or other inorganic components of INPs were

1967 suggested to have a modest contribution to total INP concentrations at most times and at the

1968 freezing temperatures probed during this study. As in most prior studies, the mineral influence

1969 became stronger at the lowest temperatures assessed. In contrast, it was found by comparison to

1970 a parameterization based on proximally regional soil particles that arable soil INPs likely

1971 explained the second most important contribution (behind biological INPs) of INPs during FIN-

1972 03, those emanating from other organic particle components that may have been internally mixed

1973 with minerals. Biomass burning influences were possible but appear to have not contributed

1974 greatly to the climatology of INPs during the study. It was critically important in arriving at these

1975 conclusions to have single particle aerosol composition data, from a mass spectrometer that

1976 could discern the sizes and fractional contribution of minerals and from a laser-based single

1977 particle fluorescence measurement to estimate the biological character of particles. Nevertheless,

1978 there is a limit beyond the instrumentation complex here utilized in that INPs may always

1979 constitute a subset of the aerosol different in composition and size than the predominant aerosol.

1980 Knowledge advance may require improvement in methods that link INP and compositional

1981 measurements on single particles to specifically isolate these factors. Hence, a great amount of

1982 work is still needed to generally parameterize the mixed INP populations that may occur

1983 temporally in the atmosphere at higher altitude sites like SPL, or anywhere for that matter.

1984   Importantly, FIN-03 included an assessment of the separate relative contributions of

1985 deposition and immersion freezing INP concentrations, one of the few existing data sets of this

1986 kind. The offline FRIDGE-DC method was used to acquire comprehensive deposition N_{INP}

1987 measurements in dependence on RH (95 and 99%), while the CSU-CFDC and MIT-SPIN

1988 instruments attempted focused deposition nucleation measurements at (nominally) 95% RH on

1989 several days. The deposition INP concentration obtained by FRIDGE-DC increased from 95%

1990 RH to 99% RH on average by a factor of 3.3. Also, deposition N_{INP} were nearly always lower

1991 than immersion freezing N_{INP} for the temperatures assessed. Deposition INP concentrations at

1992 most times at 99% RH (always at 95% RH) were lower by an order of magnitude than

1993 immersion freezing INP concentrations at –20 °C and by more than an order of magnitude at –25

1994 °C. For the online instruments, only limited periods of deposition INP measurements with the

1995 CSU-CFDC achieved statistical significance. While these data were in good agreement with

1996 FRIDGE-DC data at –25 °C and 95% RH, the most striking result was that all other

1997 measurement periods for the CSU-CFDC and MIT-SPIN gave measurements that were not

1998 significant at the 95% confidence level. Thus, currently, offline methods for measuring

1999 deposition INPs appear to offer the best chance for success in measuring the lower

2000 concentrations of INPs that activate below water saturation in the mixed-phase temperature

2001 regime. It would be useful to make such assessments at a variety of sites to confirm

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2011 measurements made during FIN-03 on the relative contributions and variability of INPs active in

2012 these conditions toward ice formation in clouds. Additional instrument developments for online

2013 measurements of these, and future intercomparisons, will be useful.

2014   In summary, the agreements amongst instruments during FIN-03, within factors ranging

2015 from nearly 1 to up to 5.5 times on average between individual measurements and rarely

2016 exceeding one order of magnitude in short time periods, match those found in the FIN-02

2017 laboratory studies. These represented state-of-the-art for measurements at the time of FIN-03 and

2018 taken together with further improvements since this time as reflected in recent studies (Knopf et

2019 al., 2021; Brasseur et al., 2022; Lacher et al., 2024) demonstrate steady improvement in the

2020 community's collective ability to detect and quantify atmospheric ice nucleation. There was not a

2021 clear divide between the ability of online and offline systems to measure immersion freezing INP

2022 concentrations from the data collected in this study, although the need to carefully consider

2023 aerosol sampling efficiencies for different instruments was highlighted as a potential issue, one

2024 requiring close attention in future studies. In principle, both types of instruments show excellent

2025 promise for future field studies. For full closure studies of ice nucleation by atmospheric

2026 aerosols, methods for identifying INP composition as demonstrated herein and recommended by

2027 other recent discussions in Knopf et al. (2021) and Burrows et al. (2022) are critical for

2028 understanding and improving INP measurements overall.

2029   There is a clear need in the future to extend measurement comparisons to the

2030 atmospherically-relevant and critically important temperature range higher than –15 °C. The low

2031 atmospheric number concentrations of INPs existing at times at these temperatures is a

2032 significant challenge for such, reflected in this study by the inability to measure INP

2033 concentrations above detection limits at the SPL site even for 3-to-4-hour filter collections at

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2040   temperatures higher than –7 °C. Longer sample times and higher volume collections can improve

2041   this situation, but introduce other technical challenges and do not appear possible for online

2042   instruments.

2043   We also herein do not address the relevance of INP measurements overall for

2044   understanding ice formation in clouds, where secondary processes may come into play. This is

2045   an additional topic for critical investigation, given a degree of confidence now established in

2046   measuring INPs. However, the fact that 5-factor to order of magnitude correspondence between

2047   measurements equate to 3.5 to 5 °C temperature uncertainties in assessment of INPs is something

2048   that also deserves scrutiny from the cloud modeling community concerning if this is satisfactory,

2049   and if not, what level of correspondence should the INP research community be seeking.

2050

2051 **Data availability** All data used for the figures in this paper can be accessed at

2052 https://radar.kit.edu/radar/en/dataset/eGhfvcOhsOyADZXN (persistent

2053 doi:10.35097/eGhfvcOhsOyADZXN)

2054 **Author contributions**

2055 Paul J. DeMott, Jessica A. Mirrielees and Sarah D. Brooks wrote the paper with assistance from

2056 all teams and authors contributing information on instrument descriptions and comments on all

2057 results and conclusions, with contributions from Jake Zenker on some data analysis. Paul J.

2058 DeMott, Ezra J.T. Levin, Thea Schiebel, Kaitlyn Suski, and Tom Hill provided data and analyses

2059 from the CSU-CFDC and IS instruments. Daniel J. Cziczo, Martin J. Wolfe, Sarvesh Garimella,

2060 and Maria Zawadowicz provided MIT-SPIN team measurements and analyses. Markus D.

2061 Petters and Sarah S. Petters provided data and analysis for the NCSU-CS instrument. Heinz G.

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2062 Bingemer, Jann Schrod, and Daniel Weber provided data and analyses for the FRIDGE

2063 instrument. Anne Perring provided data and analyses for the WIBS-4A. Karl Froyd provided

2064 data and analyses for the LAS and PALMS. Anna Gannet Hallar and Ian McCubbin oversaw

2065 field operations, coordinated with visiting teams at Storm Peak Laboratory, and provided

2066 nephelometer and meteorological measurements. Paul J. DeMott, Daniel J. Cziczo, Ottmar

2067 Möhler contributed to organize the campaign in connection with the other FIN activities.

2068

2069 **Competing interests**

2070   The contact author has declared that none of the authors has any competing interests.

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2089

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Page 16: [1] Deleted	Paul DeMott	10/14/24 12:03:00 PM

Page 44: [2] Deleted	Paul DeMott	11/5/24 10:40:00 AM

Page 44: [2] Deleted	Paul DeMott	11/5/24 10:40:00 AM

Page 44: [3] Deleted	Paul DeMott	10/10/24 1:41:00 PM

Page 45: [4] Deleted	Paul DeMott	11/3/24 8:00:00 PM

Page 53: [5] Deleted	Paul DeMott	11/14/24 8:11:00 AM

Field Intercomparison of Ice Nucleation Measurements: The Fifth International Workshop on Ice Nucleation Phase 3 (FIN-03)

