

1 **Laboratory analysis** of volcanic ash particles using a 2D
2 **video disdrometer**

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27 **Abstract**

28 Radar variables of volcanic ash clouds are dependent on microphysical processes and can be
29 expressed using physical parameters of volcanic ash particles, such as terminal velocity, axis ratio, and
30 canting angle, which are necessary for quantitative ash-fall estimations. In this study, free-fall
31 experiments of volcanic ash were accomplished using a two-dimensional video disdrometer under
32 controlled conditions.

33 Samples containing a rotating symmetric axis were selected and divided into five types according
34 to shape and orientation, i.e., oblate and prolate spheroids with horizontally and vertically oriented
35 axes and spheres. The horizontally and vertically oriented particles were present in proportions of 75.5%
36 and 21.6%, and oblate and prolate spheroids were in proportions of 76.2% and 23.8%, respectively.
37 The most common shape type was a horizontally oriented oblate spheroid (57.3%).

38 The terminal velocities were classified according to shape type. The terminal velocities of prolate
39 spheroids (vertically oriented) particles were higher than those of oblate spheroids (horizontally).
40 Terminal velocities were in the range $0.5 < \text{volume-equivalent spherical particle diameter (D)} < 1$ mm
41 for OH because of an increase in axis ratio and a sharp decrease in sample size from $D < 0.7$ mm. The
42 axis ratios fell over a wide range, from 0 to 1.5, at $D < 2$ mm, but converged to 0.94 at $D > 2$ mm.

43 The histogram of canting angles followed unimodal and bimodal distributions with respect to
44 horizontally and vertically oriented particles, respectively. The mean values were close to 0° and the
45 standard deviation for the entire particle shape types was close to that of raindrops (10°) under calm
46 atmospheric conditions.

47

1. Introduction

Volcanic eruptions are considered one of the most severe types of natural disasters, and can lead to human casualties and property damage (Wilson et al., 2012; Hillman et al., 2012; Sigurdsson et al., 2015; Wilson et al., 2015). Following an eruption, fine airborne volcanic ash flows for several tens of kilometers, which can cause major problems by increasing aviation traffic (e.g., Bonadonna et al., 2012; Langmann et al., 2012); this was seen after the eruption of Eyjafjallajökull volcano in Iceland during the period April 14– 21, 2010, for example (Bonadonna et al., 2011). From the viewpoint of volcanological hazard reduction, trajectory modeling of volcanic ash clouds is an important research topic. The aim of numerical forecasting modeling is to accurately predict the transport and deposition of ash, which is vitally important for hazard mitigation (Poulidis et al., 2017). Current numerical simulations consider the terminal velocity (V_T) of particles and incorporate wind advection, atmospheric diffusion, and particle aggregation (Folch, 2012; Bonadonna et al., 2015). The V_T of a particle is affected by its shape, density particle volume fraction, and atmospheric properties, such as air pressure (P), temperature (T), and vertical flow (Wilson and Huang, 1979; Haider and Levenspiel, 1989; Ganser, 1993; Bonadonna et al., 1998; Riley, 2003; Dellino, 2005; Coltelli et al., 2008; Alfano et al., 2011; Del Bello et al., 2017). Observational analysis would be helpful for improving the performance of eruption models; however, there is limited information on the microphysical characteristics and inner structures of volcanic ash clouds (Marzano et al., 2006; 2012). Transport and sedimentation of volcanic ash are complex processes, and the residence time and fall velocity of ash is critically dependent on particle size (Bonadonna et al., 1998), where with respect to the latter, smaller particles could be flowing in the atmosphere further from the vent. Ash consists of very fine-grained fragments, volume-equivalent spherical particle diameter (D in μm) is generally smaller than 2 mm, and these are generally dominated by broken glass shards rather than crystal and lithic fragments. Bombs, resulting from larger eruptions, may show vesicularity; they exhibit flexibility similar to

72 plastic when a volcano becomes active and erupts (Sparks et al., 1997; Marzano et al., 2006; 2012).
73 The trajectories of volcanic ash clouds can be detected by meteorological satellites (Stevenson et al.,
74 2015), and the ash amounts can be observed using a ground-based paper cup and an electronic balance
75 (Tajima et al., 2015, Maki et al., 2016). Covering the middle latitudes of the entire globe, geostationary
76 meteorological satellites are able to detect water clouds over the ocean, even when they are far from
77 land; they are also more capable of detecting the upper layers of weather systems, such as convective
78 cells and typhoons, than other weather observational instruments, albeit with relatively low spatial
79 resolution (a few kilometers). When operating in near-polar orbiting mode, these sensors have higher
80 spatial resolution but are limited by their lower temporal resolution, and only pass over a given area
81 twice per day. Weather radar operates for a similar purpose to that of meteorological satellites, and
82 provides information for determining the volume, mass, and echo top height of weather systems. Short-
83 duration eruptions, i.e., less than 1 hour, can be detected at high spatio-temporal resolution, especially
84 in the early period of an eruption. The temporal resolution of weather radar is a few minutes for a
85 single volume scan, and depends on the observation strategy and radar band. The spatial resolution of
86 weather radar is a few hundred meters and is proportional to the radar frequency. A number of ash
87 cloud detections were reported in several observational cases in the US and Japan (Maki and Doviak,
88 2001; Maki et al., 2012). Marzano et al. (2013) summarized 28 major explosive volcanic eruptions
89 detected by weather radars from 1970 to 2011.

90 Harris and Rose (1983) attempted to analyze volcanic ash particle size and total mass using a C-
91 band weather radar. Maki and Doviak (2001) found that a weather radar could measure the radar
92 reflectivity factor (z in $\text{mm}^6 \text{ m}^{-3}$) of volcanic ash columns and Donnadieu et al. (2012) detected
93 volcanic eruptions using an L-band fixed radar. Marzono et al. (2006, 2012) and Maki et al. (2012,
94 2014) detected and analyzed volcanic eruptions using weather radars, from theoretical (physical) and
95 experimental (engineering) perspectives. These weather radars were developed to detect hydrometeors

96 with a size range from millimeters to centimeters. Coarse particles ($D < 0.53$ mm) can be detected by
97 weather radars. Coarse volcanic ash particles are deposited rapidly following an eruption within a few
98 tens of kilometers from the vent (Bonadonna et al., 1998; Beckett et al., 2015); however, smaller
99 particles, such as fine ash ($D < 64$ μm), are transported over distances of several thousands of
100 kilometers and cannot be detected by operational weather radar, since the minimum detectable
101 reflectivity (MDR) of radar is inversely proportional to the distance between the radar and the target,
102 and where this depends on the wavelength of the transmitted wave (Sauvageot, 1992; Maki et al., 2016).

103 Weather radars require detailed information on the following basic parameters: particle size
104 distribution (PSD), particle density (ρ_s in g cm^{-3}), dielectric constant ($|K|^2$), V_T (in m s^{-1}), axis ratio (γ),
105 and canting angle (β in $^\circ$) to estimate ash amount quantitatively, and these can all be observed by a
106 ground-based instrument. The PSD is controlled by the microphysical processes of the volcanic
107 eruption system and plays an important role in its development; it is also essential for accurate
108 evaluation of the backscattering and absorption of particles (Seliga and Bringi, 1976; Marzano et al.,
109 2006; 2012), which affect the radar variables. There are two basic PSD models: i) the gamma size
110 distribution following the gamma function (Γ) and ii) the Weibull size distribution, mainly applied as
111 a log-normal PSD. The former is as a well-known reference in radar meteorology. For precipitation,
112 several raindrop size distribution (DSD) models can be used in a variety of cases (e.g., Ulbrich, 1983;
113 Testud et al., 2001). This implies that a disdrometer is necessary to analyze the observed radar variables
114 and evaluate them in a scattering simulation.

115 The ashfall rate (R_A) can be also defined in terms of the V_T and is determined by ρ_s , shape, and
116 orientation (Ganser, 1993; Hölzer and Martin Sommerfeld, 2008; Mandø and Rosendahl, 2010;
117 Bagheri and Bonadonna, 2016; Dioguardi et al., 2017). The V_T of particles vary widely due to their
118 irregular shapes and material components (e.g., Wilson, 1972; Harris and Rose, 1983; Bonadonna et
119 al., 2011, Maki et al., 2016)

120 Volcanic ash particles have a range of shapes, and this presents a major challenge when analyzing
121 their characteristics. To address this, Böhm (1989) analyzed the aerodynamic properties of an irregular
122 hydrometeor and Huang (2010, 2015) applied this concept to snow particles. Recently, the irregularity
123 of volcanic ash particles was analyzed in detail based on the features of various regular particles, such
124 as cubes, cylinders, and disks (Bagheri and Bonadonna, 2016), using a computed tomography (CT)
125 scanner (Dioguardi et al., 2017; Garboczi et al., 2017).

126 The particle shape observed by radar depends on oscillation or tumbling and can be expressed
127 quantitatively using the β . Research on the β of volcanic ash based on meteorological radar is scarce.
128 Marzano et al. (2012) assumed that the standard deviations of the canting angle (σ_β) were 30° and 10°
129 under unstable conditions (in which the tumbling phenomenon may occur) and stable conditions,
130 respectively. For the case of raindrops, the mean value of the canting angle ($\bar{\beta}$) is close to 0° and σ_β
131 ranged from 4° to 10° (Beard and Jameson, 1983; Hendry et al., 1987; Huang et al., 2008; Ryzhkov et
132 al., 2011). Solid hydrometeors, such as snowflakes and hail, exhibited different features compared to
133 the liquid phase. The σ_β values for graupel, rimed particles, and aggregations were 20° , 16° , and 13° ,
134 respectively (Garrett, 2015). Hail that occurs in the context of the tumbling phenomenon is described
135 by $\bar{\beta} = 60(1-f_w)$, where f_w is the wetting ratio (Synder et al., 2013; Jung et al., 2008). It is assumed
136 that the existence of a liquid phase water covering on the particles reduces the tumbling/oscillations
137 phenomenon.

138 The $|K|^2$ of the particle depends on the chemical components and phase. Water and ice
139 hydrometeors have quite different values of $|K|^2$, of 0.93 and 0.19, respectively, where the value
140 depends on the radar frequency and T. Volcanic ash has a silicate content proportional to the $|K|^2$,
141 resulting in various values of $|K|^2$, such as 0.34 and 0.39 ± 0.02 (Adam et al., 1996; Marzano et al.,
142 2006; Oguchi et al., 2009).

143 Aerodynamic properties are important for safe aviation, and for studying the effects of volcanic
144 ash on climate change, since these parameters determine the residence time of ash particles in the
145 atmosphere (e.g., Folch et al., 2009). There are two approaches to studying these aerodynamic
146 properties. The first approach is physical, where a numerical simulation model is used to calculate
147 terminal velocities, drag force, and Reynolds number (Re); examples of this approach can be found in
148 Happel and Brenner (1983). Scattering simulations of particles may be aided by the results of the
149 present study, allowing accurate detection of ash fall by weather radar; this could help verify observed
150 radar variables. In particular, the T-matrix scattering simulation developed by Waterman (1965, 1971)
151 is useful for calculation of the theoretical backscattering power of non-spherical particles. The second
152 approach is related to engineering research, in which the aforementioned relationships are determined
153 experimentally; examples of this approach can be found in Dellino et al. (2005), Bagheri et al. (2013),
154 Bagheri and Bonadonna (2016), and Dioguardi et al. (2018). Particle dispersion depends on
155 atmospheric conditions, such as T and wind, which can affect the retrieval of basic parameters. It is
156 suggested that these parameters be analyzed through free-fall laboratory experiments, to reduce
157 unknown information and obtain reliable results.

