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Captured Cirrus Ice Particles in High Definition

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1 Abstract

2 Cirrus clouds composed of small ice crystals are often the first solid matter encountered by 3 sunlight as it streams into Earth's atmosphere. A broad array of recent research has emphasized that photon-particle scattering calculations are very sensitive to ice particle morphology, 4 5 complexity, and surface roughness. Uncertain variations in these parameters have major 6 implications for successfully parameterizing the radiative ramifications of cirrus clouds in 7 climate models. To date, characterization of the microscale details of cirrus particle morphology 8 has been limited by the particles' inaccessibility and technical difficulty in capturing imagery 9 with sufficient resolution. Results from a new experimental system achieve much higher 10 resolution images of cirrus ice particles than existing airborne particle imaging systems. The 11 novel system (Ice Cryo-Encapsulation by Balloon, ICE-Ball) employs a balloon-borne payload 12 with environmental sensors and hermetically-sealed cryo-encapsulation cells. The payload 13 captures ice particles from cirrus clouds, seals them, and returns them via parachute for vapor-14 locked transfer onto a cryo-scanning electron microscopy stage (cryo-SEM). From 2015-2019, 15 the ICE-Ball system has successfully yielded high resolution particle images on nine cirrus-16 penetrating flights. On several flights, including one highlighted here in detail, thousands of 17 cirrus particles were retrieved and imaged, revealing unanticipated particle morphologies, 18 extensive habit heterogeneity, multiple scales of mesoscale roughening, a wide array of 19 embedded aerosol particles, and even greater complexity than expected. 20

21 1. Introduction

22 Understanding of cirrus cloud microphysics has advanced dramatically in the past several 23 decades thanks to continual technical innovations in satellite remote sensing, in-situ aircraft measurements, sophisticated laboratory experiments, and modeling that incorporates this new 24 25 wealth of data. In combination, the au courant picture of cirrus clouds has emerged: a highly 26 complex system that results in a vast array of cirrus formations, varying in time and location 27 through interdependent mechanisms of microphysics, chemistry, dynamics, and radiation (e.g. 28 Heymsfield et al. 2017). While the net magnitude of cirrus radiative forcing is clearly not as large as thick low-altitude clouds, an intricate picture of climate impacts from cirrus is coming 29 30 into focus. It now seems clear that both the sign (positive or negative) and strength of cirrus 31 radiative forcings and feedbacks depend on variables that can change with a wide array of 32 parameters: geography, season, time of day, dynamical setting, and the concentrations, shapes, 33 sizes, and textures of the cirrus ice particles themselves (e.g. Burkhardt and Kärcher, 2011; 34 Harrington et al. 2009; Järvinen 2018b, Yi et al. 2016). Furthermore, many of these factors may 35 be changing markedly over time, as contrail-induced cirrus and changing temperature, humidity, 36 aerosol in the high troposphere are affected by evolving anthropogenic influences (Randel and 37 Jensen, 2013; Kärcher et al. 2018, Zhang et al. 2019). Undoubtedly, a sophisticated, high-38 resolution understanding of cirrus is critical to accurately model the impacts to global and 39 regional climate.

40 Satellite-derived measurements of cirrus properties have become vastly more 41 sophisticated with the advent of increased spatial and temporal resolution, a broader array of spectral channels, specialized detectors, and advances in scattering theory (e.g. Yang 2008; 42 43 Baum 2011; Sun 2011; Mauno 2011; Yang 2013; Cole 2014; Tang 2017, Sourdeval et al. 2018; 44 Yang et al. 2018). Where a generation ago it was challenging to even isolate the presence of 45 cirrus clouds in much satellite imagery, it is now routine to derive estimates of ice cloud optical 46 depth, cloud top temperature, cloud top height, effective particle size, and in some cases even to infer the dominant particle habit and roughness of crystal surfaces (McFarlane 2008; King 2013; 47 Cole 2014, Hioki et al. 2016, Saito et al. 2017). The emerging ubiquity of this sophisticated 48 49 satellite data and highly-developed retrieval schemes can sometimes obscure the fact that major 50 fundamental uncertainties remain regarding cirrus microphysical compositions and their intertwined dynamic evolution. In reference to scales of observation and small physical features 51

52 on ice particles we refer to refer to several distinct regimes, defined as follows: nanoscale, 1-100 53 nm; mesoscale, 100 nm – 1 μ m; and microscale, 1 μ m – 500 μ m.

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55 Cloud particle imaging probes on research aircraft have also contributed to major leaps in 56 understanding, helping to constrain cirrus property satellite retrievals and climate modeling 57 representations (Baumgarnder et al. 2017; Lawson et al. 2019). These probes deliver particle 58 imaging and concentration measurements that yield unique insights into ice particle habits and 59 distributions in cirrus, though several significant limitations remain. The SPEC Inc. CPI probes have flown for nearly 20 years and can achieve 2.3 μ m pixel size and ~5 μ m optical resolutions 60 61 and SPEC's 2D-S stereo imaging probe yields 10 µm pixel sizes (Lawson et al. 2019). For 62 example, CPI images of cirrus ice were featured on the June 2001 cover of the Bulletin of the 63 American Meteorological Society (Connelly et al. 2007) and have contributed to many other 64 cloud physics field programs since (for complete list, see Appendix A in Lawson et al. 2019). 65 Other recent in-situ particle measurement innovations include the HOLODEC (Fugal 2004), 66 SID3 (Ulanowski et al. 2012, Järvinen et al 2018a), and PHIPS-Halo (Schnaiter 2018), with 67 imaging resolutions on the order of 5-10 microns, as well as multi-angle projections, and indirect scattering measurements of particle roughness and complexity. High speed aerodynamics and 68 69 concerns about instrument-induced crystal shattering have produced some uncertainties 70 regarding inferred particle concentrations, size distributions, and orientations, but perhaps more 71 importantly, the limited optical resolving power means that in-situ imaging instruments are not 72 able to determine fine-scale details of crystal facet roughness or highly complex habit geometry, 73 particularly for small ice crystals. Several groups have also achieved recent in-situ measurements 74 of cirrus particles using balloon-borne instruments (Miloshevich and Heymsfield 1997; Cirisan 75 et al. 2014; Kuhn and Heymsfield 2016; Wolf et al. 2018). Though this has been a relatively 76 sparse set, some slight momentum appears to be building toward exploiting advantages of this 77 slower-speed probe.