Supporting Information

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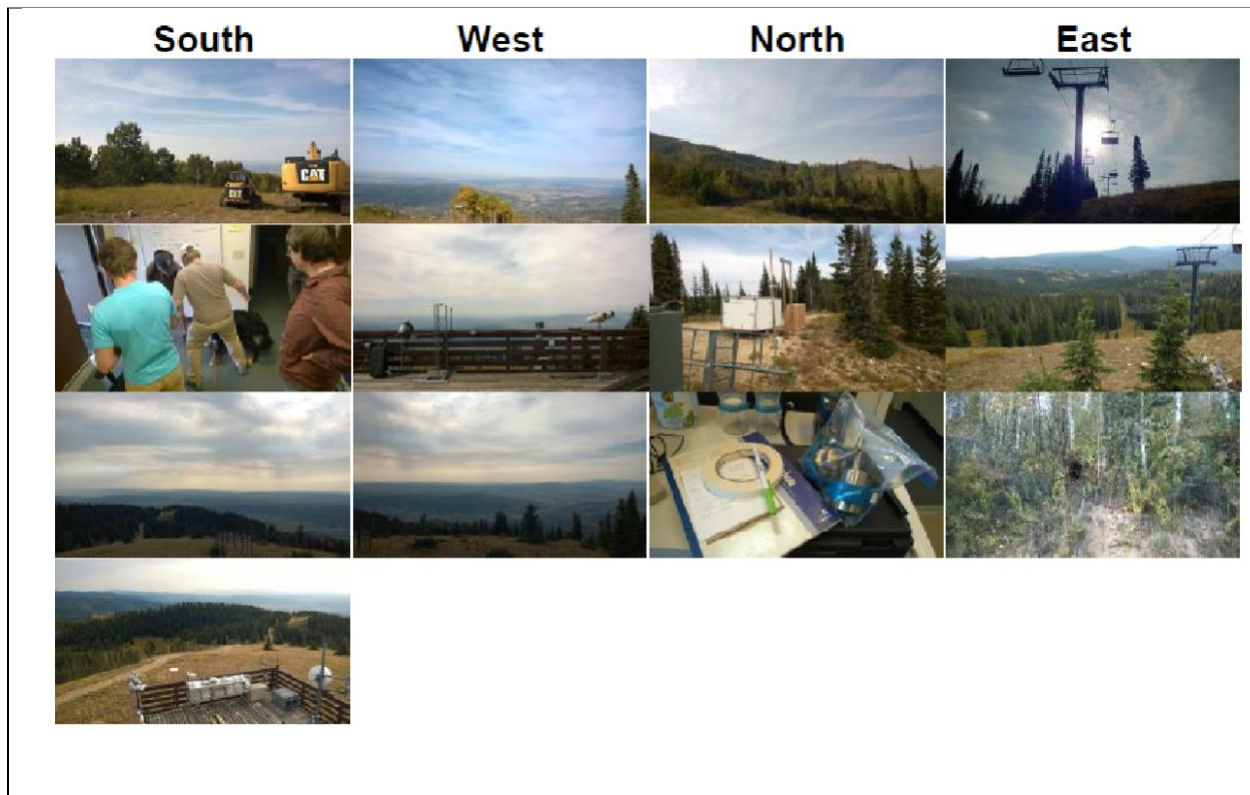
S1. Weather conditions and photos

Photographs shown here are available at https://sspetters.github.io/fin03/index.html or upon request to Sarah Petters.

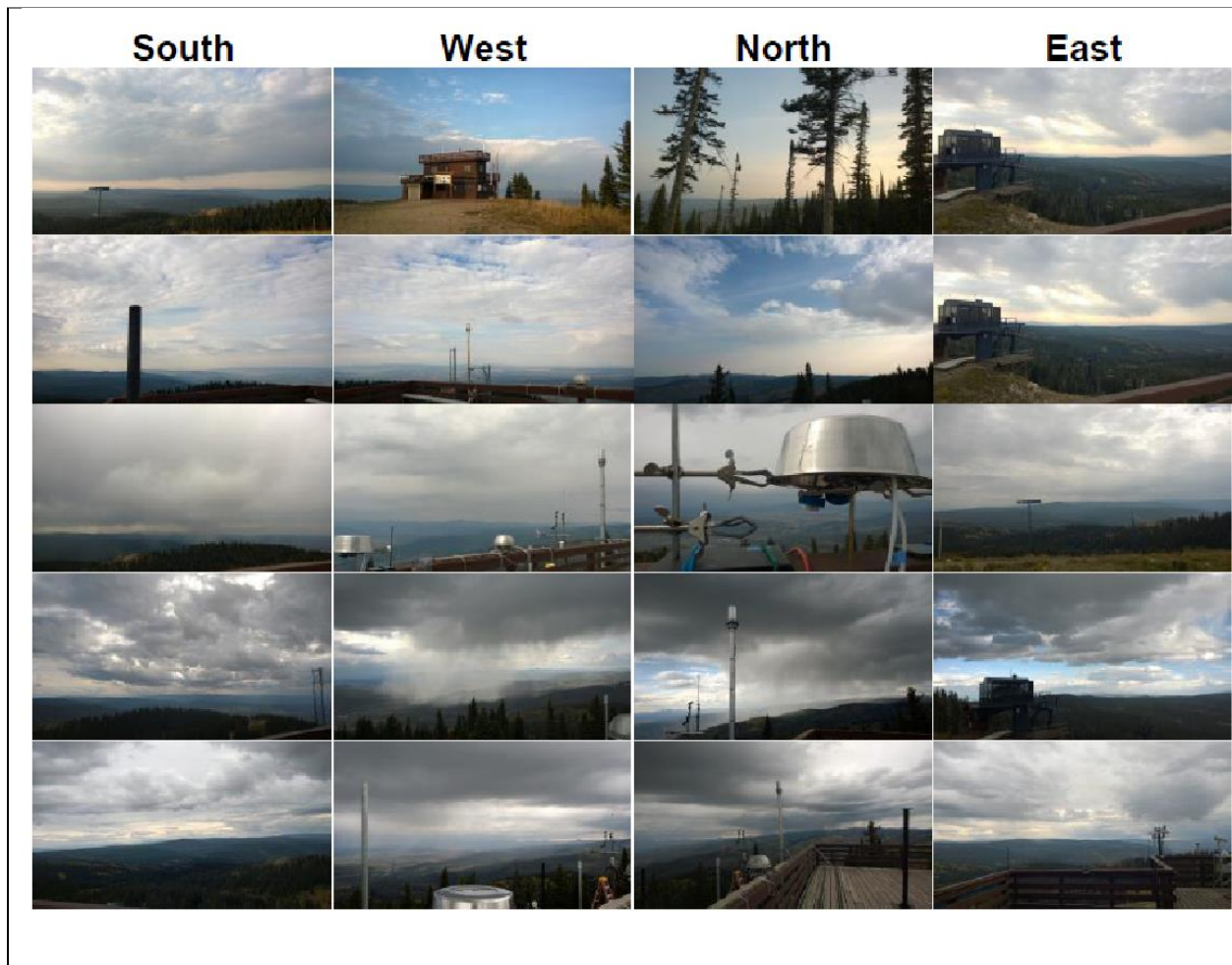
Date: September 12, 2015

Observations

Sky 100% clear, warm, dry.
First day of sampling.

Date: September 13, 2015

Observations

Clouds arriving. The road was again dusty in the morning (not pictured). Brown haze layer is present.
Stronger winds from West in the daytime (10 AM - 4 PM).
1 PM: Very hazy now, thicker cirrostratus
3 PM: Very breezy. 70-80% coverage with altostratus.

South	West	North	East

Date: September 14, 2015

Observations

Arrival of rain. Wind picked up after 6pm.