158 The present study focuses on the second approach, and its purpose is to analyze the physical
159 parameters of volcanic ash particles statistically using a ground-based disdrometer, and to develop
160 quantitative ash-fall estimation (QAE) methods for accurate detection of volcanic ash clouds by
161 weather radar. The rest of this paper is organized as follows. Section 2 introduces the free-fall
162 experiment for samples collected with a ground-based disdrometer and the classification method for
163 particle shapes. Section 3 presents the V_T , γ and β results for each type of volcanic ash particle shape.
164 Section 4 summarizes the results.

165

2. Data and Methods

a. Two dimensional (2D) video disdrometer

The 2D video disdrometer (2DVD) was developed by Joanneum Research (Graz, Austria) to detect single raindrop particles, and the instrument has been modified to cover the errors caused by turbulence effects (Nešpor et al., 2000). The device is able to observe the shape, V_T , and β of a single particle using optical light. The ability to analyze a single particle is a significant advantage compared to other disdrometers, such as the Joss-Waldvogel disdrometer (JWD), the Precipitation Occurrence Sensor System (POSS), and Parsivel (PARTicle Size and VElocity). For instance, Parsivel considers a fixed measurement area without any consideration of particle shape (e.g., Tokay et al., 2014), while 2DVD observes particles by passing them through a 100 cm² observation area consisting of two light sources and reflecting mirrors and two cameras, one set 6.2 mm above the other, and collects data with a resolution of 630 pixels; this results in a pixel size of 0.2 mm at 55 kHz (Kruger and Krajewski, 2002). Particles passing through the observation area yield shape information according to the radiation intensity of the light sources, which is helpful for calculation of γ and β . The V_T of particles is calculated using the height difference between the two cameras. Based on these advantages, the oscillation and particle shape of raindrops can be analyzed by 2DVD (Thurai and Bringi, 2005; Thurai et al., 2007). Huang et al. (2010, 2015) used 2DVD to analyze the features of irregularly shaped snow. There have been few previous aerodynamic analyses of volcanic ash particles performed using 2DVD, which is able to detect and analyze volcanic ash particles with a range of irregular shapes. Thus, 2DVD offers a unique approach as a new observation strategy.

b. Definition of particle shape type

190 Volcanic ash particles have various shapes that can be detected by 2DVD (Fig. 1). In the case of
 191 raindrops, the DSD is dependent upon the break-up and coalescence processes occurring via up and
 192 downdrafts, since the forces of gravity and buoyancy can easily affect raindrop shapes (Rosenfeld and
 193 Ulbrich, 2003). Solid particles do not readily change shape when falling without the influence of forces
 194 such as collision. It is thus inferred that many particle shapes would be found in the atmosphere, and
 195 that it would be possible to define and classify each particle shape type if we were able to accurately
 196 detect a single particle. Thus, the range of γ for solid particles would be expected to be wide compared
 197 to that of raindrops, and various values of V_T and β would likely be observed. The γ_x of a particle is
 198 defined as the ratio of height to width for the observation direction x , and its representative value is
 199 calculated using the geometric means of the two γ (γ_1, γ_2) detected by cameras 1 and 2, respectively
 200 (Eq. 1):

$$\gamma_{1(2)} = \frac{\text{Height}_{1(2)}}{\text{Width}_{1(2)}}, \quad \gamma = \sqrt{\gamma_1 \gamma_2}$$

Eq. (1).

206 The β is defined as the difference in angle between the rotating symmetric axis and vertical axis.
 207 The counter-clockwise (clockwise) movement of the rotating symmetric axis has a positive (negative)
 208 value and the entire range is 180° (from -90° to 90°) with 0° as the center.

209 It is necessary to consider the true axis ratio (γ_T) to correctly define the particle shape (Fig. 2). The
 210 apparent axis ratio (γ_A) considers the effect of β but the γ_T does not. The 2D coordinates (x, z) of the
 211 particle shape with β are defined as follows:

212

213 $x_A = r \cos(\theta + \beta), \quad z_A = r \sin(\theta + \beta)$

214

215 $x_T = r \cos\theta, \quad z_T = r \sin\theta$

216

217 where subscript A is the coordinate of the original data coordinate considering the β and subscript
218 T is the modified data coordinate. The symbol r refers to the length from the data point to the center
219 and the symbol θ represents the degrees of data coordinates from the positive x axis, which range
220 between 0° and 180° . In this paper, γ stands for γ_A for convenience.

221 An objective criterion for particle shape type was considered since particle shapes can be highly
222 diverse and irregular (e.g., Bagheri and Bonadonna, 2016, Dioguardi et al., 2017; Garboczi et al., 2017,
223 Dioguardi et al., 2018). In the case of irregular particles, the γ can change according to the observation
224 direction; however, any criterion should be able to define the particle shape types strictly and reliably.
225 To solve this problem, particles with a rotating symmetric axis were the main target of the present
226 study. Therefore, we considered oblate spheroid (O), prolate spheroid (P), and sphere (Sp), which all
227 have a rotating symmetric axes. Among these particle types, the major axes of the oblate and prolate
228 spheroids could be horizontally (H) and vertically (V) oriented with respect to the ground, respectively.
229 Thus, the various particle shapes were divided into five types as follows; OH, OV, PH, PV, and Sp.

230 To define these particle shape types, a strict definition of the γ_T is required, which can be calculated
231 from the β . As with the γ , the two β values are automatically calculated by 2DVD. In the case where
232 the β is assumed as 0° , the rotating symmetric axis for OH and PV can be defined, since it is observed
233 for any observation direction parallel to the ground. However, in the case of OV and PH particles, the
234 rotating symmetric axis cannot be defined when the observation direction is parallel. In the case where
235 the β is not 0° , γ_T for all particle shape types would not change when oscillation occurs in a direction
236 orthogonal to the observation direction, but it is difficult to estimate both γ_T and β when particle

237 oscillation appears in a direction parallel to the observation direction. The ability to restore the γ_T and
238 β relative to this observation direction is limited, which is one of the main disadvantages of the 2D
239 observation strategy.

240 Based on these facts, a major β was selected based on the following reasoning: i) a β for the
241 observation direction with lower (higher) γ_T for OH (PV) is selected. ii) in the case of OV (PH), for
242 which the rotating symmetric axis was observed for only one observation direction, β was considered
243 where the value of β had a higher (lower) γ_T than that of the other observation direction. Therefore, β
244 with a lower (higher) γ_T in two observation directions for the case of an oblate (prolate) particle was
245 considered as a meaningful value. The Sp could not have their value of β determined theoretically,
246 because there is the possibility of a rotating symmetric axis in any direction.

247 After removing β , each particle shape was defined using γ_T (Table 1). Note that a 10% bias range
248 was allowed, to take observational error into account. For example, a particle was considered as a
249 sphere when $0.9 < \gamma_T < 1.1$, which is an applied 10% bias range from $\gamma_T = 1$. In addition, the particle
250 types OH and PV (OV and PH) were classified when the value of $|\gamma_1 - \gamma_2|$ was smaller (larger) than
251 $0.1\gamma_T$, to consider particles with only a rotating symmetric axis.

252

253

254 **c. Calculate the terminal velocity for the various particle shape types**

255 The V_T of volcanic ash is required to estimate the R_A ($\text{kg m}^{-2} \text{s}^{-1}$) on the ground where this depends
256 on atmospheric density (ρ_g in g cm^{-3}), T , Re , drag coefficient (C_D), D , shape, and ρ_s . Kunii and
257 Levenspiel (1969) developed a theoretical V_T equation:

258

$$259 \quad V_T = \left(\frac{4(\rho_s - \rho_g)gD}{3\rho_g C_D} \right)^{0.5} \quad (10^0 < Re < 10^4)$$

Eq. (2)

Later, Suzuki (1983) developed a theoretical V_T equation for tephra. Bonadonna et al. (2011) then modified the theoretical V_T equation suggested by Kunii and Levenspiel (1969) with observed ash data, which implied that the result of the theoretical V_T equation could be unsuitable for non-spherical particles. Based on these equations, various C_D equations considering non-spherical particles were subsequently developed. Tran-Cong et al. (2004) developed a new equation for C_D using the function of circularity and Hölzer and Somerfeld (2008) introduced a progressed C_D equation considering two types of sphericity: lengthwise ($\Phi_{||}$) and crosswise (Φ_{\perp}). This equation is as follows:

$$C_D = \frac{8}{Re} \frac{1}{\sqrt{\Phi_{||}}} + \frac{16}{Re} \frac{1}{\sqrt{\Phi}} + \frac{3}{\sqrt{Re}} \frac{1}{\Phi^{3/4}} + 0.42 \times 10^{0.41(-\log(\Phi))^{0.2}} \frac{1}{\Phi_{\perp}}$$

Eq. (3)

The Re is defined as:

$$Re = \frac{\rho_g V_T D}{\mu}$$

Eq. (4)

where μ is the dynamic viscosity ($\text{kg m}^{-1} \text{s}^{-1}$), which we assumed to be 1.983×10^{-5} based on atmospheric conditions at a T of $25 \text{ }^\circ\text{C}$. Three types of sphericity were defined as follows:

$$\Phi = \frac{\pi D^2}{SA}$$

Eq. (5)

where SA is the surface area of the particle (mm²). The lengthwise sphericity is defined as the ratio between the cross-sectional area of the volume-equivalent sphere and the difference between half the surface area and the mean of the projected vertical cross-sectional area (A_V) of the particle (Eq. 6):

$$\Phi_{\parallel} = \frac{\pi D^2}{4(0.5 \times SA - A_V)}$$

Eq. (6)

The crosswise sphericity is the same as the lengthwise sphericity, except for the denominator, which includes the projected horizontal cross-sectional area of the particle (A_H), defined as follows:

$$\Phi_{\perp} = \frac{\pi D^2}{4A_H}$$

Eq. (7)

It is noteworthy that the V_T is required to calculate the Re and V_T , which refers to the final product. To solve this problem, the theoretical V_T (Eq. 2) was used as the input value of Eq. 4 until Eq. 2 converged.

d. Sakurajima volcano

Japan has around 10% (110) of all of the active volcanos in the world. Sakurajima (1,117m, 31.58° N, 130.65° E, Kyushu, Japan) is an active volcanic island formed around 13,000 years ago, and its

305 tephra is approximately 57–66 % SiO_2 Peléan-type. The major eruptions periods of Sakurajima were
306 1471–1476 (Bunmei era), 1779–1782 (An-ei era) and 1914 (Taisho era). The Japan Meteorological
307 Agency (JMA) reported that the eruption frequency of Sakurajima would increase significantly from
308 2009 (Maki et al., 2014) and the accumulated ash fall exceeded 3.5 kg m^{-2} in Kagoshima city in 2012
309 (Maki et al., 2016). The Ministry of Land, Infrastructure, Transport, and Tourism (MLITT) installed
310 an operational X-band radar 10.7 km from the vent, as well as 16 automatic volcanic ash weight
311 measurements, to observe volcanic eruptions in 2011 (Fig. 3).