The synthesis that has been emerging describes cirrus clouds that are often, but not
always, dominated by combination of complex particle morphologies, and with crystal facets that
usually show high roughening and complexity at the microscale (Baum et al. 2011; Yang et al.
2013; Yi et al. 2013; Tang et al. 2017; Heymsfield et al. 2017; Lawson et al. 2019). Particle
complexity has been considered to encompass an array of potential geometric deviations away

83 from a simple hexagonal, single ice crystal: intricate polycrystalline morphological shapes, 84 aggregations of individual particles, partial sublimation of particles, post-sublimation regrowth 85 of microfacets, and inclusions of bubbles and aerosol particles (Ulanowski et al. 2012; Schnaiter et al. 2016; Voitlander et al. 2018). Even where crystals may present mainly planar facet 86 87 surfaces, these surfaces are often characterized by regular or irregular patterns of roughening at 88 multiple scales. All aspects of increased complexity and roughening have been shown to smooth 89 and dampen the characteristic peaks in the scattering phase function of hexagonal ice crystals 90 (van Diedenhoven 2014). The angular integral of the phase function yields the asymmetry parameter, which has been broadly applied as an indicator of net radiative impact of underlying 91 92 particle microphysics (Baran 2015). With mesoscopic crystal roughness and complexity 93 contributing to less total forward scattering, the asymmetry parameter and net downwelling 94 radiation is reduced (e.g. Yang and Liou 1998; Um and McFarquhar 2011, van Diedenhoven et 95 al. 2014). The calculated impacts on cirrus cloud radiative effect are shown to be 96 climatologically significant compared to assumptions that cirrus composed of less complex 97 crystals (Yang et al. 2013; Järvinen et al. 2018b). Furthermore, beyond questions of particle 98 morphology and radiative balances, major uncertainties around cirrus cloud evolution remain 99 regarding particle nucleation pathways and the interconnected roles of aerosol chemistry, high-100 altitude humidity, and the subtle dynamics of vertical motion and turbulent eddies in cirrus.

101 2. ICE-Ball in-Situ Capture Methods

102 2.1 ICE-Ball System

103 The ICE-Ball experiment has been designed, refined, and implemented from 2015-2019. The 104 basic system consists of a ~2 kg payload ("Crystal Catcher") carried aloft by a 300 g latex 105 weather balloon. The payload components are enclosed in a mylar-wrapped Styrofoam cube 106 (Fig. 1) to prevent electronics from freezing and to comply with FAA regulations for weight, 107 density, and visibility. Figure 1 shows the ICE-Ball system ready to launch, along with a cross-108 section diagram of the cryo-collection and preservation mechanism. The instrument suite 109 consists of standard balloon sonde sensors (pressure, temperature, and dewpoint), and also 110 includes HD video (GoPro Session) and dual real-time GPS position tracking (SPOT and 111 GreenAlp). The cryo-capture vessel and ice encapsulation cell comprise the novel ice particle 112 capture and preservation mechanism. Several versions of this mechanism have been employed, but in each case, it has consisted of a vacuum-insulated stainless steel vessel (250 ml volume) 113

filled with crushed dry ice and containing a custom-machined sweep tube and ice encapsulation
cell. The sweep tube extends slightly above the top of the payload, and passively collects
particles in its path due to the upward motion of the balloon (~5 m/s). When the collection

aperature is open, the particles settle to the bottom of the collection tube and are gravitationally

118 deposited in the ice encapsulation cell, which is nestled in the surrounding dry ice. The

encapsulation cell interior diameter is 7 mm, and has an open volume of 0.2 cm^3 .

120 During ascent, the balloon is ~ 5 m above the payload and does not appear to affect 121 particle concentrations impacting the top of the payload. Several sweep tube geometries and 122 opening sizes have been tested (from 0.5 to 5 cm²), but in each case, computational fluid 123 dynamics streamline modeling and sample analyses suggest that collection efficiencies are high 124 for particles larger than 50 µm diameter and decrease to less than 10% for particles smaller than 125 20 µm diameter (Supplement 3.E.). Cirrus cloud conditions and the in-flight collection operation 126 is recorded via the go-Pro video. Cirrus particles are routinely observed passing the camera, and 127 either 22° halos and/or circumzenith arcs can often be observed on the video record of each successful flight. 128