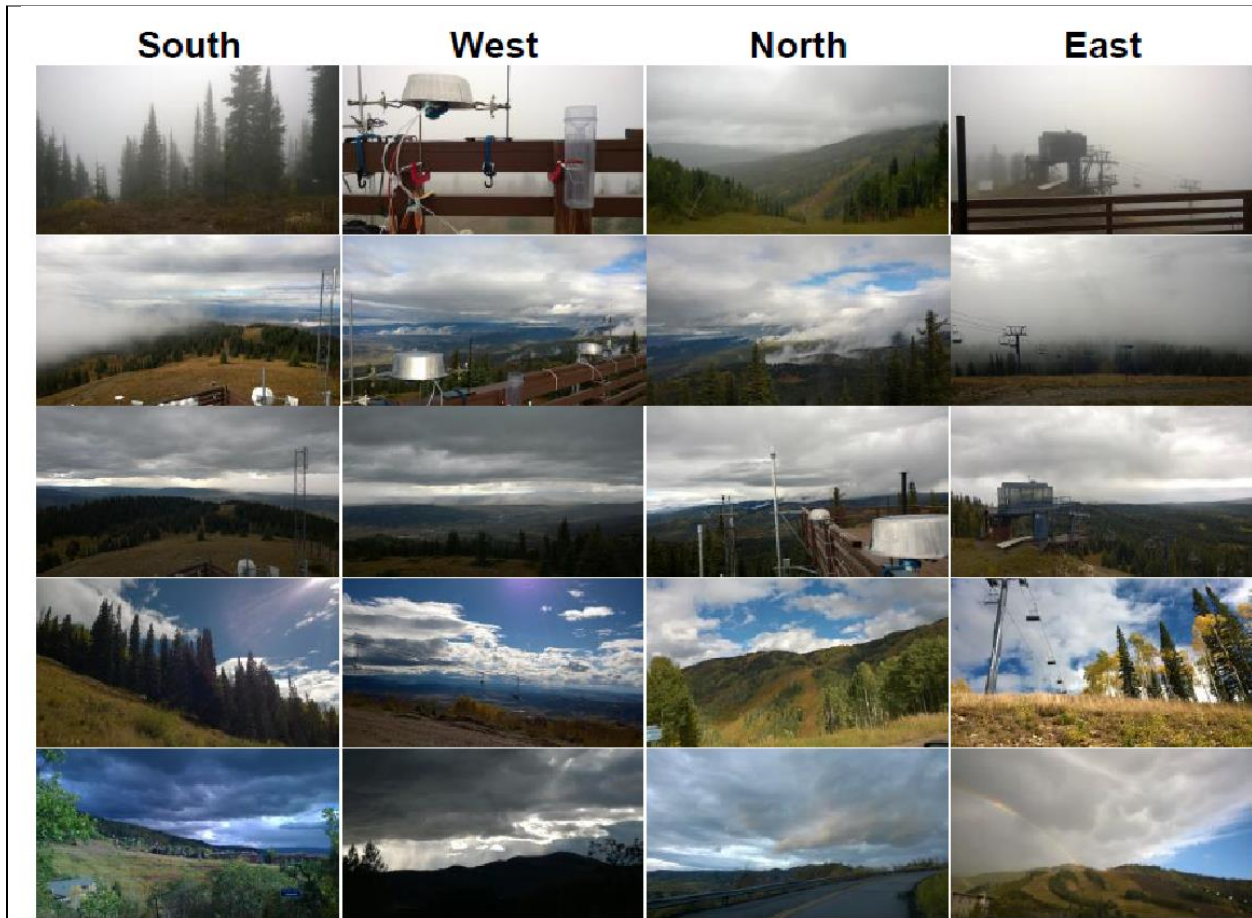
8-8:30 AM: Wind from direction of anvil cloud; transition to overcast; continued wind, clouds break up

8:45-9 AM: Vehicles arriving on dirt road; 8:50 AM: Sun breaking behind Skilift, patches of broken stratocumulus

9 AM: Clear in valley, hazy; 10:28,10:58 AM: Vehicles on dirt road; 1:20-1:50 PM: Wind dies, first drops of rain; rain stops

5:02 PM: Rain (8 minutes); 5:30 PM: Meeting. For road dust, later arrivals will park lower on road;

6:11 PM: Rain begins again, stays

| South | West | North | East |

Date: September 15, 2015

Observations

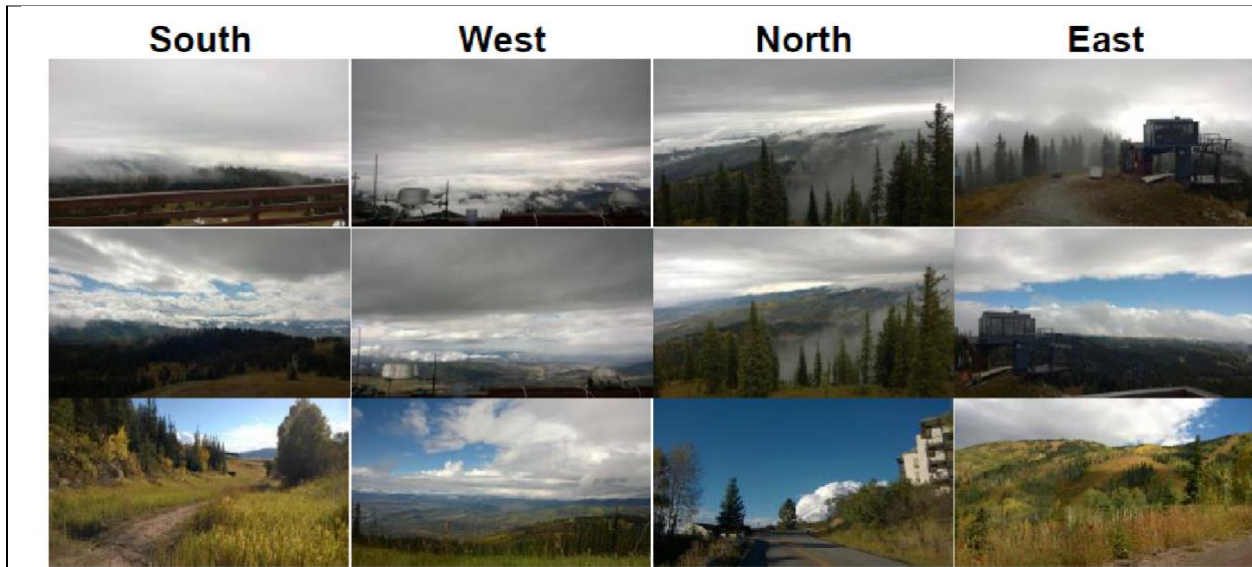
Rain predicted through 2:30 PM, "elective downtime" today (intercomparison cancelled due to road conditions).
1:35 PM: Rain
1:45 PM: Raining quite hard, sideways
2:10 PM: Separation of cloud layers