312

313

314 **e. Data collection and reanalysis**

315 The data were collected by automatic volcanic ash weight measurements performed on the
316 Sakurajima volcano (Tajima et al., 2015; Maki et al., 2014; 2016). The free-fall experiments were
317 divided into two types; one was performed for each phi scale ($\Phi = -\log_2 D$) from $\Phi = 3$ to -4 ($0.125 <$
318 $D < 16 \text{ mm}$), and the other was not considered on a particle size scale. The former data, expressed by
319 A and B (Type 1), were collected at two sites and reanalyzed by size (Fig. 4); the latter data, expressed
320 as C–E (Type 2), were collected at 18 sites (Table 2). Free-fall experiments on collected volcanic ash
321 particles were carried out in the large-scale rainfall simulator of the National Research Institute for
322 Earth Science and Disaster Prevention (NIED) in Tsukuba, Japan. The collected particles were dropped
323 manually by a manager around 17 m from the ground and re-collected by a third-generation 2DVD
324 (Maki et al., 2016). Half a cup of each sample was dropped for 30 s to stimulate dispersion, and the
325 measurement period was 1 min. To avoid wind effects including turbulence, the 2DVD was surrounded
326 by a 27 m^3 windbreaking wall (Fig. 5).

327 The free-fall experiments were conducted at intervals of 1 min over 6 h 30 min, as shown in Fig.
328 6. The number of particles detected by 2DVD was less than 10,000 for 1 min, and the particle size

329 range of **The** former data set (A, B) was proportional to its phi-scale, since small particles may be
330 observed in collisions or break-up from aggregates. Bonadonna et al. (2011) showed that the dominant
331 distribution areas of volcanic ash particles of $\Phi = 1\sim 2$, $1\sim 3$, and $3\sim 9$ corresponded to around 10, 20,
332 and 30 km from the vent of Eyjafjallajökull, Iceland, respectively. Dellino et al. (2012) reported that
333 the dominant volcanic ash particle size is centered at $\Phi = 0\sim 3$ and $1\sim 2$ at 3 and 9 km from the vent
334 of the same volcano, respectively. Based on these studies, the results of the present study represent the
335 range between 2 and 20 km from the vent under natural conditions.

336 Figure 7 shows the distribution of raw data (the number of data: 274,215) for V_T and γ with D .
337 There were various γ values, from 0 to 2, when the volume-equivalent particle $D < 2$ mm, and most of
338 the data were concentrated near 0. The γ values converged around 1 and their distributional range
339 decreased with D . It should be noted that this feature is different from that of raindrops, where smaller
340 raindrops converge around 1 and gradually decrease with D (Thurai and Bringi, 2005). Beard and
341 Chuang (1987) and Andsager et al. (1999) suggested that these raindrop features denote polynomial
342 relationships (Fig. 7a). The median value with a 0.25 mm D interval corresponded well to the center
343 of the data contour. The median line converged around $\gamma = 0.93$ based on the correlation coefficient
344 value (CC). When this was higher than 0.95 for each D interval, the data converged. According to this
345 condition, the range of $2 < D < 5$ mm was satisfied and the mean value was calculated using these data.

346 The V_T had a wider range when $D < 2$ mm but the median line corresponded well to the center of
347 the data (Fig. 7b). The line representing the largest amount of data is higher than the V_T of raindrops
348 suggested by Atlas et al. (1973), and lower than the volcanic ash discussed by Bonadonna et al. (2011).

349 To select a reliable range for particle D , a theoretical terminal velocity equation ($V_{T,Ref}$)
350 corresponding to Eqs. 2–7 was used as the reference. The particle density associated with the eruption
351 of Sakurajima volcano is between 2.43 and 2.59 g cm⁻³ (Oguchi et al., 2009), but the actual particles
352 contain air vacuoles (Van Eaton et al., 2012). This means that the bulk density, including vacuoles, is

353 smaller than the particle density. Therefore, the minimum particle density was considered to be 2.43 g
354 cm^{-3} , and this was used as an input parameter. The atmospheric conditions of T and P were considered
355 using ground observation data from automatic weather stations (AWS), supported by the JMA. The
356 falling height of a particle when $D = 4$ was lower than that under laboratory conditions when it reached
357 90% of V_T (13.9 m) (17 m); therefore, the available data range is considered to be $D \leq 4$ mm, and this
358 would satisfy a steady-state oscillation. The detailed equations used in the present study are shown in
359 Appendix A.

360

361

362 **f. Quality control procedures**

363 The 2DVD was originally developed to detect raindrop hydrometeors. For this reason, additional
364 quality control (QC) checks were deemed necessary to ensure applicability to non-hydrometeors, such
365 as volcanic ash particles. Specifically, we performed the following two QC procedures for accurate
366 analysis of the data:

367 i) Particle $D > 0.25$ mm was selected in consideration of the minimum spatial resolution of 2DVD.

368 ii) If the major axis observed by 2DVD was 10% longer than that of the value calculated directly
369 based on data coordinates, the data were considered erroneous and thus removed. A 10% bias range
370 was considered based on mathematical error, the irregular particle shape, and the limitation of the
371 spatiotemporal resolution of 2DVD.

372 iii) Based on the definition of β (Fig. 2), the perfect condition with respect to ellipsoids is satisfied
373 when $|\beta| = 0^\circ$ (90°) for OH and PV (OV and PH); these values are defined as the center values. However,
374 2DVD calculated that the β for each particle shape type was concentrated around $|\beta| = 0^\circ$ (90°) with
375 respect to horizontally (vertically) oriented particles, which correspond to OH and PH (OV and PV).
376 Furthermore, analysis of particles with an orthogonal center angle from 0° is difficult, since they have

377 two center angles ($\pm 90^\circ$). To address observation errors and enhance the convenience of analysis, all
378 center angles were set to $|\beta| = 0^\circ$ and modified to give the representative canting angle, β_R , using the
379 following equation:

380

$$381 \quad \beta_R = \beta - \beta_0$$

382

383 where β_0 is the orienting angle, defined by the central angle of oscillation. In the case of vertically
384 oriented particles (OV and PV), β_0 could be defined as $\pm 90^\circ$. The sign of β_0 follows that of β . After
385 selecting the particle shape types and applying these QC procedures, 19.31% of the data (62,953)
386 remained (Table 1).

387

388

389 **3. Results**

390

391 **a. Size and shape distribution**

392 The particle size distribution over the entire volcanic ash sample was skewed leftward, and the
393 particle shape distribution differed with particle size (Fig. 8). The particles were predominantly
394 horizontally oriented (75.51%) or vertically oriented (21.60%). Oblate and prolate spheroids made up
395 76.26% and 23.85% of the particles, respectively. Hence, the particles were mainly OH (57.38%) or
396 PH (15.88%) (Table 2). The particles were predominantly $0.25 < D < 0.5$ mm (63.00%) or $0.5 < D <$
397 1 mm (32.80%). Relatively few particles had $D > 1$ mm (4.20%).

398 There was large variation in shape among particles $0.25 < D < 0.5$ mm, but the variation decreased
399 with increasing D . All of the particle shape types had the largest number of particle at $D > 1.0$ mm and
400 the next were shown at $0.5 < D < 1.0$ mm, except for OH. In total, 95.80% of OH particles had $D < 1$
401 mm; at $D < 0.5$ mm, the value was 75.68%, and at $D = 0.5$ – 1 mm, it was 22.36%. In the cases of PH
402 and OV, 93.63% and 93.87% of these particles, respectively, had $D < 1$ mm. Beyond $D > 1$ mm, the
403 differences in the number of particles for each particle shape type were considerably decreased. The
404 PSD of volcanic ash particles depended on the distance from the vent (Beckett et al., 2015; Stevenson
405 et al., 2015). Based on the results, this particle size and shape distribution would represent the area
406 located over 2–20 km from the vent.

407

408

409 **b. Terminal velocity**

410 It is difficult to apply qualitative analysis approaches to V_T since it depends on the scale, magnitude,
411 and duration of an eruption. Furthermore, it is impossible to develop a model of V_T without knowledge

412 of the aerodynamic properties of the volcanic ash particles, since V_T is a function of the density, shapes,
413 and oscillations of particles. In this study, we analyzed the basic features of volcanic ash particles in a
414 laboratory experiment to obtain reliable results.

415 The observed values of V_T were well classified by particle shape and were generally higher than
416 those of raindrops, as previously suggested by Atlas et al. (1973) (Fig. 9). The values were estimated
417 using a multiple regression analysis, with V_T defined for each particle type, and with additional QC
418 measures applied. If we consider a single V_T QC measure for the entire particle types, a number of
419 available data might be removed, since V_T critically depends on particle shape. Therefore, we applied
420 a $\pm 60\%$ V_T QC threshold (Jaffrain et al., 2011).

421 The highest values of V_T were recorded in the order of prolate, sphere, and oblate. Vertically
422 oriented particles had higher V_T values than horizontal ones. For raindrops with a single particle shape
423 type, V_T changes with atmospheric conditions, especially vertical air motion (Kim and Lee, 2016).
424 However, the V_T values for volcanic ash particles change independent of external conditions, since
425 they have various particle shapes. The V_T for every particle type was considered to be in a power-law
426 form, except for OH. OH particles followed the regression line relatively closely, and showed the
427 highest CC and root mean square error (RMSE) values, of 0.94 and 0.46 m s^{-1} , respectively.
428 Horizontally orientated particles had relatively higher correlations (OH: 0.94, PH: 0.87) compared to
429 those with a vertical orientation (OV: 0.75, PV: 0.71). All of these data are summarized in Table 3.

430 To verify the reliability of the particle data obtained by 2DVD, which was originally developed to
431 detect liquid raindrops, particle density, C_D , and Re , as well as theoretical V_T values according to these
432 parameters, were analyzed. To calculate the parameters of interest, including the surface area and
433 cross-sectional area of irregular particles, we applied the irregular particle volume estimation equations
434 of Huang et al. (2010).

435

436

437 **c. Aerodynamic properties**

438 Particle densities were estimated using the $V_{T,Ref}$, converged to 2.37 g cm^{-3} when $D < 1.5 \text{ mm}$ (Fig.
439 10a). This value corresponds well to that of the minimum particle density (2.44 g cm^{-3}) reported by
440 Oguchi et al. (2009). The slight difference is likely due to observation errors and the presence of
441 vesicles (e.g., Seligman et al., 2016). The median value of particle density is changed to $0.5 < D < 1.5$
442 mm, and this range corresponds to that of V_T . Horizontally oriented particles (OH and PH) have
443 relatively smaller particle density and vertically oriented particles (OV and PV) have higher density
444 (Fig. 10b); spheres have particle densities that accord best with D . The median particle density values
445 for all particle shapes converged when $D < 2 \text{ mm}$, and the converged particle density values ranged
446 from 2.35 to 2.50 g cm^{-3} .

447 The Re and C_D for the all particle shapes ranged from 10 to 4,000 and 0.6 to 20, respectively (Fig.
448 11a). Higher values of C_D were observed when $Re < 70$; above this threshold, C_D dramatically
449 decreased. These results were derived according to the number of OH particles, which mainly had D
450 $< 0.5 \text{ mm}$, and had higher C_D and lower Re . Particle shape was divided into two types: OH and others
451 (Fig. 11b). OH particles had higher C_D compared to the other particle shapes, which in turn showed
452 few differences among themselves. The OH particles experience strong drag forces under the same
453 flow conditions, leading to lower V_T . The differences between OH and other particles diminished with
454 $Re < 1,000$. The other particle shapes had relatively higher C_D in the range $10 < Re < 3,000$. Rong et
455 al. (2015) analyzed the relationship between Re and C_D for oblate and prolate particles, and showed
456 that OH particles had higher C_D compared to the reference line (Clift and Gauven, 1971) in the range
457 $0 < Re < 400$. Each relationship is summarized in Fig. 11.