129 2.2 Ice Crystal Preservation

130 The apertures to the cryo-vessels' sweep-tubes can be opened and closed using a rotational servo 131 motor that is driven by an Arduino microprocessor (a previous version used robotic clamshell 132 seals, as seen in Supplement 2 video). The Arduino is programmed to open the path to each 133 collection vessel individually at cirrus altitudes that are prescribed before each launch. 134 Immediately after transiting the prescribed collection zone(s), the apertures are closed and a magnetic sphere is dropped down the collection tube to seal the collected crystals in the small-135 136 volume encapsulation cell (see Fig. 1b). This onboard preservation system has been tested to 137 preserve collected crystals in pristine condition for approximately 6 hours, which usually 138 provides ample time for recovery. Upon ICE-Ball landing and recovery, the small volume 139 encapsulation cell is hermetically double-sealed and stored in dry ice to ensure that crystals are 140 preserved as pristinely as possible. After returning to the lab, the sealed ice-crystal samples can 141 also be stored in liquid nitrogen for medium-term storage of up to several days prior to transfer 142 and imaging in the cryo-SEM.

143 2.3 Flight Record

Intensive field campaigns were conducted during June and July of 2016-2019, consisting of 5-10 144 145 flights per campaign. In order to proceed with mission launch, the following conditions were 146 required: 1) greater than 50% projected cirrus coverage at the time of launch, 2) horizontal wind speeds (trajectory mean) less than 30 m/s, 3) modeled trajectory allowing for a safe launch zone 147 148 and an open landing zone within a 1 hour drive of TCNJ, 4) FAA/ATC approval, requiring flight 149 plan filing 24 hours prior to launch. Conditions that prevented launches on particular days 150 mainly included high wind speeds at altitude, and clear skies or poorly predicted cirrus cloud 151 coverage. During mid-Atlantic summer, high altitude mean wind speeds meet the speed 30 152 m/smaximum launch threshold approximately 60% of the time; regional climatological 153 proximity to the jetstream often results in prohibitively high winds in the upper troposphere 154 during other seasons. High wind speeds result in a longer flight trajectory (a typical 25 m/s mean 155 wind yields an ~80 km flight), degrading landing zone accuracy (nominal landing position 156 prediction error radius of 10% of the trajectory length). Longer flight paths also require 157 additional drive time and increase the risk of landing in an inaccessible or unsafe location (e.g. 158 Atlantic Ocean, Military Base, Airport, or Interstate). In the summertime mid-Atlantic region, 159 cirrus coverage is approximately 20%. The accuracy of cirrus coverage forecasts by NCEP operation weather models (GFS, NAM, and HRRR) were found to be a significant challenge to 160 161 launch planning. Models of high-cloud forecasts appear not to produce significant skill beyond 162 ~48 hour lead times, though it is likely that these fields have not been refined as carefully as 163 others due to modest influence on surface weather.

164 It is important to note that this flight planning framework meant that the most successful 165 ice-particle collections have occurred in moderately thick synoptic cirrus cloud systems. This is 166 the case for the focal data set in Fig. 2, Fig. 3, and table 1, and several of the additional data 167 shown in the Supplement (1A & 1D) also constitute moderately thick frontal cirrus, although in 168 none of the sampled cirrus were thick enough to be optically opaque. It is likely that some of 169 these systems have include liquid-origins, which may be contributing to particle complexity (e.g. 170 Luebke et al. 2016 and Wernli et al. 2016). Several of the Supplement data collections are also from thin, high, or scattered cirrus (1B,1E,1F,1G) or convective-origin cirrus (1C). In order to 171 172 further analyze, quantify, and model the implications of ICE-Ball data it will be essential to 173 target a broad range of cirrus clouds at various heights, thicknesses, growth/dissipation stages, and dynamical origins (Spichtinger and Gierens, 2009). 174

175 The novel experimental system has failed to recover ice crystals on more occasions than 176 it succeeded (38% crystal recovery rate). As the team gained more experience, the success rate 177 improved (65% during the final campaign), but systemic experimental challenges remain. 178 Conditions that resulted in failure to capture or recover cirrus ice crystals were somewhat varied: 179 system technical failures including premature balloon bursts and frozen electronics (6 180 occurrences); ICE-BALL landing zone (often high in a tree canopy) resulted in recovery time 181 that was too long to preserve crystals (6 times); flight trajectory missed scattered cirrus clouds (4 182 times); failure of Cryo-transfer or SEM outage (2 times). Perhaps the most difficult obstacle to 183 the further development and deployment of the experimental system is the challenge associated 184 with difficult to access landing zones. This is especially challenging in the mid-Atlantic where 185 geography results in only small pockets of public property and high fractions of tree coverage. 186 Remarkably, all 28 flights were eventually recovered, but 4 of these included instances of the 187 system caught higher than 15 m up in a tree, which typically resulted in a complex multi-day 188 effort to retrieve.

189 2.4. Vapor-lock transfer and cryo-SEM imaging

190 A unique cryo-SEM imaging capability for captured samples is provided by a Hitachi SU5000 SEM, equipped with a Quorum 3010 Cryosystem and EDAX Octane Enegry Dispersive 191 192 Spectroscopy (EDS). The Hitachi SU5000 employs a Schottky field emission electron gun and 193 variable pressure sample chamber. The combination of the variable-pressure FE-SEM chamber 194 with the Quorum cryosystem is a unique configuration that allows samples to be transferred, 195 held, and imaged uncoated at very low temperature (usually approximately -160°C), while 196 simultaneously ensuring that excess water vapor is not deposited or removed from the sample 197 surfaces. The Quorum 3010 Cryosystem integrates a cryo-airlock that transfers a frozen 198 encapusulation cell into the SEM chamber while maintaining cryo-cooling and hermetic sealing 199 throughout the transfer process. Once the SEM chamber has been loaded with the crystal sample 200 and balanced cryo-temperature and pressure achieved, the magnetic seal is removed and imaging 201 can commence.