South	West	North	East

Date: September 16, 2015

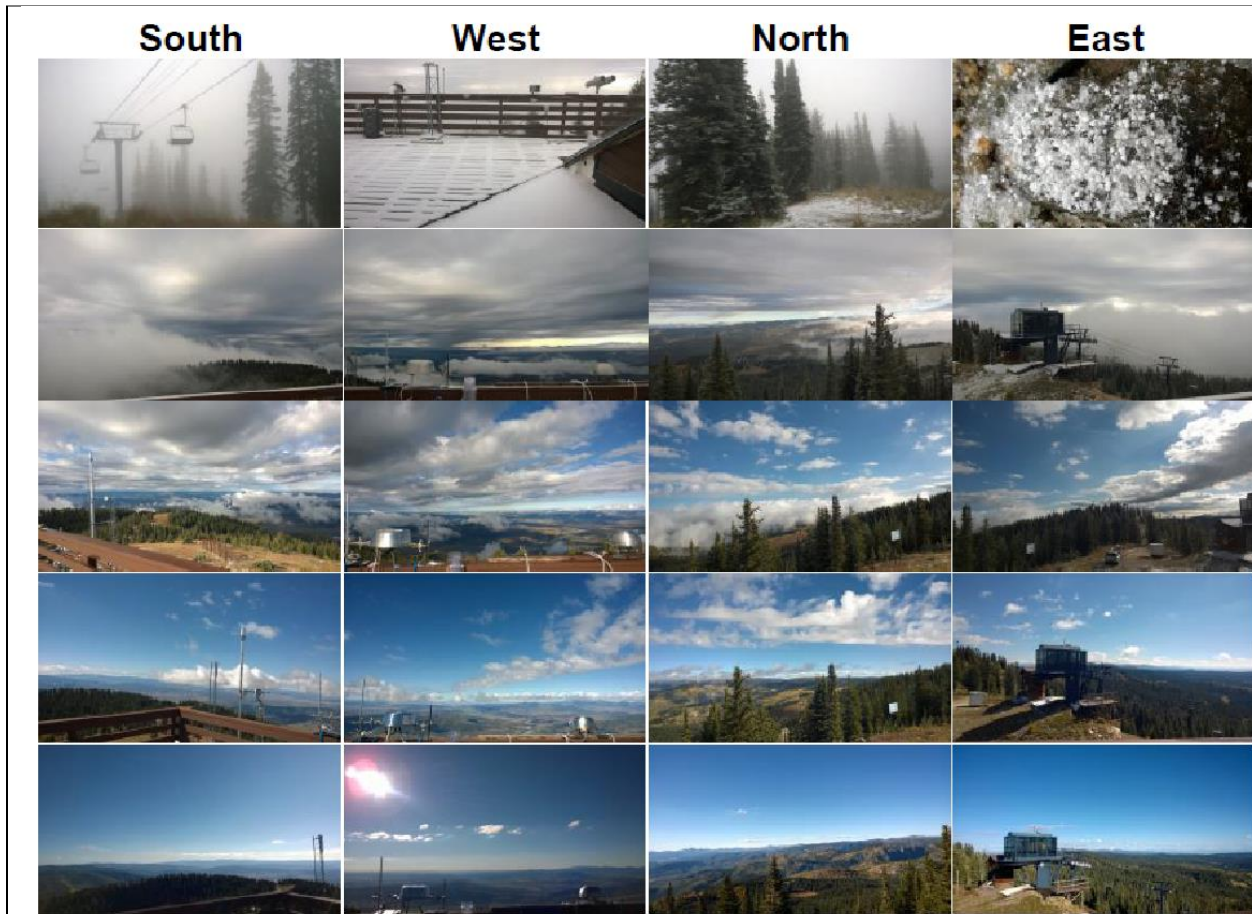
Observations

Rain intermittently.
8:44 AM: Rain approaching; wind coming from downpour to west; 9 AM: Rain has begun and stopped
9 AM: Meeting: Bike race on dirt road over weekend; Free troposphere peaks at 6am; Starting this weekend there will be smoke from California on the West wind; Children will visit on Monday
1:06 PM: Raining again; 3:18 PM: 1-mm hail, roundish, for a couple of minutes; 3:25 PM: Small amount of rain
3:40 PM: Intercomparison cancelled; SPIN background issues; 3:41 PM: Hail again, 5 minutes
4:40 PM: Rain has stopped in valley, patchy cumulus
5:05 PM: 2 minutes of hail

South	West	North	East

Date: September 17, 2015

Observations

Site closed after half day due to weather.
10 AM: Arrive in fog/rain/snow; everything is wet
10:30 AM: Rain has stopped
11 AM: Snow and sleet again
12:40 PM: Still raining

South	West	North	East

Date: September 18, 2015

Observations

Early snow, clear later.
8 AM: Arrive in snow/graupel; 8:20 AM: Snow depth 1/2 inch; 8:26 AM: Fog arrives, visibility low
9 AM: Sun is out; power plant plumes clearly visible in valley
10:32 AM: Clear sky above high cloud, clumpy low cumulus (much lower than past couple days), generally clear above, sunny, RH 51%
11:38 AM: Sunny, scattered cumulus in valley. Upper level cumulus height above mountain is constant (to East)
1:00 PM: Vehicles departing; 2:40 PM: Sunny, small amount of haze; 3:25 PM: Vehicles; 4:15 PM: Vehicle

	South	West	North	East

Date: September 19, 2015

Observations

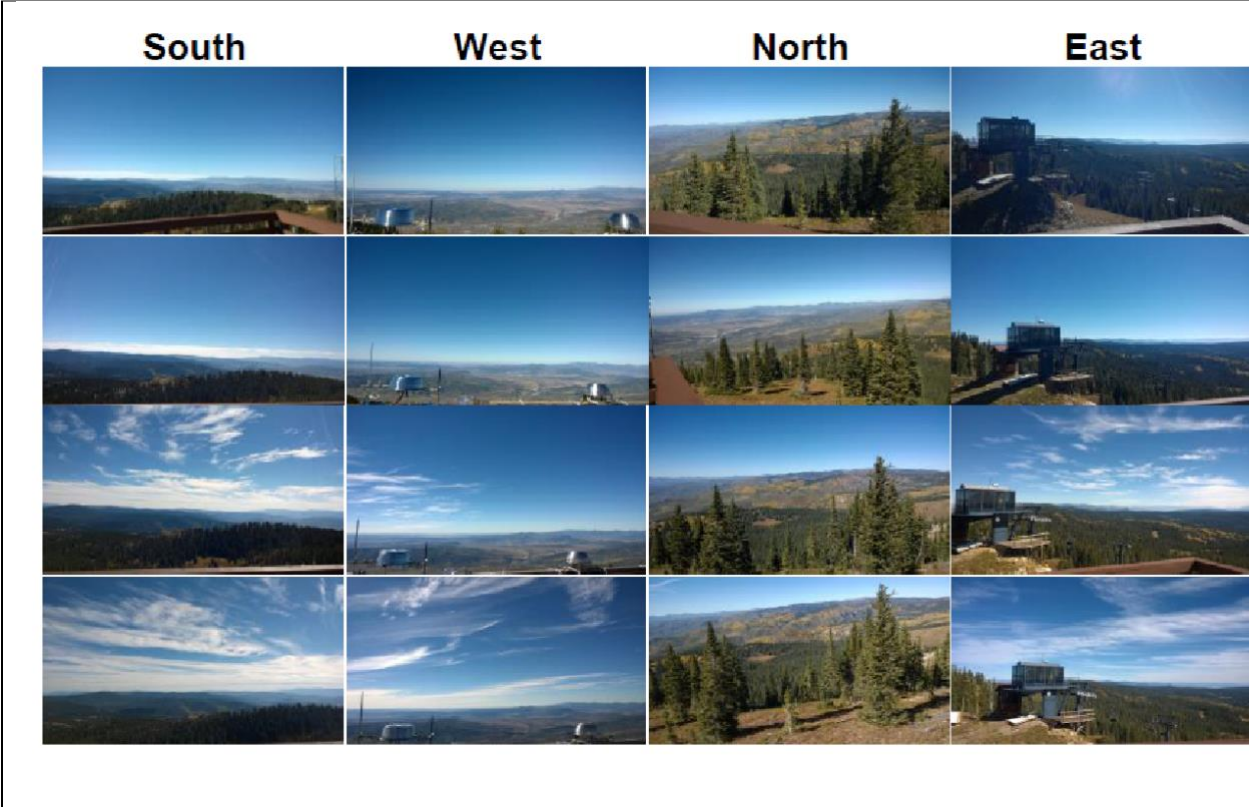
Winds from West 10am-9pm. No precipitation was anticipated.
12 PM: No rain/snow overnight