458

459

460 **d. Axis ratio**

461 The γ of a particle affects the backscattering power of electromagnetic waves, and it is necessary
462 to calculate the horizontal reflectivity (Z in dBZ), differential reflectivity (Z_{DR} in dB), and specific
463 differential polarization phase shift (K_{DP} in $^{\circ} \text{ km}^{-1}$). Note that previous studies have analyzed the γ
464 distribution for raindrops and snow, including hail; however few studies have been reported on
465 volcanic ash.

466 Smaller raindrop particles exhibit higher values of σ . However, most of the particles are
467 concentrated around $\gamma = 1$ when D is close to 0 mm, and the γ for raindrops was shown to be inversely
468 proportional to D (e.g., Pruppacher and Beard, 1970; Beard and Chuang, 1987; Goddard et al. 1994b;
469 Andsager et al. 1999; Thurai and Bringi, 2005). Snowflakes showed linear relationships of γ and D
470 that were different from those of raindrops. Brandes et al. (2007) suggested that an equation for γ where
471 (D) = $0.01714D + 0.8467$. Hail had a similar γ to snowflakes, of 0.77 (Matson and Huggins, 1980)
472 versus $0.75 < \gamma < 0.79$ (Jewell and Brimelow, 2009), respectively.

473 Figure 12 shows the quartiles and median values of γ for the entire particle shapes, and for each
474 individual particle shape type. The γ had a higher standard deviation (σ_{γ}) of > 0.25 when $D < 0.75$ mm,
475 which decreased and converged to $\sigma_{\gamma} = 0.15$ when $D > 1$ mm (Fig. 12a). The γ in the lower σ_{γ} range
476 converged to $\gamma = 0.94$ and could be expressed as follows:

477

$$478 \quad \gamma(D) = 0.94 - 0.25\exp(-1.90 D)$$

479 Eq. (8)

480 The particles are more easily classified by shape than by V_T (Fig. 12b). Every particle shape type
481 was independent of D , except for OH. The relationships of the particle shape types are expressed as
482 follows (Eqs. 9–12):

483

484
$$\gamma(D)_{OH} = 0.37 \tanh(1.84D - 1.88) + 0.38$$

485 Eq. (9)

486
$$\gamma_{OV} = 1.15$$

487 Eq. (10)

488
$$\gamma_{PH} = 0.88$$

489 Eq. (11)

490
$$\gamma_{PV} = 1.24$$

491 Eq. (12)

492

493 The parameter $\gamma(D)_{OH}$ was calculated using the hyperbolic tangent (tanh) for the following reasons:

494 i) its range of values was wider than those of other particle types; ii) the data distribution changed
495 continuously with D; and iii) it was present in a higher proportion (30.44%) compared to the other
496 parameters. We found that variations in γ decreased with D and the proportion of Sp shapes increased
497 when $D > 2$ mm. The particle types OV and PH showed a wide distribution over $1 < \gamma < 1.5$ and $0.4 <$
498 $\gamma < 1.0$, respectively, when $D < 2$ mm, but the variability in median values was relatively low. The
499 relationships of γ for each particle shape type could be expressed by constant values at $\gamma = 0.75$ (OH),
500 0.88 (PH), 1.15 (OV), and 1.24 (PV), respectively, and these differences are around $\gamma = 0.12$. Maki et
501 al. (2014) analyzed dual-pol radar variables and found that Z_{DR} increased with time; the dominant
502 values at 10 and 18 min after the eruption were close to 1 and 2 dB, respectively. With respect to
503 hydrometeors, the dominant γ of volcanic ash can be inferred from the relationship between γ and Z_{DR} ,
504 as suggested by Herzegh and Jameson (1992). The values correspond to the γ of volcanic ash when the
505 median γ of rain and ice is 0.85 and 0.73, respectively; this assumption is based on the $|K|^2$ for
506 volcanic ash being between that of ice and liquid water. These representative values correspond to D

507 of 1 and 0.15 mm, respectively, based on Eq. 9. It is implied that the amount of particles was largest
508 when $D < 1$ mm (95.8%) and the γ corresponded well to the observed Z_{DR} .

509

510

511 **e. Canting angle**

512 Statistical analysis of β is required to understand the aerodynamic properties of volcanic ash
513 particles, and the input parameters of T-matrix scattering simulations, to verify the observed radar
514 variables. A histogram was used to analyze the distribution and dominant characteristics of β_R . The
515 interval of the histogram was set to 4° .

516 More than 95% of β values for each particle shape type were concentrated in the range $|\beta_R| \leq 30^\circ$,
517 with 0° as the center (Fig. 13). The particles were symmetrically distributed around 0° and more than
518 50% were concentrated in the range $|\beta_R| < 4^\circ$. The horizontally oriented β distribution was relatively
519 narrow ($|\beta_R| < 20^\circ$) and exhibited a unimodal distribution. It is noteworthy that 90% of OH particles
520 were concentrated in the range $|\beta_R| < 4^\circ$. The vertically oriented β distribution was relatively broader
521 ($|\beta_R| < 30^\circ$) and followed a bimodal distribution. PV exhibited a bimodal form, but this was not
522 symmetrical about 0° . For spheres, the β distribution was narrower, similar to horizontally oriented
523 particles, and its had a bimodal distribution, similar to vertically oriented particles, indicating that the
524 independent features of both orientations were combined.

525 The values of $|\overline{\beta_R}|$ and $|\sigma_\beta|$ for OH and PV (OV, PH) were 0° and 3.5° (0.4° , 13.1°), and 1.3° and
526 12.7° (0.2° , 10.9°), respectively. OV (OH) had the highest (lowest) value of $|\sigma_\beta|$. Thus, $|\overline{\beta_R}|$ and $|\sigma_\beta|$ for
527 the all volcanic ash particles were quite similar (0.3° and 10.1° , respectively) to those of raindrops and
528 the value of $|\sigma_\beta|$ for Sp (11.5°) was higher than that of the entire data set (10.1°).

529 In the case OH, the $|\sigma_\beta|$ value was about half that for raindrops while the values for OV, PH, and
530 PV particles were all similar to that of raindrops. This validates the assumption of Marzano et al. (2012)

531 under stable conditions ($|\sigma_\beta| = 10^\circ$). Therefore, we assumed that the tumbling phenomenon ($|\sigma_\beta| > 30^\circ$)
532 of the particles under calm atmospheric conditions was likely to be minor.

533 To analyze the correlation between particle D and β , quartiles for each particle D interval were
534 calculated (Fig. 14). The particles were concentrated at $|\beta_R| < 30^\circ$ regardless of D, and median values
535 were stable when $D < 1$ mm for the entire particle shape types; however, fluctuation increased with D
536 (Fig. 14a). The $|\sigma_\beta|$ values gradually increased from 10° to 13° when $0.3 < D < 1.3$ mm and variability
537 was greatest around the center (13°). This increase in $|\sigma_\beta|$ would not be expected in the case of a
538 relatively small number of particles (Fig. 8a), since their standard deviation is largely maintained at
539 about 13° regardless of dataset size.

540 Variability in the median values for individual particles was more apparent. The values converged
541 around 0° but fluctuation increased with greater D from the zero line. The median $|\beta_R|$ values
542 exceeded 3° , 5° , 10° , and 15° when $D > 1$ mm, $1 < D < 2$ mm, $2 < D < 3$ mm, and $D > 3$ mm,
543 respectively (Fig. 14b).

544

545

546 **e. Verification**

547 To verify the basic parameters of volcanic ash estimated in the present study, the $V_{T, \text{Ref}}$ (Eq. 2) was
548 compared to the observed values (V_T) obtained in the free-fall experiments (Fig. 15). The particle
549 density (Fig. 10) and γ (Fig. 12) estimated in the present study were applied to the C_D (Eq. 3) and the
550 sphericity (Eqs. 5–7) relationships; these parameters were used as input values for $V_{T, \text{Ref}}$. The
551 atmospheric conditions were the same as in Fig. 5.

552 There was wide range of V_T over $0.25 < D < 1.2$ mm for the entire particle shape types, and V_T
553 decreased when $D > 1.2$ mm. The V_T in the present study corresponds to that obtained by Miwa et al
554 (2015), who analyzed Parsivel data using the same laboratory experiments (Fig. 5); correspondence

555 was also good with other previous results (Beckett et al., 2015). A polynomial regression analysis was
556 used to explore the nonlinear relationship between V_T and D (Table 3). The inflection point of V_T at
557 $0.7 < D < 1.3$ mm results from an increase in the number of OH when $D < 1$ mm (Fig. 8).

558 The $V_{T,Ref}$ was lower (higher) than the observed value when $D < 1.2$ mm ($D > 2.4$ mm). One of the
559 reasons why a difference between the theoretical and observed values appeared when $D > 2.4$ mm was
560 that the upper limit of D when 90% of the terminal velocity was reached was set at 4 mm for a height
561 of 17 m (Fig. A1). This is nonetheless meaningful because the values of $V_{T,Ref}$ calculated based on the
562 basic parameters in this study were within $\pm 5\%$ of the observed value.

563

564 **4. Summary and Conclusions**

565 To inform the development of QAE methods, basic parameters (V_T , γ and β) of volcanic ash
566 particles were analyzed using 2DVD. Data were collected according to 18 automatic volcanic ash
567 weight measurements performed on Sakurajima volcano, Japan (31.58° N, 130.65° E). To identify the
568 aerodynamic properties of the volcanic ash particles in the samples, a free-fall experiment using 2DVD
569 was conducted with the large-scale rainfall simulator of the NIED, and 274,215 raw data points were
570 obtained.

571 Radar variables are highly dependent on the $|K|^2$, size, and shape of particles. Particle types with
572 rotating symmetric axes were selected, because volcanic ash particles have a wide variety of irregular
573 shapes. Their orientation was also considered, and horizontally (OH) and vertically (OV) oriented
574 oblate spheroids were therefore studied, as well as horizontally (PH) and vertically (PV) oriented
575 prolate spheroids, and spheres (Sp).

576 The particle shape types were defined according to their γ_T , with a 10% bias range; the β among
577 two values was selected to be representative when the value of γ_T was smaller (larger) than that of the
578 other observation direction in the case of the oblate (prolate) particle.

579 The dominant particle shape comprised horizontally and vertical oriented particles, present in
580 proportions of 75.51% and 21.60%, respectively. Regarding particle shape, oblate (prolate) spheroids
581 comprised 76.26% (23.85%) of all particles in the samples. The most common particle shape type was
582 OH, accounting for 59% of all particles when $D < 1$ mm, and 69% when $D < 0.5$ mm. Overall, 95.80%,
583 93.87%, and 93.63% of the OH, OV, and PH particles had $D < 1$ mm, respectively.

584 The V_T of the particles were in the order of prolate, sphere, and oblate, and vertical oriented
585 particles had higher V_T than horizontal orientated particles. These results are consistent with the V_T ,
586 $V_{T, Ref}$, which suggests that 2DVD is a reliable for observing volcanic ash particles and the tumbling
587 phenomenon under stable weather conditions. A noticeable increase in V_T for OH in the range $0.5 < D$

588 < 1 mm occurred through an increase in γ and a decrease in the number of OH when $D \sim 0.7$ mm; this
589 was not observed for other particle types.

590 The estimated ρ_s converged to 2.37 g cm^{-3} when $D > 1.5$ mm, and the median value changed over
591 the range $0.5 < D < 1.5$ mm. This is consistent with the particle density value for the Sakurajima
592 volcano reported by Oguchi et al. (2009). The distributions of C_D and Re were divided into two particle
593 type categories, OH and other, and C_D increased dramatically when $Re < 70$. These results were derived
594 from the particle concentration of OH, which was highest when $D < 0.5$ mm; at this threshold, C_D was
595 higher and Re was lower. The range of V_T over $0.7 < D < 1.3$ mm was informed by both γ and particle
596 density.