Electron beam accelerations of 2kV - 20 kV have been successfully employed with Hitachi backscatter and secondary electron detectors to produce micrographs of the captured ice crystals. The backscatter images in particular produce a dramatic contrast between the ice and higher-density embedded aerosol particles that often include silica minerals and metal oxides.

The image resolutions of individual micrographs depend on multiple factors including SEM 206 207 beam energy, spot size, working distance, and beam scanning speed. Generally, lower 208 magnification micrographs near 100x magnification achieve resolutions of 500-1000 nm, while 209 moderate magnification images near 2000x have resolutions of 25-50 nm. Although used 210 somewhat less frequently for these samples due to limited field of view, higher magnification 211 images of 5000x or above routinely achieve 10 nm resolution. At magnification above 30kx, 212 resolution approaching 2 nm is possible in this configuration, however, this results in a very 213 small field of view without prominent ice facet features, and appears to alter the ice surface 214 unless very low beam energies are used. It is someone easier to achieve sub-5 nm resolution, 215 crisp focus and high contrast images without deforming the ice surfaces if the samples are cryo-216 sputter coated and then imaged in high vacuum. However, this process has not been used 217 frequently because the cryo-sputtering process appears to obscure the smallest nanoscale surface 218 roughness patterns, and also complicates the prospects for using EDS to measure composition of 219 aerosol particles.

220 2.5. ICE-Ball upgrades in progress

221 Although the ICE-Ball instrument as flown over the past several years has already successfully 222 enabled a new view of cirrus ice particles, several significant modifications to the system are 223 currently in progress. Perhaps most significantly, a high-definition, high-contrast macro-video 224 imaging system will now be integrated into the ICE-Ball payload. This imaging system will be 225 capable of measuring particle concentrations at each point during the cirrus penetrations. A 226 second key upgrade includes the ability to separate captured particles from different regions of a 227 single cirrus layer (e.g. cloud base, cloud middle, cloud top). Together, these improvements will 228 allow better correlation of cloud-scale properties with the cryo-SEM micrographs, promoting the 229 ability to use these measurements for quantitative measures and models of cirrus properties 230 (Sourdeval et al. 2018).

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232 3. Results: Cirrus Ice Crystal Capture

233 Particularly with respect to detailed visualization of mesoscale roughness and complexity, the Ice

234 Cryo Encapsulation by Balloon (ICE-Ball) probe demonstrates the capability to dramatically

enhance knowledge of fine-scale details of cirrus ice particles. In the four successful collection

flights from November 2015-August 2017, small numbers (min. 3, max. 20) of intact ice crystals

- were recovered and imaged by Cryo-SEM. In Spring 2018, the collection aperture was
- significantly enlarged, which resulted in collection of thousands of crystals on six successful
- flights during Spring and Summer 2018-2019. The flight on April 24, 2018 was particularly
- successful, and provides the focus of the results presented here (Fig. 2, Fig. 3, and Table 1) due
- to the large number of very well-preserved crystals and the synchronous alignment with well-
- 242 defined NASA A-Train satellite measurements.
- 243 The other successful recoveries also yielded significant data, including some marked differences in the morphology of ice crystals captured from the high-altitude clouds. Example ice particle 244 images for these additional flights are provided in supplemental data, along with a description of 245 246 the synoptic context. Within this sample set, high thin in-situ cirrus (Fig. 5b & 5c., Supl. 1-E and 247 1-G) and ice particles within proximity of convection (Suppl. 1-C) tended to be smaller and more 248 compact than examples collected from actively growing warm-advection cloud shields (e.g. Fig. 249 2, Fig. 3, Table 1, and Suppl. 1-D). The lone convective-origin ice cloud that was sampled 250 (Supl. 1-C) included several high-resolution images of frozen droplets, but did not capture ice 251 particles from a well-developed cumulonimbus anvil. Although it may be challenging to get the 252 instrument into an ideal position, future ICE-Ball flight missions will target anvil outflows, 253 aiming to gather high-resolution details of convective-origin ice that have implications in the 254 thunderstorm electrification process (Stith el at. 2016 and Um et al. 2018).
- 255 **3.1** Synoptic Atmospheric Context on 4/24/2018
- 256 On the morning of April 24th, 2018, a surface low pressure system was moving from the 257 Carolinas toward the Northeastern United States. Warm advection aloft generated a shield of 258 ascending air to the north and east of the low, resulting in the emergence of a large region of 259 cirrus and cirrostratus. At mid-morning over central New Jersey, this cirrus deck extended from a 260 9.3 km base to a 11.5 km cloud top, with an optical depth between 3 - 40 (NOAA/CIRA analysis 261 algorithms on GOES-16 data and MODIS Terra). Satellite images, skew-T diagrams, and back-262 trajectories for this flight context are provided in Supplement 3. For much of the morning, a 263 faint 22 degree optical halo was visible from the ground in the filtered sunlight, and is also 264 clearly visible from in-flight video (available in supplemental data). The ICE-Ball system was 265 deployed at 11:05 am from near Bordentown, NJ. The payload ascended at approximately 6 m/s, 266 penetrating the ~2 km thick cirrostratus near Ewing, NJ at 11:45 am. Winds at this altitude were 28 m/s from the south, with a cloud base temperature of -40°C and a cloud top temperature of -267