No photos taken

Date: September 20, 2015

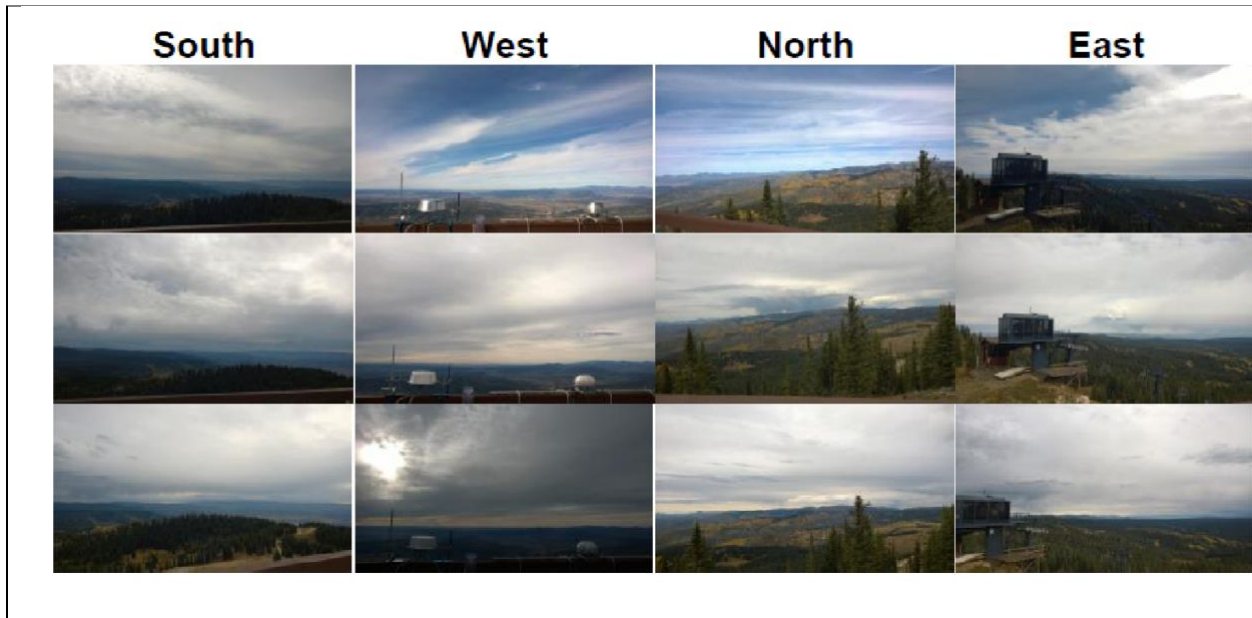
Observations

None

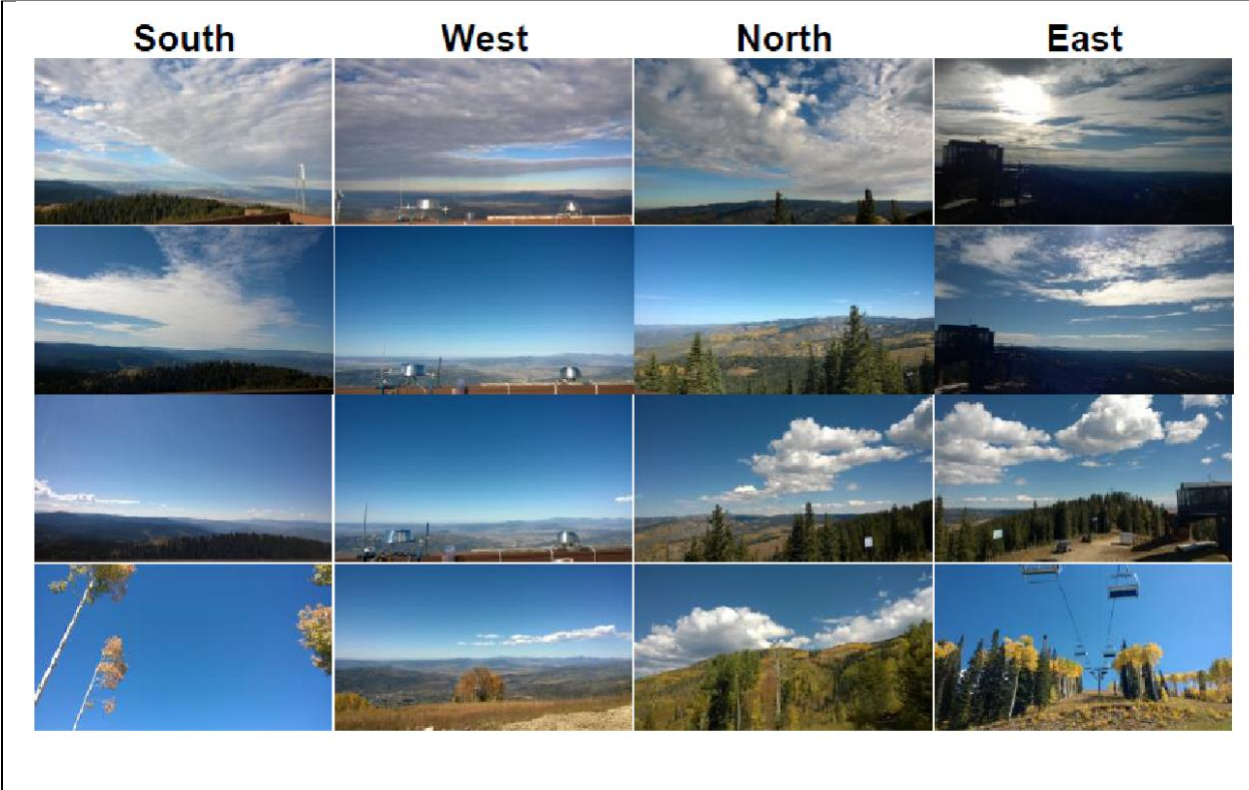
South	West	North	East

Date: September 21, 2015

Observations

Clear with high clouds.
10 AM: Clear and slightly hazy
11 AM: Vehicles

South	West	North	East

Date: September 22, 2015

Observations

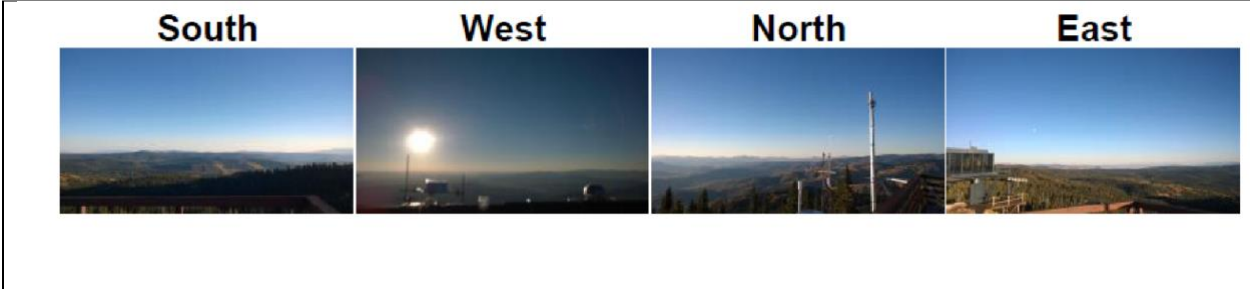
Overcast without rain.
2 PM: Hazy with a couple of clouds downwind; Ian goes to block road after some discussions with Forest Service
3:48 PM: Virga descending on horizon; wind has died; thunderstorm downwind

Date: September 23, 2015

Observations

Breezy, scattered clouds.