597 The σ_γ decreased when $D > 0.75$ mm, to 0.15, and converged to $\gamma = 0.94$. This is in contrast to the
598 results obtained for raindrops, but is highly consistent with results for actual eruptions. Most measured
599 values of Z_{DR} for Sakurajima volcanic ash clouds on August 18, 2013 were in the range 1–2 dB,
600 corresponding to a horizontally oriented oblate spheroid.

601 The $|\sigma_\beta|$ of OV particles with $|\beta_0| = 90^\circ$ was largest (13.1°) among all particle types. OH particles
602 had the lowest $|\sigma_\beta|$ at 3.5° , and this value was also smaller than that of raindrops (7° – 10°). Based on the
603 $|\sigma_\beta|$ results, the tumbling phenomenon would not be dominant under calm atmospheric conditions. The
604 quartiles were stable when $D < 1$ mm for the entire particle shape types, but increased with D . The
605 value of σ_β was higher when $D < 1.3$ mm and started to converge around 13° due to a decrease in the
606 number of OH particles.

607 The estimated γ and ρ_s were applied to the $V_{T,Ref}$ for validation. The observed V_T was within $\pm 5\%$
608 of the theoretical value when $D \leq 4$ mm, implying that the estimates of the basic parameters were
609 reasonable for the particle range considered.

610 These results could inform the development of new approaches for detecting non-hydrometeors
611 using weather radar, thus facilitating observations of ash clouds and preventing the damage to human
612 life and property caused by large amounts of ash fall. In future work we will analyze the eruption of
613 Sakurajima volcano that occurred on August 18, 2013 with a dual-pol radar and 2DVD, using basic
614 parameters of volcanic ash particles obtained from laboratory experiments. Differences in
615 characteristics between the experimental, and actual basic parameters, and PSD, will be investigated.

616

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622 their large-rainfall simulator.

623

624 **Appendix A**

625

626 The theoretical fall velocity and falling distance with time are calculated as follows;

627

628

$$F = ma = F_g - F_D$$

629

Eq. (A1)

630

$$F_g = mg$$

631

Eq. (A2)

632

$$F_D = \frac{1}{2} \rho_g V^2 C_D A$$

633

Eq. (A3)

634

635 where F_g is universal gravitation, F_D is drag force, m is mass, and a is the free-fall acceleration.

636 In Eq. A2, g is the acceleration due to gravity, considered to be 9.81 m s^{-2} , and V and C_D in Eq. A3

637 correspond to the values in Eq. 2–7. The symbols μ and ρ_g are the dynamic viscosity and density of

638 the atmosphere and were assumed as $1.837 \times 10^{-5} \text{ kg m}^{-1} \text{ s}^{-1}$ and $1.194 \times 10^{-5} \text{ g cm}^{-3}$, respectively.

639 The results were based on conditions at an atmospheric T as $25 \text{ }^\circ\text{C}$.

640 To ensure accuracy, we considered the surface roughness effect of a volcanic ash particle (1.07^{-1})

641 on the fall velocity, as suggested by Bagheri and Bonadonna (2016), and the results for $D = 4 \text{ mm}$

642 are shown in Fig. A1.

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644

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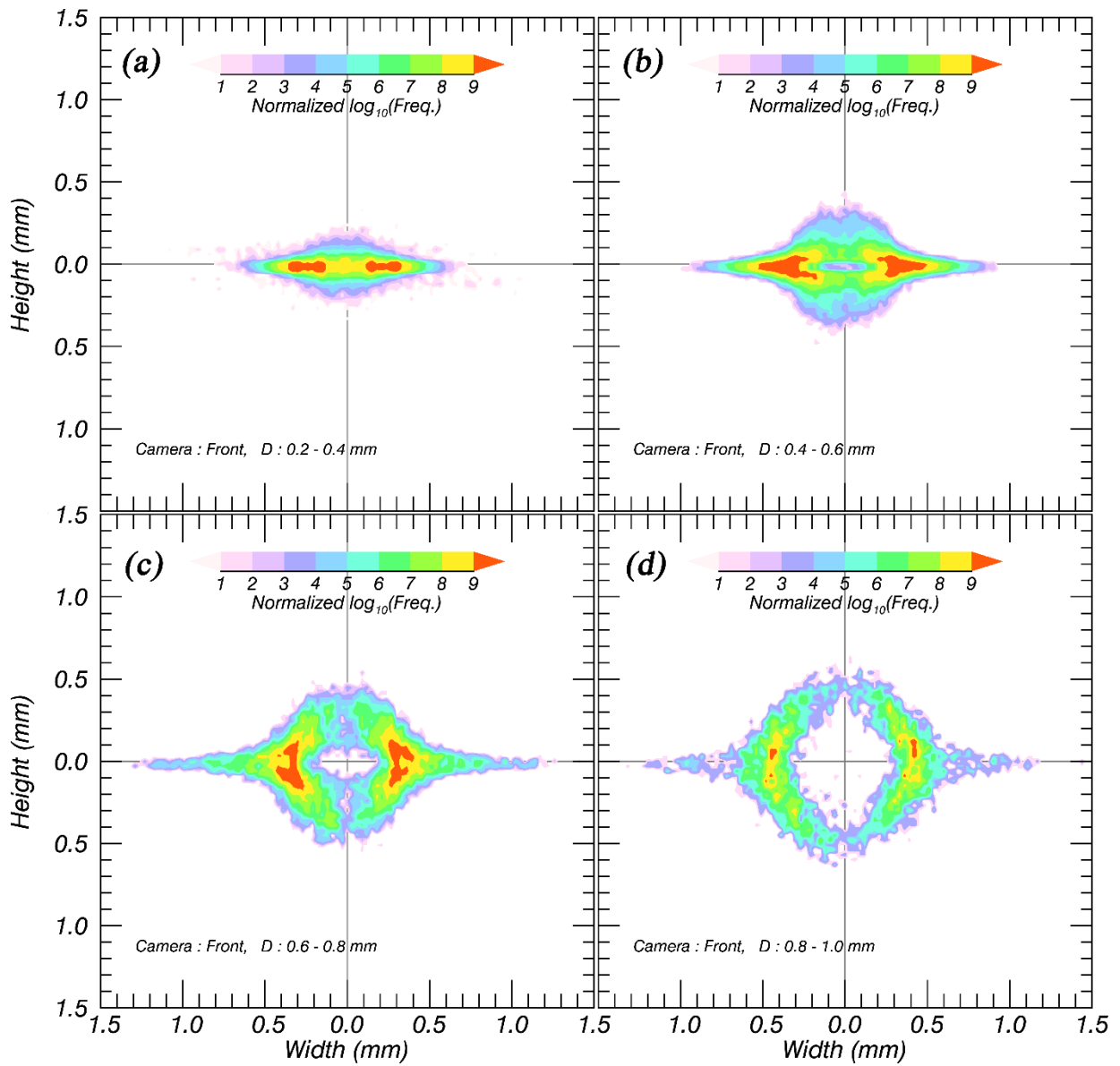
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Figures

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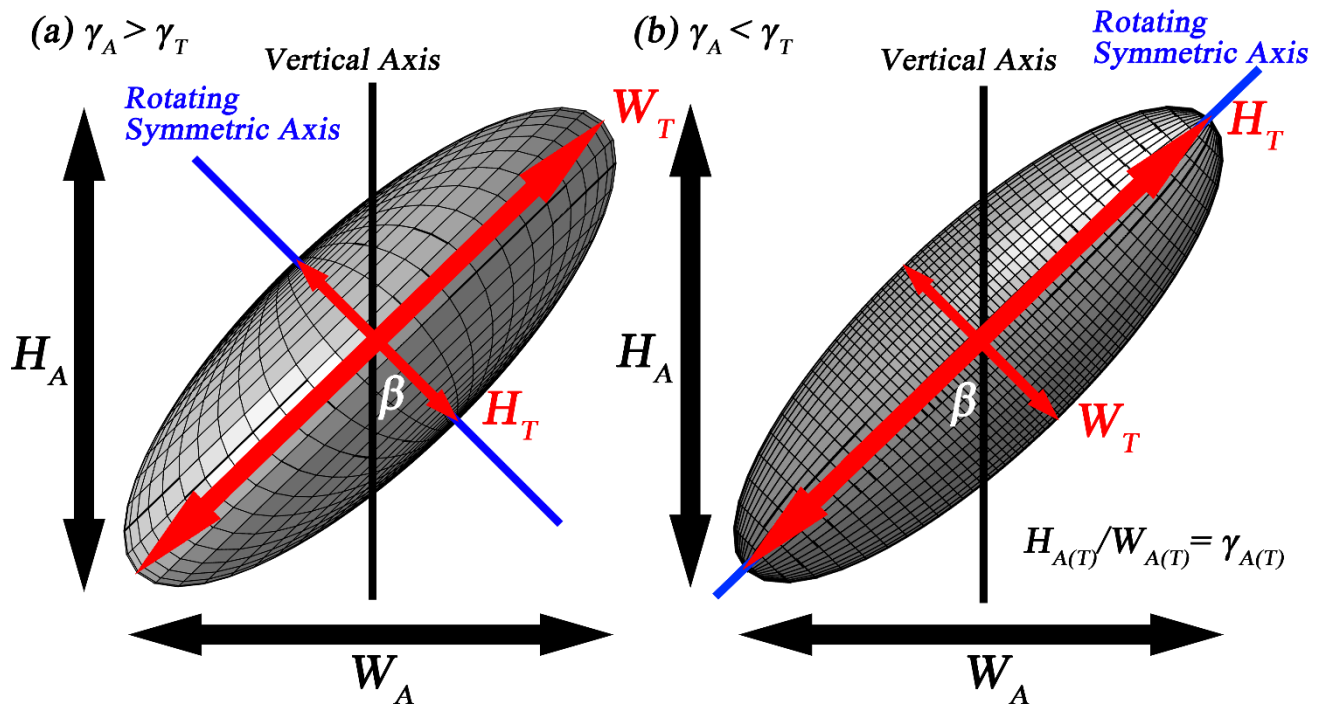
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864 Figure 1. Accumulated contoured images of volcanic ash particles with D measured by a two-
865 dimensional video disdrometer (2DVD).