55°C. Video from the flight payload recorded ice particles impacting ICE-Ball for approximately
7 minutes as the instrument ascended through the cirrus thickness. While the 22 degree halo was
clearly evident, no distinct circumzenith arc was visible on this flight, which was often observed
in video at altitude on other ICE-Ball cirrus penetrations (for example in the Supplement 2:
Flight video montage). The balloon burst at 14 km altitude, and the payload descended via
parachute, landing in Hillsborough, NJ. Recovery occurred approximately 10 minutes after
landing, and the captured and sealed ice particles were transferred into the Cryo-SEM for

imaging at approximately 3:00 pm.

276 **3.2 Multiform and Intricate Particle Morphology**

277 Captured ice particles from 4/24/2018 and from other flights show striking morphological 278 diversity and complexity. Particularly in instances where thousands of ice particles were 279 collected from a single cirrostratus (e.g. Fig. 2), it is clear that the imaged particles represent just 280 the top-most section of the cloud ($\sim 2\%$ of 4mm deep collection is visible in Fig. 2), with 281 particles collected from the lower and middle parts of the cloud buried below the particles on 282 top. Despite a collection mechanism that principally reveals particles from near the top of a 283 single cirrus layer, an extraordinarily wide variety of habits are apparent from each single cloud penetration, including particles of nearly every cirrus habit classification that is already 284 285 recognized (e.g. from Bailey and Hallett, 2009) and several other discernible geometric forms 286 that have not been reported elsewhere. Among the most striking features of the particle images is 287 that every aspect of particle morphology is present in multifarious patterns. Even from one 288 section of one cirrus cloud, and among recognizable particle habits, major inhomogeneities are 289 present including wide ranges of particle size, aspect ratio, varying degrees of hollowing, 290 trigonal to hexagonal cross symmetry, broad variations in polycrystallinity, and particles that 291 range from highly sublimated to those with pristinely sharp edges and facets. Perhaps the best 292 way to appreciate this immense diversity in particle form is through the stitched mosaic 293 micrograph from 4/24/18 (Fig. 2). This mosaic of 50 lower-magnification Cryo-SEM images 294 (100x) captures the entirety of one ICE-Ball sample collection cryo-cell, with a circular inside 295 diameter of 7.0 mm. Each individual image field is 0.97mm tall x 1.27 mm wide, with a pixel 296 resolution of 992 nm. An automated multi-capture algorithm on the Hitachi SEM drove the 297 sample stage to consistent overlap with a high-quality reconstruction; only in the bottom left of 298 the mosaic is some minor mismatch apparent. The mosaic figure uses false-color to highlight

299 several particle habits (bullet rosettes, columns, and plates) that fit classic definitions of 300 morphology; particle categories where manually identified and by consensus among 3 co-301 authors. In total, these distinct-habit particles number ~185 of the approximately 1600 individual ice particles that are distinguishable within the depth of focus visible from the top of the sample. 302 303 The remaining $\sim 88\%$ of ice particles resolved in Figure 2 include the following: a) complex 304 polycrystal assemblages, often not radiating outward from a single point (~75%), b) highly 305 sublimated particles where the original habit is no longer distinct ($\sim 5\%$), c) single bullets 306 apparently broken off from rosettes (~5%), and d) compact particles with convoluted facets 307 $(\sim 1\%)$. Comparable convoluted crystal forms do not appear to have been reported in the 308 literature and these particles are labeled as "outre olyhedral". Measurements of cross section 309 area, ellipse-fit dimensions, solidity, and aspect ratio for these particles are provided in Table 1. 310 These measurements are automatically generated by the standard ImageJ/Fiji particle analysis package on the separately false-colored Figure 2. Particles; solidity is defined as the cross-311 312 section area in the plane of view divided by the convex hull particle area enclosure. Bullet 313 rosettes with thin bullets have the lowest solidity (minimum S = 0.44), while compact single 314 crystals have solidity near S = 1. In this sample, the top focal plane reveals only the first several 315 layers of collected crystals. The full sample collection was accumulated 4 mm deep with an 316 estimated ~35 particle/mm linear packing, and thus estimates that approximately 200,000 317 individual cirrus ice particles were captured and preserved in this sample alone. The large ~ 2 318 mm solid chunk at the top-left of Figure 2 is believed to be a dislodged remnant from the 319 collection-tube machining process; no similar mm-sized solid particulate objects have been 320 observed in any other samples.