Date: September 24, 2015

Observations

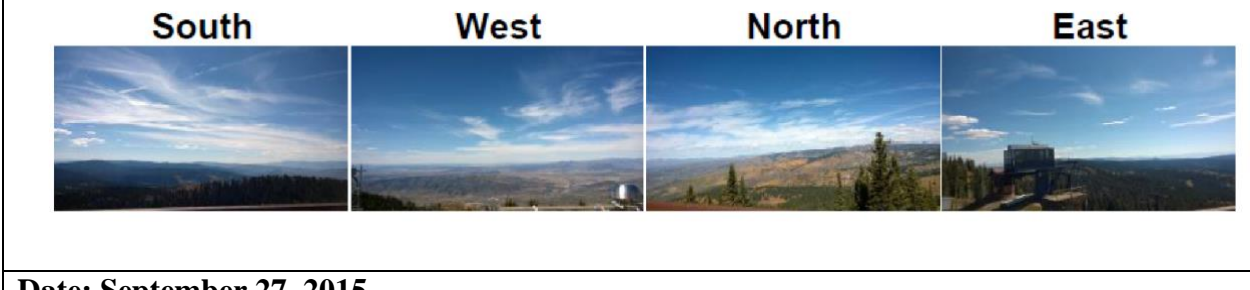
None (mandatory downtime)

Date: September 25, 2015

Observations

Hazy day. Wind from NW after 10am.
8 AM: Sunny and hazy
11:30 AM: Fairly hazy
6:20 PM: Very hazy, slight breeze, no clouds

Date: September 26, 2015

Observations

Scattered high clouds.

Date: September 27, 2015

Observations

Scattered high clouds.

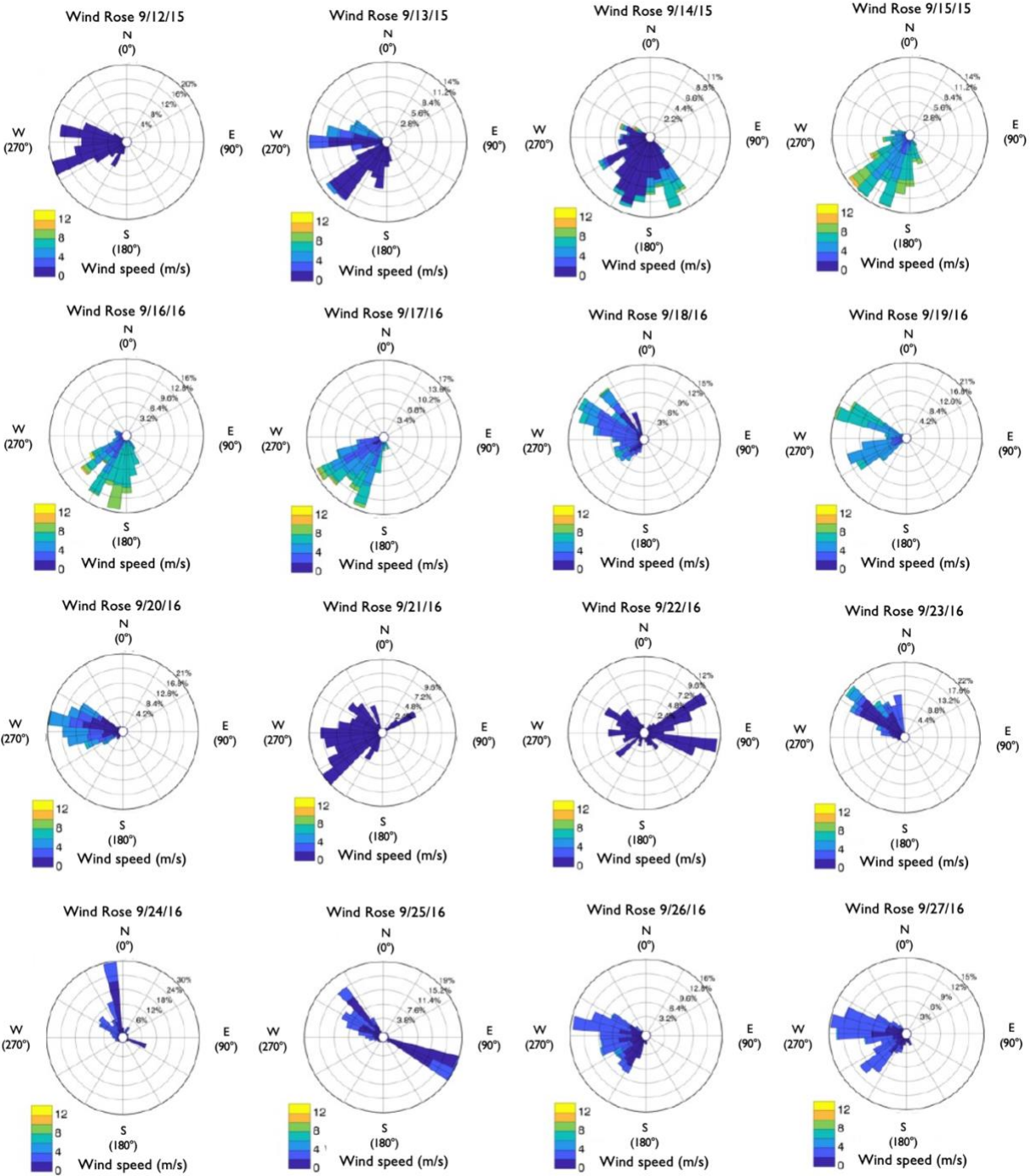



Figure S1. Wind roses displaying wind speed throughout FIN-03.