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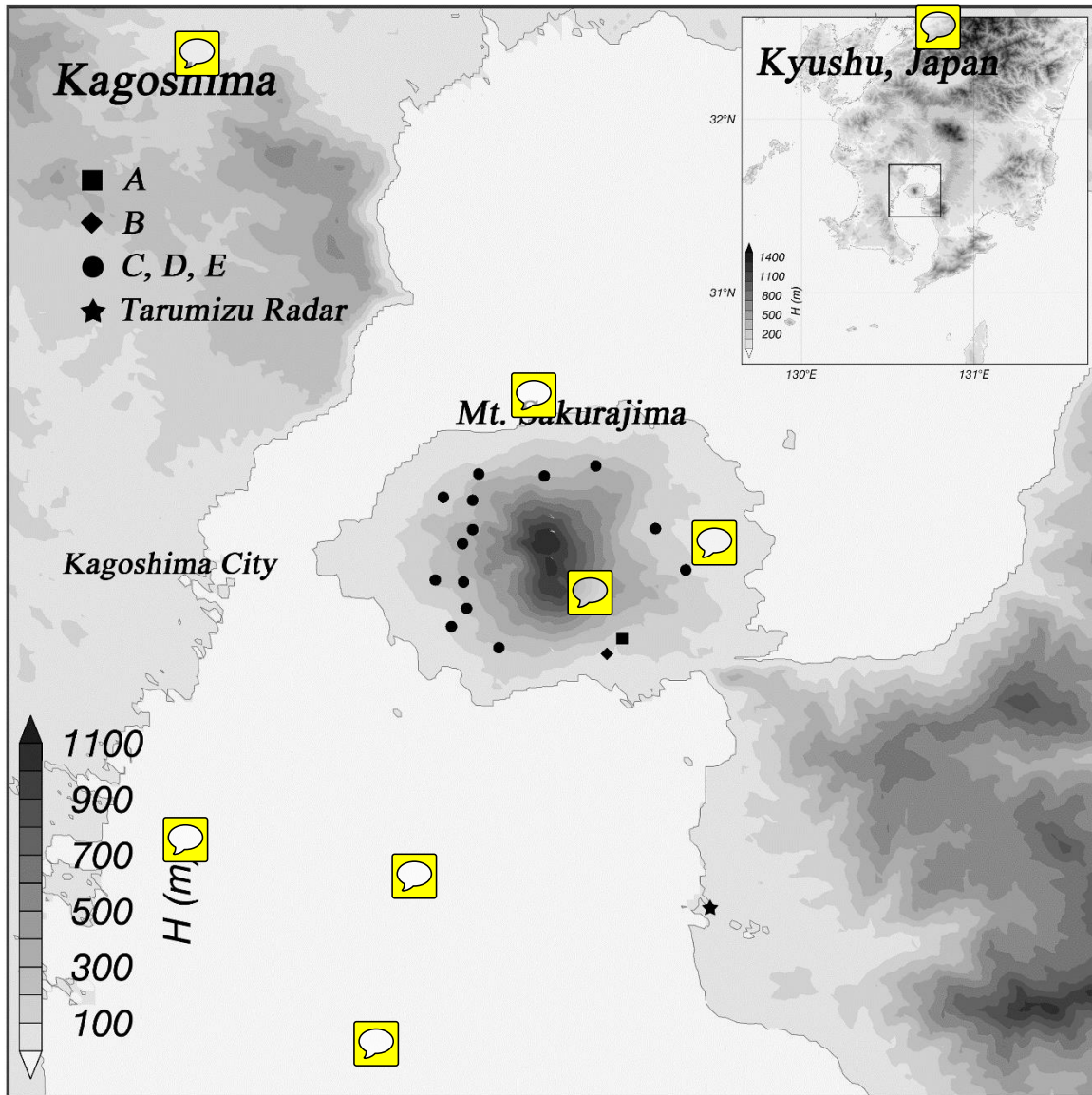
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870 Figure 2. Conceptual model of an (a) oblate and (b) prolate spheroid with the same canting angle
 871 (β). $W_{A(T)}$ and $H_{A(T)}$ are the apparent (true) width and height of the particle, respectively. $\gamma_{A(T)}$ is the
 872 apparent (true) axis ratio.

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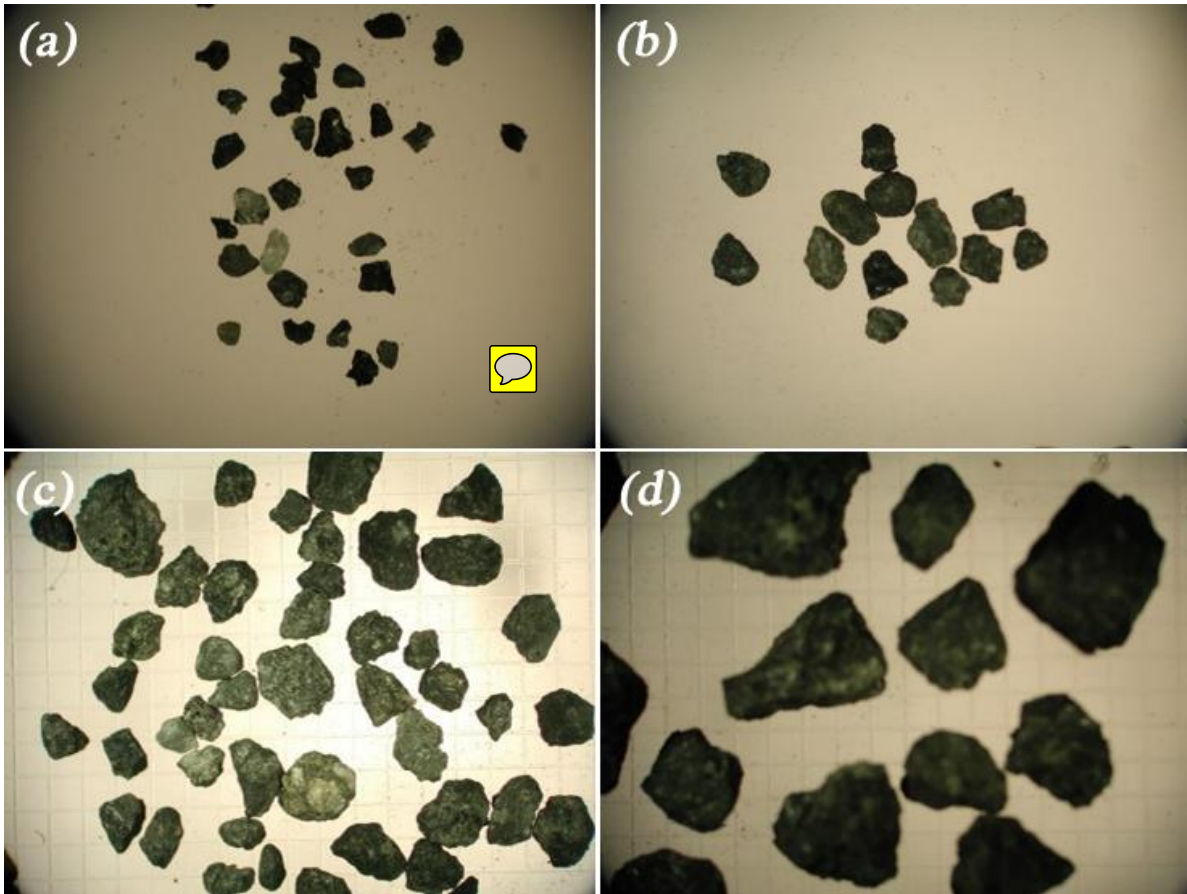
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Figure 3. The locations of tephrometers and Showa crater on Sakurajima volcano, Japan. Black symbols indicate the locations of tephrometers and the star, square, and circle symbols correspond to datasets A, B and C–E, respectively. The white circle symbol represents the location of Showa crater.



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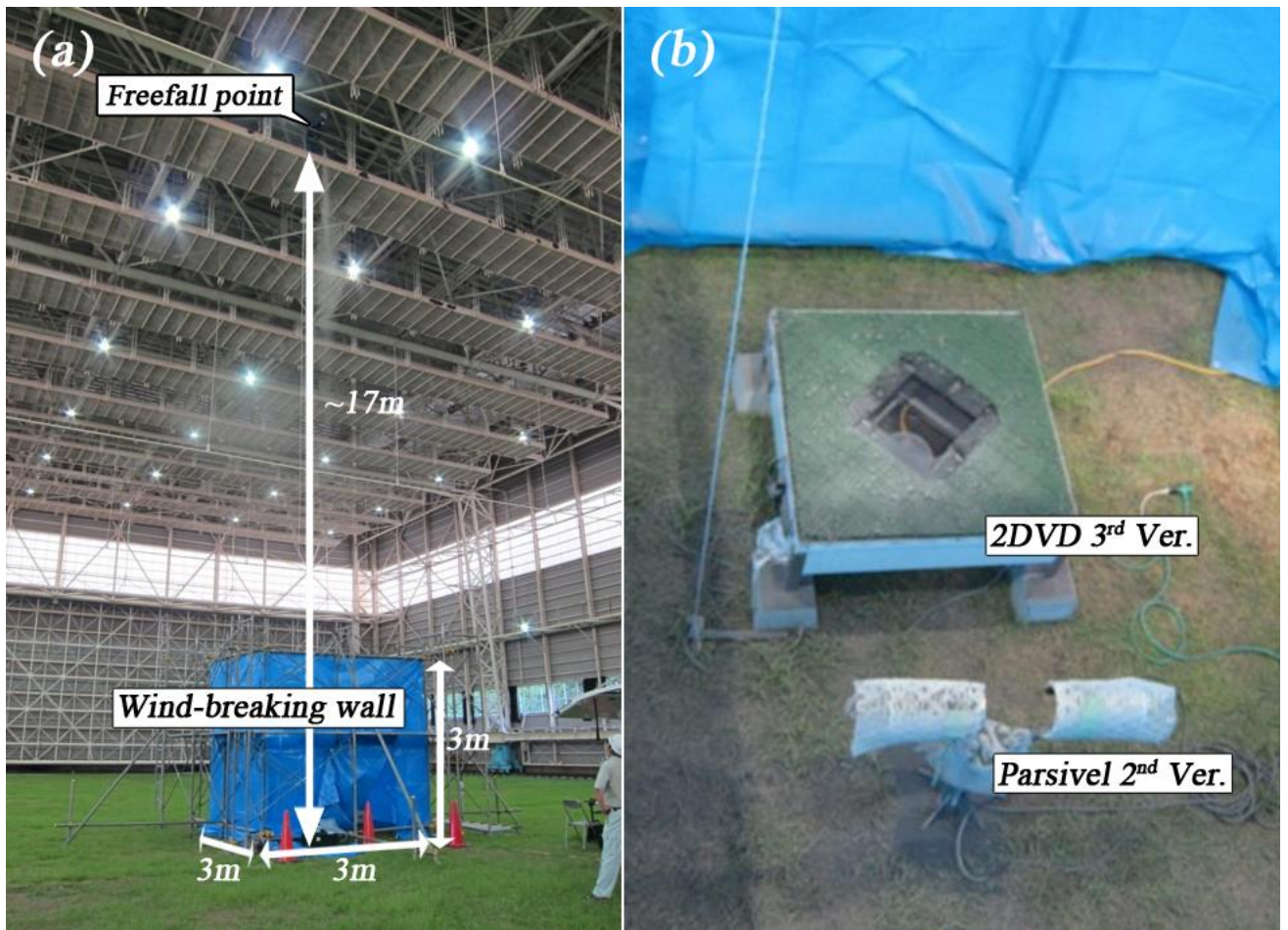
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Figure 4. Real images of volcanic ash particles used in the present study. The particles were classified as (a) $0.125 \text{ mm} < D \leq 0.25 \text{ mm}$, (b) $0.25 \text{ mm} < D \leq 1 \text{ mm}$, (c) $1 \text{ mm} < D < 2 \text{ mm}$, and (d) $2 < D \leq 4 \text{ mm}$.