321 3.3 Surface Texture Roughness with Multiple Scales and Patterning

322 Higher resolution images reveal the topography and textures of crystal facets and edges in 323 greater detail. Even in the most pristinely faceted crystals that show no evidence of sublimation, 324 mesoscale texture on the facet surfaces is nearly always apparent at some scale. On some 325 particles and facets, the roughening is dramatically apparent, with micron-scale features in depth 326 and wavelength of the roughening pattern. On other facets, the roughness is significantly more 327 subtle, with dominant patterning at scales less than 200 nm. In addition, some particles show 328 roughness at multiple scales simultaneously. While particle complexity and micron-scale roughness are apparent at 100x, resolving the smaller-scale surface textures requires micrograph 329

resolutions of at least 100 nm and carefully tuned contrast. Figure 3 highlights varying degrees of 330 331 surface roughness in six-panel micrographs from April 24th, ranging from 250x to 20000x 332 magnification. Panel A. and B. show examples of the outre polyhedron designation; panel c. 333 demonstrates the open scrolling seen on a subset of particle facets. It is straightforward to 334 achieve crisp image focus (from both secondary and backscatter electron detectors) from 335 magnifications of 10x to 5000x in the Hitachi SU5000 with Quorum cryo-stage, operating at 10-336 20 Pa in variable pressure mode with stage temperature near -160°C. Beyond 5000x 337 magnification, crisp focus in variable pressure mode is harder to achieve, particularly while 338 balancing with a goal of avoiding high beam currents which can induce slight in-situ sublimation 339 at higher beam energy, density, and exposure times. Nevertheless, at -160°C and medium beam 340 density, ice particles have extremely low vapor pressure, and even smaller vapor pressure 341 gradients, such that they can be imaged for hours without noticeable changes in shape or surface 342 texture at the nm scale. Particles can even be re-sealed while under cryo-vacuum and removed 343 from the cryo-stage for short-term storage in low-temperature freezers or liquid nitrogen immersion. 344

345 3.4 Ice-embedded Aerosols and Particulates

346 All ice crystal retrievals (and those that did not capture ice) have also collected numerous aerosol 347 particulates. On flights when no cirrus crystals are captured, the ICE-Ball system nevertheless 348 typically captures several dozen large interstitial aerosols particles (>25 µm dia.), but very few 349 smaller aerosols (<25 µm dia.). This disparity provides high confidence that the many small 350 aerosol particles observed on ice crystals' surfaces adhered to the surfaces within the cloud and 351 not separately deposited post-capture. Although it has not yet been tractable to measure a large 352 fraction of these scavenged and embedded particulates, several dozen have been measured by 353 Energy Dispersive Spectroscopy (EDAX-EDS), revealing wide-ranging compositions that 354 include mineral dust, soot, fly-ash, and confirming previous reports of biogenic aerosol (e.g. 355 Pratt et al. 2009). Figure 4 includes three examples of aerosols collected by ICE-Ball, along with 356 EDS spectra of a fly ash aerosol (Fig. 4b) and iron-rich aerosol particle (Fig. 4c) adhering in the 357 shallow hollow of a trigonal single crystal. The particulates are highly variable in size, 358 concentration, and composition, with particles on the surface of crystals, and many additional 359 particles revealed in the residual samples left by a post-imaging sublimation process in the SEM. 360 As the complex ice particles sublimate, the embedded aerosol particulates collapse and coagulate

with neighboring particles, and leave a cohesive collection of mixed aerosol particulates near the 361 362 center of the original ice crystal. This sublimation-chemical coagulation process may point to a 363 potentially important cloud-processing effect that could occur during cirrus particle sublimation, possibly enhancing the ice-nucleating efficiency of the original particulates (Mahrt et al. 2019). 364 365 As aerosols of different origin and chemistry conjoin in close proximity under intense sunlight, 366 the post-sublimation ice particle residuals may serve as an unexpected chemical mixing-pot, 367 altering the course of their impact on subsequent cirrus formation. Ice particle residuals have 368 been captured during several previous aircraft field campaigns, but these techniques are 369 primarily restricted to small ice particles (less than 75 µm) and typically can not provide 370 morphological imaging of aerosol (Czico and Froyd 2014). With additional flights and increased 371 sampling statistics, the ICE-Ball aerosol collection technique promises to provide an important 372 complement to research on the origin and processing pathways of particulates in cirrus clouds 373 within the high troposphere and across the tropopause.

374

375 4. Conclusions

376 Perhaps unsurprisingly, this higher-resolution view of the ice particle constituents of cirrus 377 reveal new and unanticipated complexities compared to existing laboratory, aircraft, and satellite 378 measurements. The measurements from ICE-Ball do not contradict laboratory measurements 379 (Bailey and Hallett 2004) nor do they really dispute the first-order habit diagrams that encompass 380 cirrus temperatures (Bailey and Hallet 2010). Many of the recent particle observations based on 381 in-situ imaging from aircraft field campaigns and analysis are also largely corroborated (e.g. 382 Fridlind et al. 2016, van Diedenhoven et al. 2016, and Lawson et al. 2019). Nevertheless, 383 present results heighten the appreciation of cirrus particle complexity in four broad themes:

384 4.1 Immense whole-particle habit heterogeneity within single cirrus clouds

In all cases where multiple crystals were recovered, we observe that the synoptically-forced cirrus clouds contain a multiplicity of recognizable habit types, even within the same region of the cloud, and often existing outside of their expected habit temperature and pressure regime. In addition (and in concurrence with Fridlind et al. 2016 and Lawson et al. 2019), we also find that a high fraction of particles could be classified as "irregular", in that they do not fit within an established habit category. The high-resolution images demonstrate that these non-categorized particles are mainly divided between a) highly-sublimated forms where the original habit is no