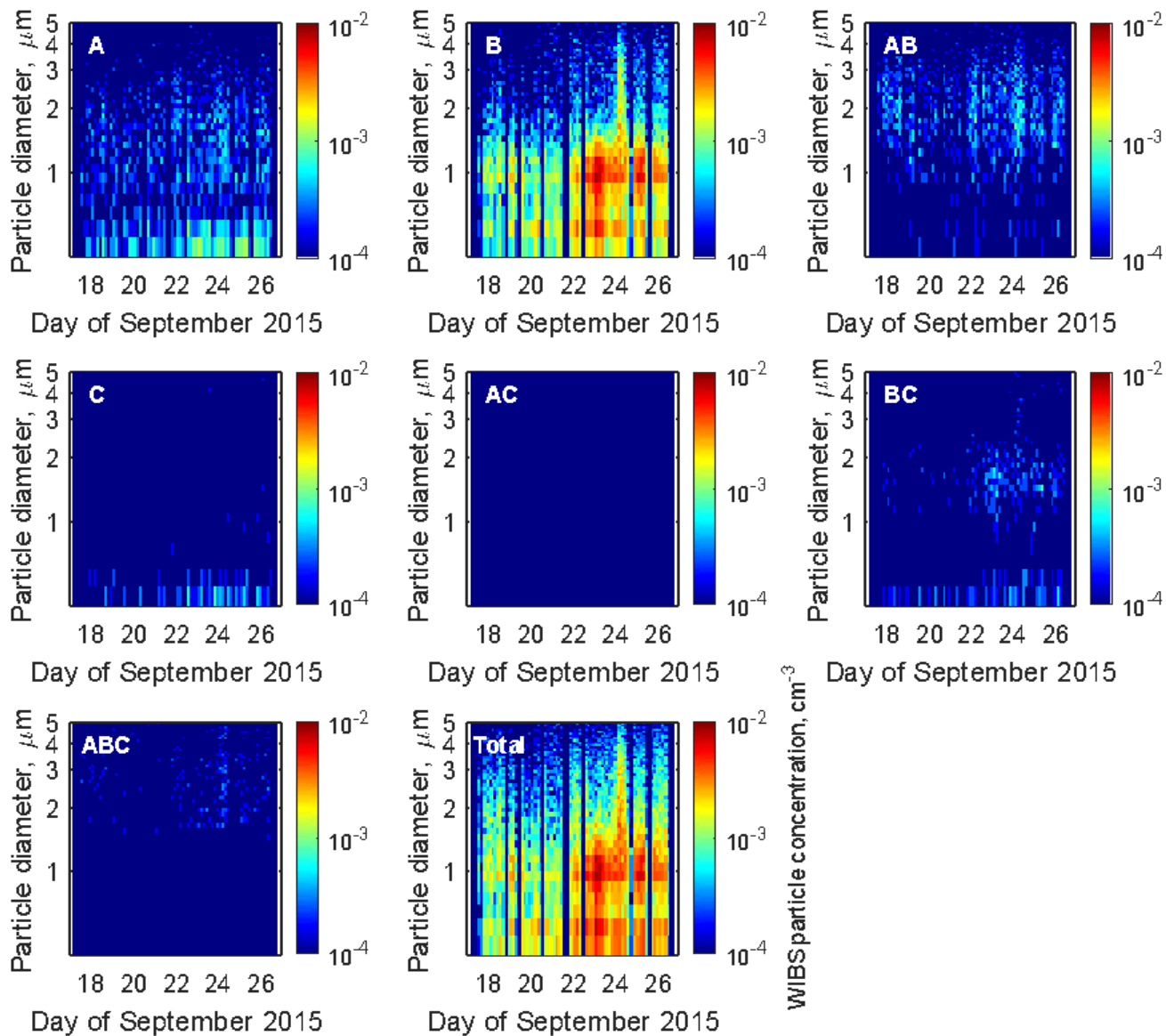



Figure S2. WIBS time series heatmaps by particle type (A, B, AB, C, AC, BC, and ABC) as well as total (all fluorescing particles measured by the WIBS which are categorized into one of the seven types). The WIBS measures the concentration of particles with diameter 0.4 - 20 µm, but the y-axis has been truncated at 5 µm to show more detail in the range that held most particles.

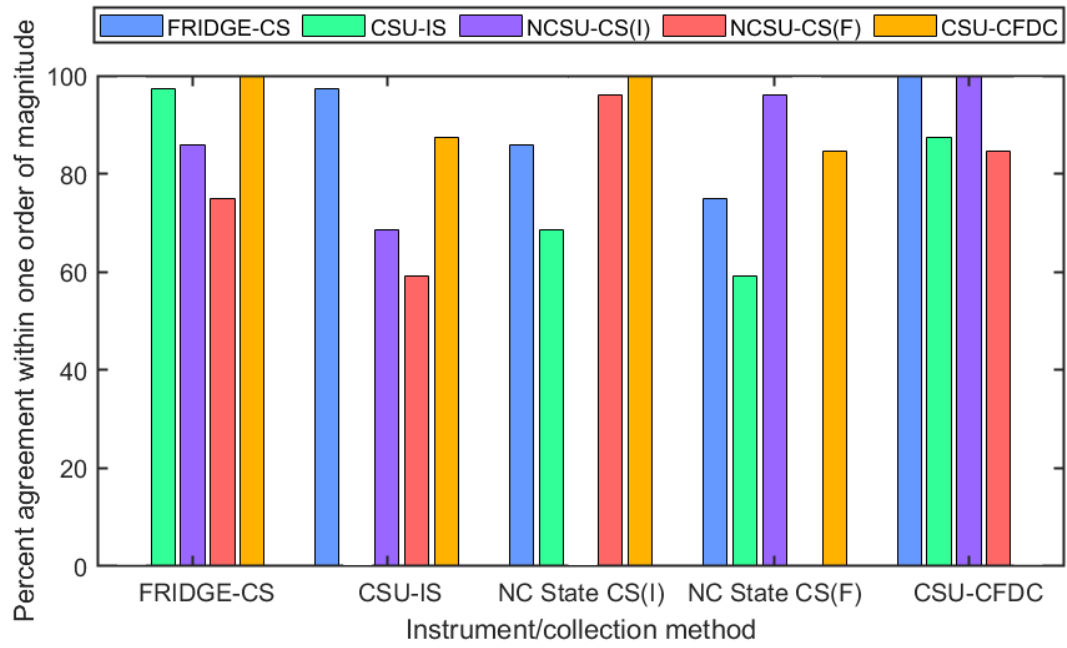



Figure S3. The percent of immersion INP measurements in which all instrument pairs agreed within one order of magnitude are shown for each pair of ice nucleation instruments.

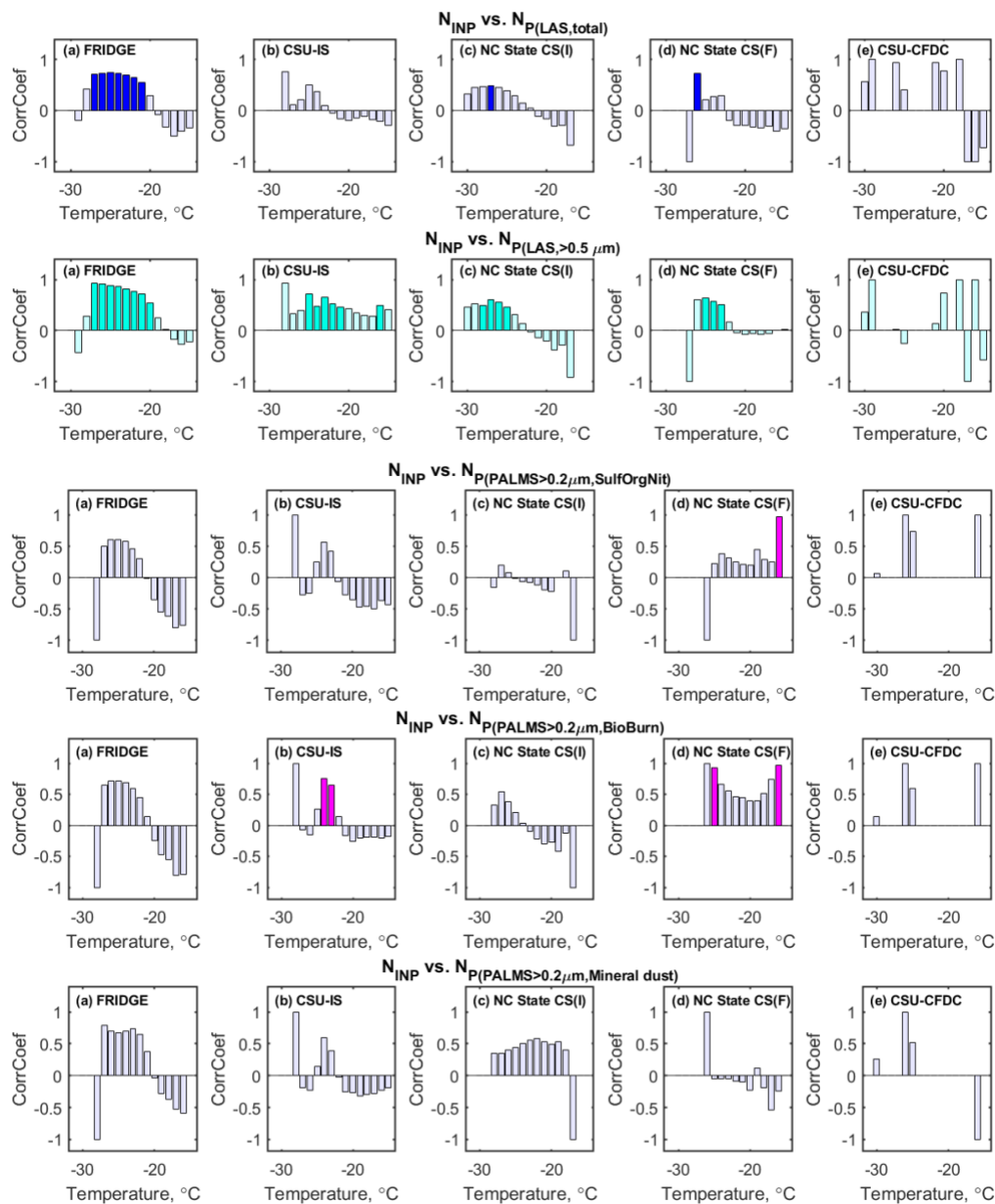



Figure S4a. Correlation coefficients in 1°C temperature bins for INPs measured by different methods with aerosol number concentrations from the LAS in all size bins and at sizes > 0.5 μm, and with number concentrations of PALMS sulfate/organic/nitrate, biomass burning and mineral dust particle types at sizes > 0.2 μm (Bright-colored bar = p < 0.05, Light-colored bar = p > 0.10 or p = NaN when there were too few samples per bin to derive the p-value)