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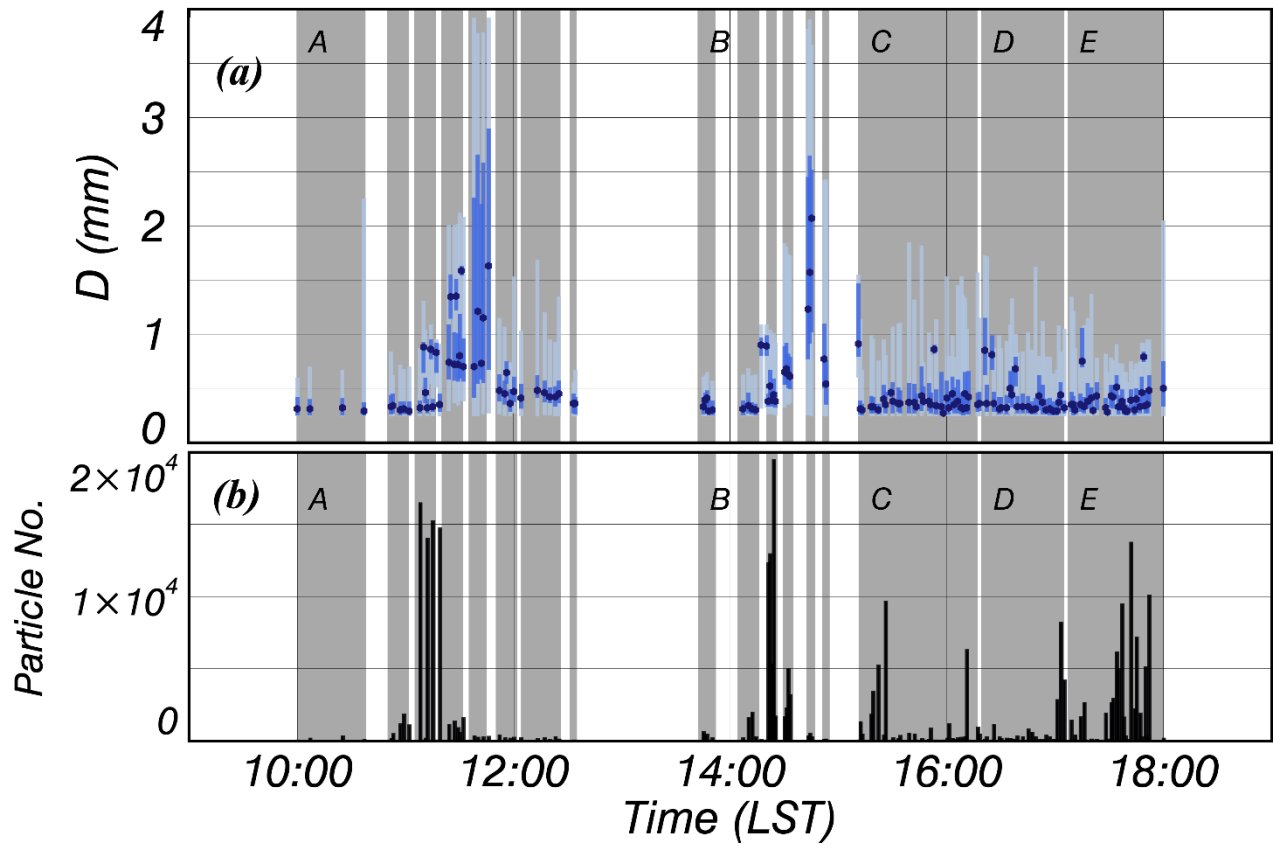
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Figure 5. Free-fall experiment conditions of volcanic ash particles on the (a) outside and (b) inside of the wind-breaking wall covering the disdrometers in the large-scale rainfall simulator of the National Research Institute for Earth Science and Disaster Prevention (NIED).



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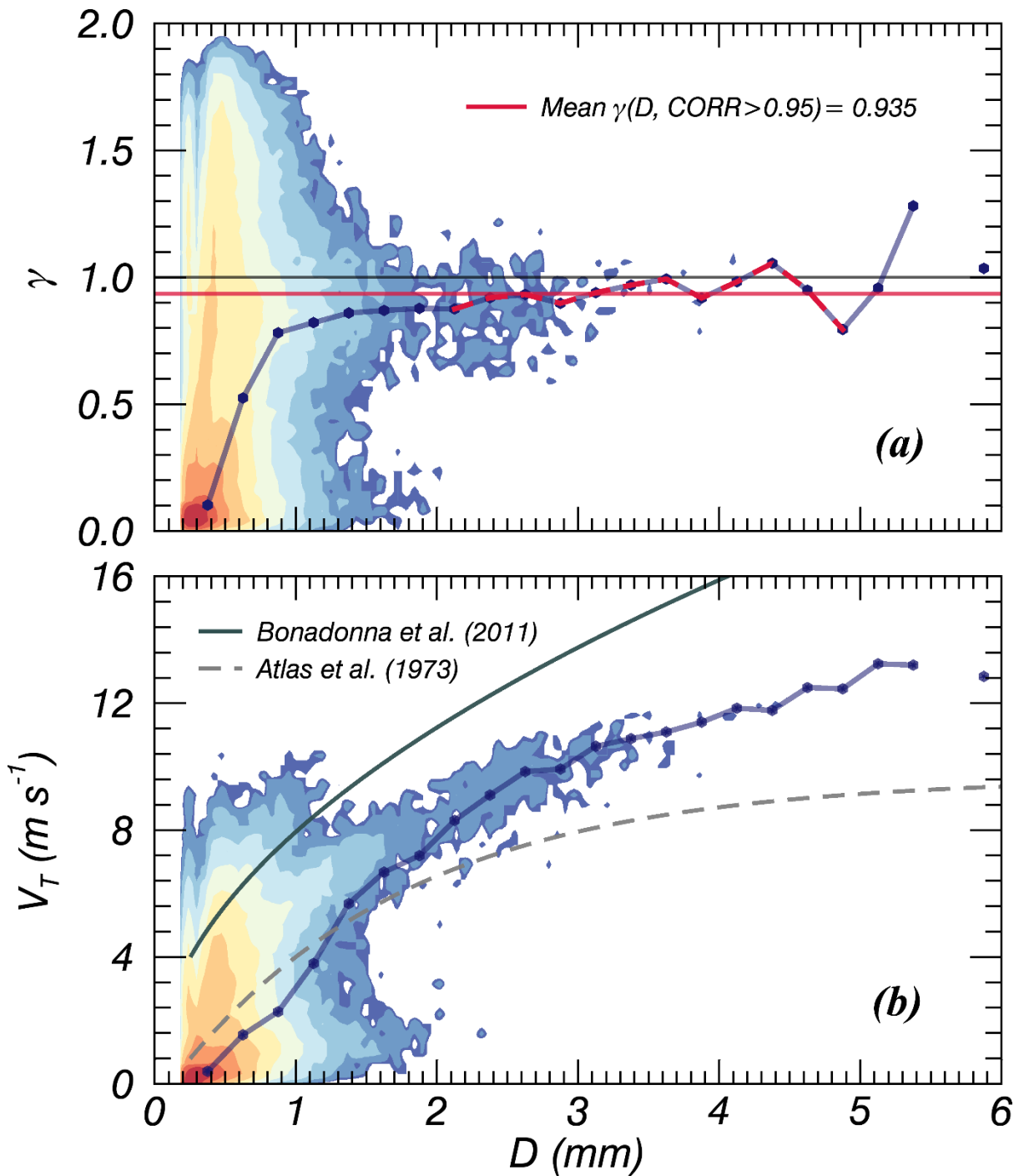
898 Figure 6. The 1-min interval time series of D and the number of particles in the free-fall

899 experiment conducted at the NIED.

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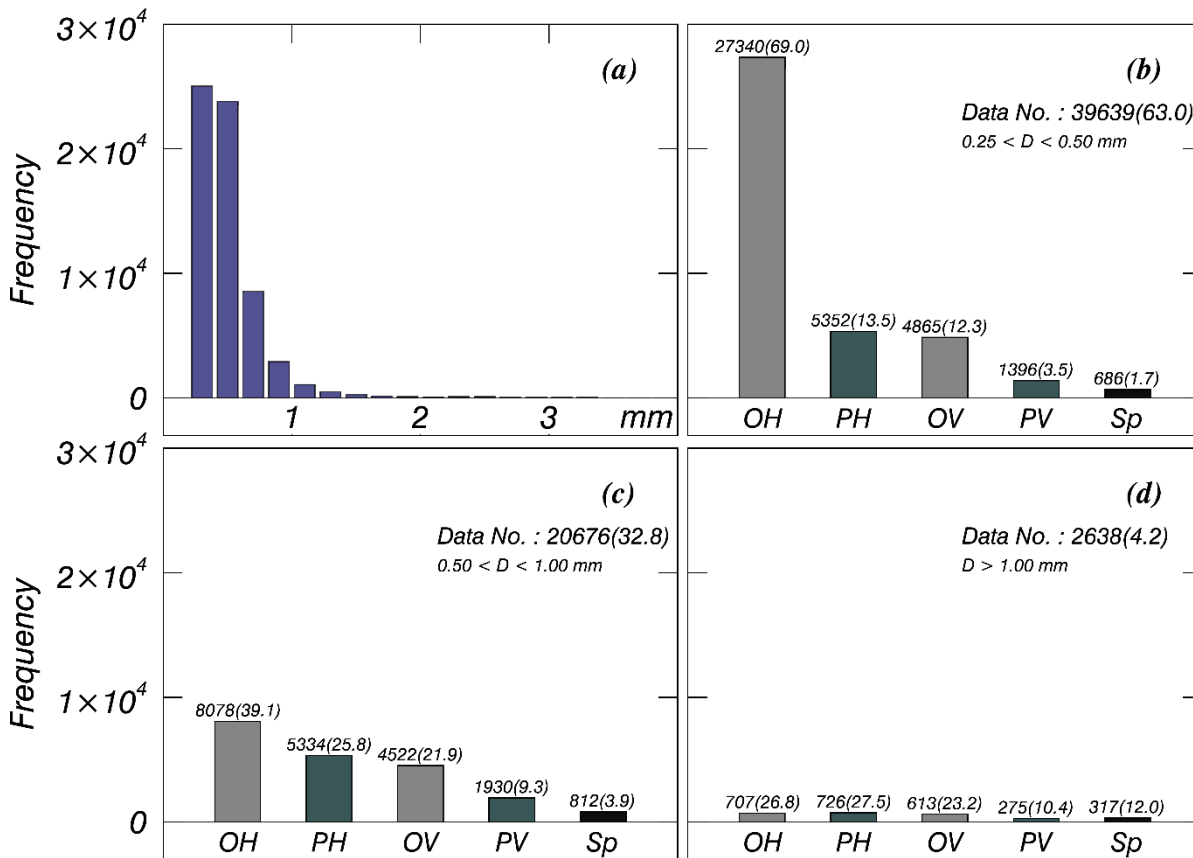
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904 Figure 7. Contour image of volcanic ash particles for (a) the axis ratio (γ) and (b) terminal velocity
 905 (V_T) with respect to the raw data. Grey solid and broken lines are the relationships between volcanic
 906 ash particles and raindrops suggested by Bonadonna et al. (2011) and Atlas et al. (1971), respectively.
 907 The red solid line is the averaged γ satisfying the condition that the correlation coefficient exceeds
 908 0.95.