longer recognizable and especially b) sharp-edged, faceted particles with complex 392 393 polycrystalline morphology that does not neatly fit in established habit categories. In most 394 instances, these polycrystalline assemblages can not truly be described as bullet or plate rosettes, 395 because the multiple crystals often do not radiate outward from a single focus, and they also 396 frequently contain plate-like and columnar crystal forms in a single particle. Furthermore, all of 397 the sharp-edged, and neatly-faceted crystals with no hint of sublimation commonly occur in 398 direct intermixture with highly sublimated particles. Due to our inherent cloud-top sampling bias, 399 this observation may only be particularly apparent at the very upper edges of cirrus clouds where 400 entrainment mixing may be prevalent and particles also affected by incoming solar radiation. 401 This upper-edge region is of particular radiative importance, especially in optically thicker cirrus, 402 where the dynamics of the upper cloud supersaturation zone and it's uncertain interactions with 403 above-cloud air may play a significant role in governing the life cycle of cirrus (Spictinger and 404 Gierens 2009; Wall et al. 2020). Despite this diverse morphology within each single cloud, the 405 set of 9 flight samples also reveals significant patterns of particle variations that appear to be 406 linked to the dynamical and air-mass characteristics of the cloud. For example, degree of aerosol 407 loading, average particle size, mean aspect ratio, in-cloud particle concentration, and degree of 408 polycrystallinity are fairly consistent within each single collection. On one flight, several sets of 409 collected particles appear as an aggregated chain (Fig. 5c; Supplement 1G.); this cirrus cloud was 410 not near active convection, but the frontal cirrus original may have derived from modest 411 convection several hours prior to collection.

412 4.2 Widespread non-hexagonal faceting, hollowing, and scrolls

413 In addition to unexpectedly convoluted whole-particles, captured ice particle sub-structures and 414 facets also show a sizeable fraction of trigonal (e.g. Fig. 4c and Fig. 5a), rhomboid (Fig. 3b) and 415 other non-hexagonal symmetries. In fact, facets with hexagonal symmetry appear to be a slight 416 minority. For example, columnar single-crystals in Figure 2 are shaded in yellow for trigonal or 417 other-shaped basal cross-sections (53) and green for hexagonal basal cross-section (30). Bullet 418 rosette cross sections also appear to follow similar proportions. For both bullet rosettes and columnar habits in Figure 2, approximately 80% of crystals demonstrate some degree of 419 420 hollowing. This proportion is similar, though slightly higher than reported by Schmitt and 421 Heymsfield (2007). Smith et al. (2015) also report experiments on the single-scattering impacts 422 of column hollowing, pointing out that greater hollowing extent tends to increase the asymmetry

parameter, but that the topographical character of the hollowing itself is also important. In 423 424 addition to typical center-hollowing, a small fraction ($\sim 1\%$) of ice particles from multiple flights 425 have prominent "scrolled" geometry (purple in Fig. 2, Fig. 5d) which has been reported in lab experiments but rarely observed in the atmosphere. Figure 5b shows a set of fairly compact and 426 427 relatively small crystals; their unusual convoluted faceting would likely not be recognizable 428 without resolutions of 1 µm or less. A recent paper by Nelson and Swanson (2019) combine lab 429 growth experiments with adjoining-surface molecular transport kinetics to explain the 430 development of "protruding growth" features at laterally-growing ice facets that may be important contributions to these secondary morphological features. This proposed mechanism 431 432 also highlights the role of growth and sublimation cycling in these formations, and helps to 433 explain the origins of terracing, sheaths, pockets, and trigonal growth, all of which are frequently 434 observed in ICE-Ball samples.

435 4.3 Mesoscopic roughening at multiple scales and diverse texturing

436 In high-magnification micrographs with resolution finer than approximately 200 nm, mesoscale 437 surface roughening on crystal facets and non-faceted sublimation surfaces is nearly always 438 apparent, but does not appear to occur at a characteristic scale-size or texture pattern in 439 individual clouds, or even on a single particle. With the smoothest, flattest facets, roughening 440 patterns may only become apparent with resolutions near or better than 200 nm combined with 441 carefully tuned contrast. In these instances, the smoothest facets show only subtle topographic 442 variations with amplitudes smaller than the wavelength of visible light (nano-scale roughening). 443 Many facets show roughness scales (amplitude and pattern wavelength) on the order of 500 nm 444 (mesoscopic roughening) and yet others reveal more dramatic roughening with scales in excess 445 of 1 µm (microscopic scale roughening). In our sample retrieval from 4/24/2018, particles in the 446 mesoscopic roughening scale range appeared most commonly. We observe that these natural 447 cirrus particles typically (but not universally) present linear roughening on prism facets and 448 radial, dendritic, or disordered roughening patterns on basal facets (Fig. 3 panels, and 449 Supplement 1). These observations of roughening are quite similar to those observed for ice 450 particles grown within environmental SEM (Magee et al. 2014; Pfalzgraff et al. 2010; Neshyba 451 et al. 2013, Butterfield et al. 2017) as well a new experimental growth chamber built specifically 452 to investigate ice surface roughening (Voigtländer et al. 2018). These observations of roughness at amplitudes and patterning agree with in-situ reports of multi-scale roughness by Collier et al. 453

454 2016. The marked similarities in roughness seen on ICE-Ball samples and lab-grown samples

- 455 substantiates ESEM and other growth chamber methods as important tools for understanding
- 456 mesoscale roughening patterns in cirrus ice growth and sublimation, especially given their
- 457 unique ability to observe facets dynamically as they experience growth and sublimation cycling.