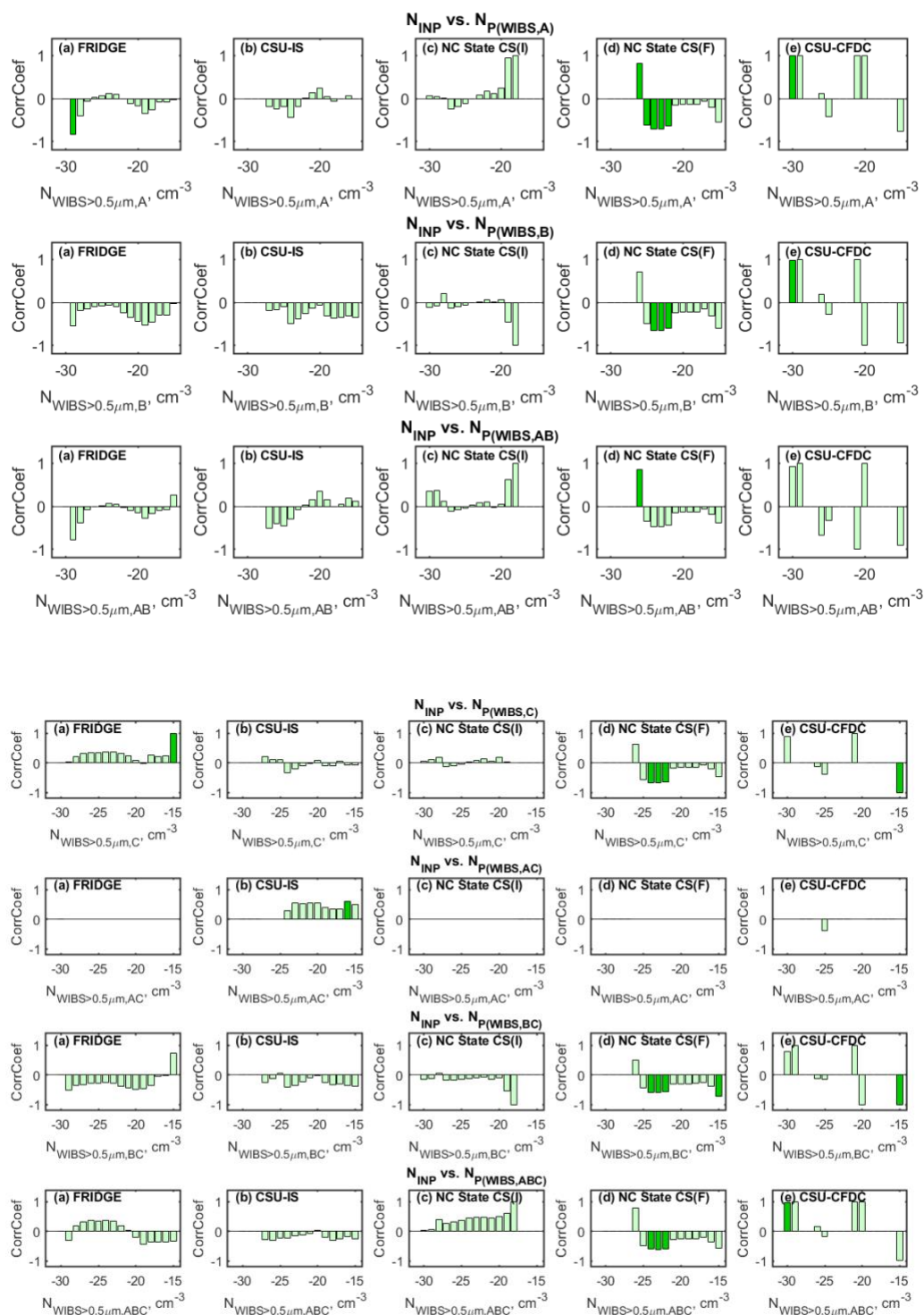



Figure S4b. Correlation coefficients in 1°C temperature bins for INPs measured by different methods with WIBS category concentrations at sizes > 0.5 μm (Bright-colored bar = p < 0.05, Light-colored bar = p > 0.10 or p = NaN)

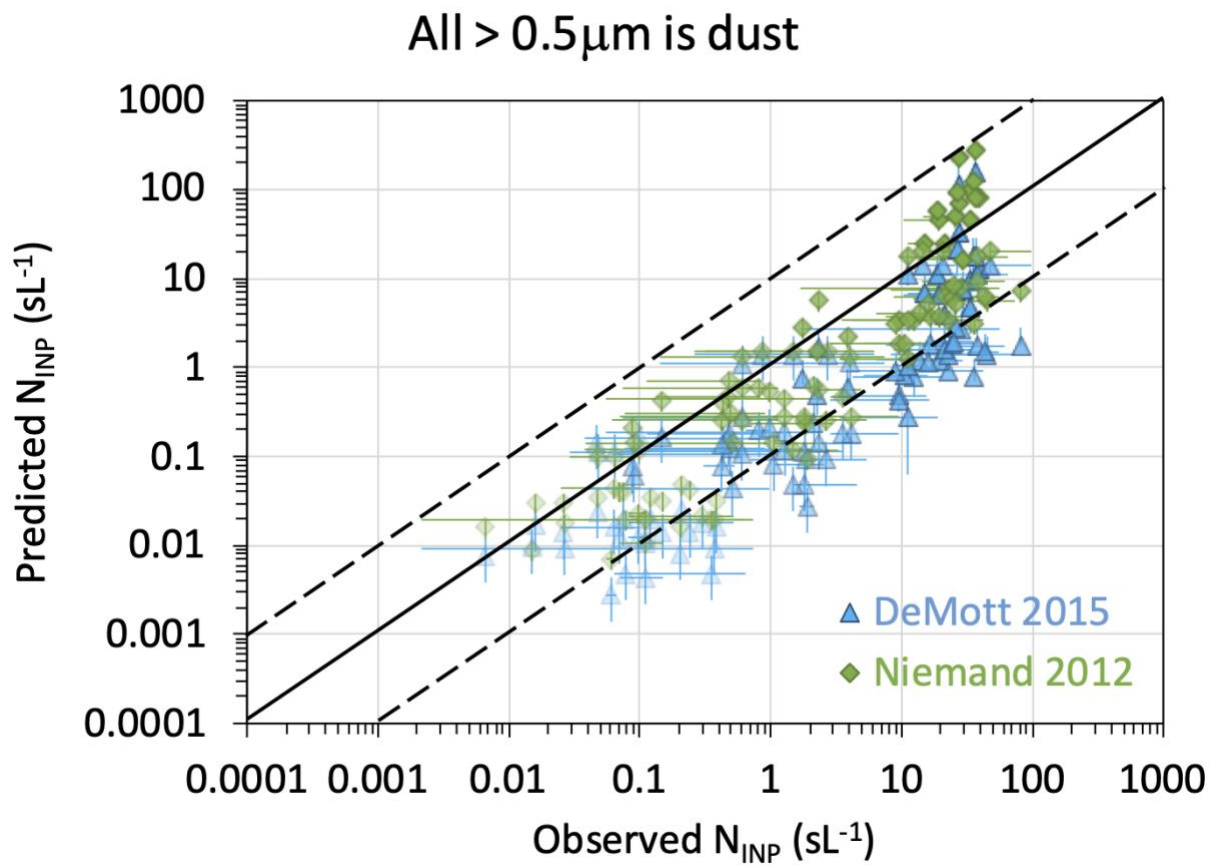



Figure S5. As in Figure 9 of the main manuscript, but for dust INP parameterizations on the assumption that all particles at sizes above 0.5 μm are dust particles.