458 4.4 Composition and morphology of embedded and nucleating aerosol

459 Cirrus particles also show high variability with respect to the presence of aerosol particulates

460 adhered to the crystal surfaces, and embedded within the sub-surface. In the cleanest cases, most

461 ice particles revealed no obvious (>50 nm radius) non-ice aerosols on the surface (e.g. Fig. 3a),

462 while in the dirtiest cases (Fig. 4a.; Supplement 1D.), each ice particle averaged several dozen

463 mineral or pollutant aerosols. Biogenic particulates are also seen with some frequency (Fig. 5e).

464 While the presence of diverse, rough, complex crystals was striking in every sample collection,

the degree of particulate contamination was highly correlated among individual sample

466 collections, suggesting that airmass-effects play a dominant role in widely-varying degrees of

467 aerosol loading. The opportunity to directly image aerosol particle morphology, relationship to

the ice particle surface, and measurement of composition may help to strengthen understanding

469 of connections between aerosol particles, ice nuclei, ice particle growth, and macro-scale cirrus

470 properties.

472 Figure 1. ICE-Ball balloon and payload photo at pre-launch (a), with co-authors Lynn, Tusay and Zhao
473 (left to right). Diagram of servo-driven sealing of cryo-capture vessels and positioning within the ICE474 Ball payload (b).



Figure 2. Mosaic of 50 Cryo-SEM micrographs of cirrus ice particles captured on 4/24/2018 from ~11
km altitude, -50°C. Each micrograph in this group was acquired at 100x magnification, with resolution of
~900 nm. Actual large circle diameter 7 mm. False color shading groups similar crystal habits, or highly
sublimated particles (orange). Grey-scale particles are sharply-faceted crystals that do not easily fit in
habit classification categories. Table 1 provides class counts and geometric measures.



495 Figure 3. Moderate magnifications (250x to 4500x) of particles, highlighting a wide variety of surface 496 roughening characteristics. (a) example of a compact-convoluted "outre polyhedron" near several bullet 497 rosettes and non-classified sharp-faceted particles. On close inspection, multiple patterns of roughness 498 visible and several mineral aerosols (bright white). (b) rhomboid column with prismatic linear roughening speckled with discrete surface adhesions, possibly from multiple growth cycles. (c) Rosette with mixed 499 aspect crystals and an array of geometric surface pits and high mesocopic roughening. (d) Geometrically 500 501 tiered and hollowed column of irregular basal cross-section with high roughening. (e) Outre polyhedron 502 with central hole and irregular roughening. F. High magnification of small, uniform angular roughening.



Figure 4. Three-panel Particle images and Energy Dispersive X-ray Spectroscopy (EDAX Octane)
statistics on ice particle contaminants. (a) 100x image of high-aerosol loading on 6/13/2018 (Supplement
1D. for additional details). (b) Fly Ash particle (not ice) captured outside cirrus cloud, with EDS
composition. (c) Shallow hollowed trigonal ice particle with iron-rich embedded aerosol (6/25/2019).



- 511 Figure 5. Ice particles with non-classical facet features. A. Trigonal crystal with 3 six-sided etch pits,
- 512 moderate roughening and aerosol loading. B. Relatively small, compact ice crystals (mean diameter \sim 55
- 513 μm) with convoluted hollowing patterns and moderate mineral dust aerosol load. C. Curving chain of a
- ~15 particle aggregate including rosettes, compact crystals, and outre polyhedra. D. Moderately
 roughened, scrolled plate with corner fins. E. Complex rosette with twisted biogenic particle (left side). F.
- 515 roughened, scrolled plate with corner lins. E. Complex roselle with twisted biogenic particle (left side). F 516 Flattened natterned heyagon with many small adhered aerosals and outre polyhedron (below)
- 516 Flattened, patterned hexagon with many small adhered aerosols and outre polyhedron (below).



520 521

Table 1. Statistics for particle habit categories in Figure 2.

Particle Type	Fig 1. Color	Count	Fit ellipse semi-major mean, μm median	Fit ellipse semi-minor mean, μm median	X-section area mean, µm ² median Tot. area%	Aspect ratio mean median	Solidity ratio mean median	Note
Columns	Green and Yellow	83	206 167	90 105	18200 10900 4.0%	2.39 2.29	.88 .90	Green columns with hexagonal cross section, yellow non-hexagonal. 90% show hollowing
Bullet Rosettes	Blue	81	189 101	90 57	18800 13400 4.0%	1.84 1.60	.69 .70	Mean of 6 visible bullets per rosette. Bullets range from thick to very thin and solid to hollow.
Highly sublimated	Orange	62	139 93	77 50	18700 3640 3.1%	1.86 1.69	.82 .85	Sublimated to extent original habit and facet shapes not distinguishable.
Plates	Red and Pink	20	218 204	142 125	29300 23800 1.5%	1.64 1.56	.88 .91	Red plates nearly hexagonal; pink are non- hexagonal.
Open Scrolls	Purple	11	183 165	124 80	21100 17900 0.6%	1.51 1.53	.89 .89	Scroll features overlap with other habits; these show dominant scroll features
Outre Polyhedra	Teal	6	250 230	214 193	43000 34200 0.7%	1.17 1.14	.88 .91	Compact particles with convoluted intersecting facets
Complex polycrystals and broken bullets	Gray	~13 00	not measured	not measured	~86%	not measured	not measured	Sharp-faceted polycrystal particles, often of mixed aspect ratio, including broken bullets (10%)

Supplementary Files

- 1. Particle images from additional flights (7-slides)
- 2. Flight video from 4/18/2018 and flight montage
- 3. Concurrent map, satellite, weather data, and collection efficiency for 4/24/2018 (5 slides)

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