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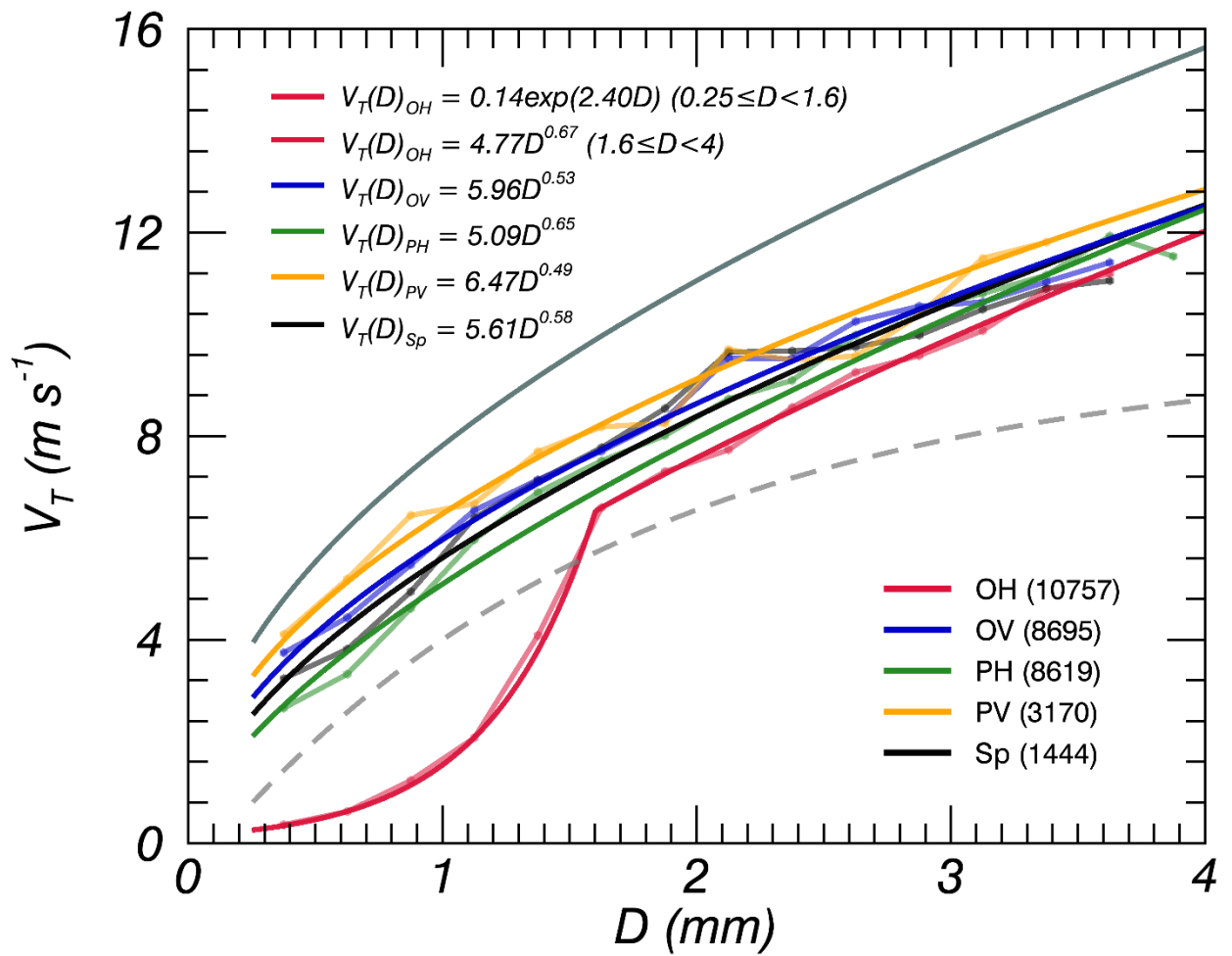
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911 Figure 8. Histograms of volcanic ash particles for (a) all particle types and (b–d) each particle
 912 shape type of the phi scale. The grey- and dark grey-shaded (patterned) bars indicate horizontal oblate
 913 (OH) (vertical oblate [OV]) and horizontal prolate (PH) (vertical prolate [PV]), respectively. The black
 914 bar corresponds to spherical (Sp) particles. The number on the top of each bar plot is the number of
 915 data points and that in parenthesis is the percentage for each phi scale.

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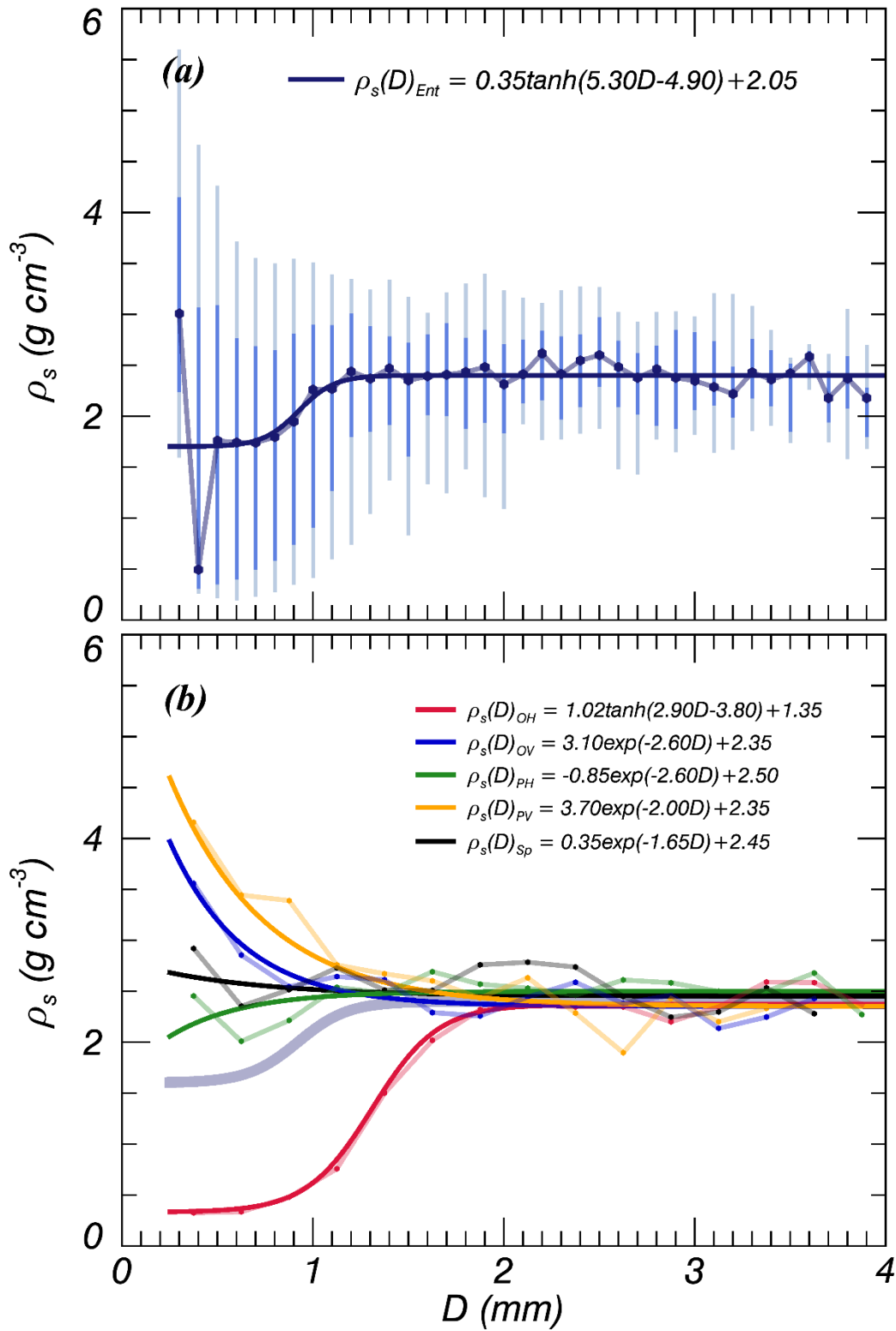
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920 Figure 9. Distribution of median V_T values after applying a 60% V_T quality control (QC) threshold
 921 for each particle shape type. The grey solid and broken lines show the relationships of volcanic ash
 922 particles and raindrops suggested by Bonadonna et al. (2011) and Atlas et al. (1971), respectively.

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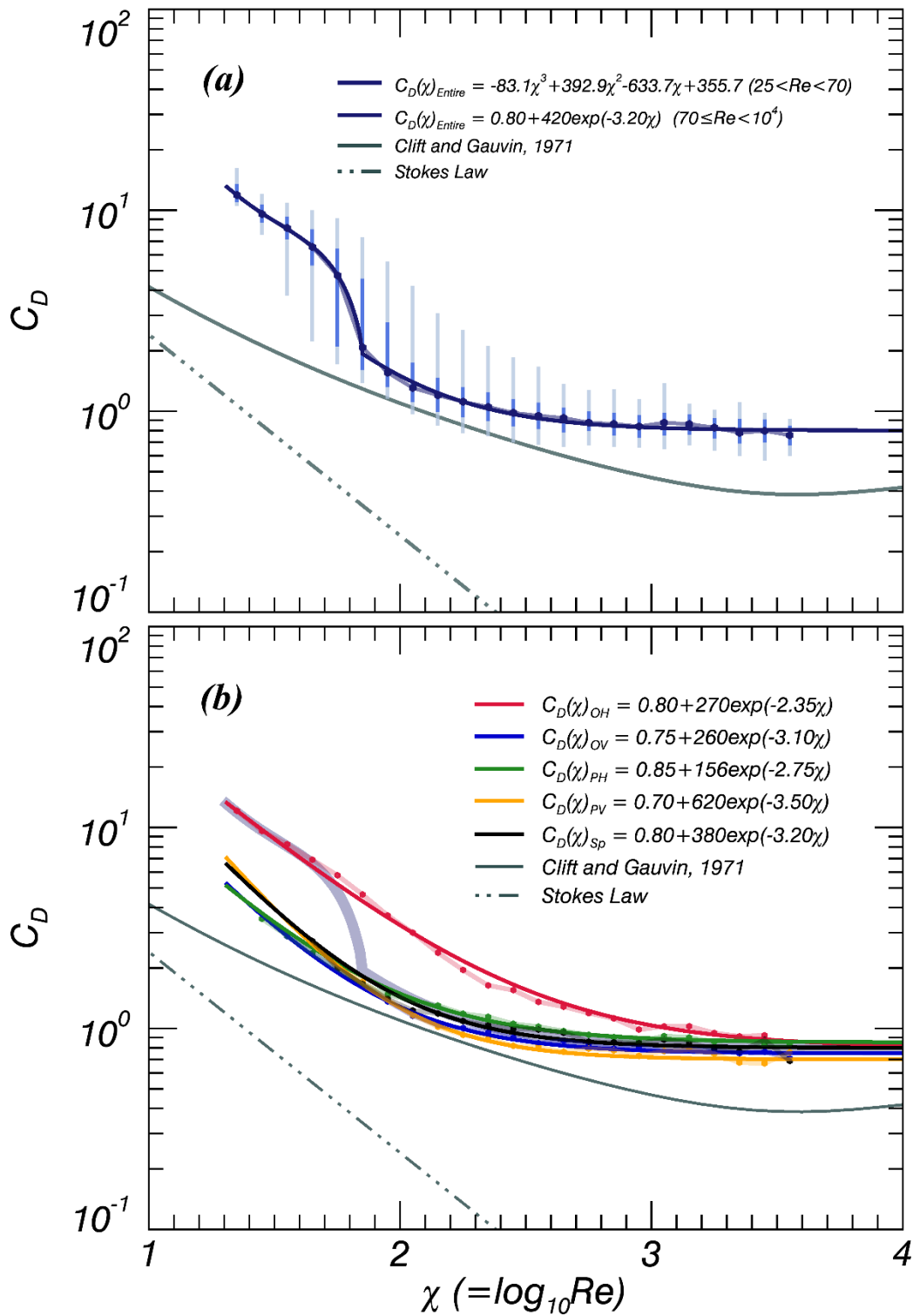


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927 Figure 10. Distribution of (a) quartile and (b) median particle density (ρ_s) values after applying the

928 60% V_T QC threshold for all, and each individual, particle shape type, respectively.

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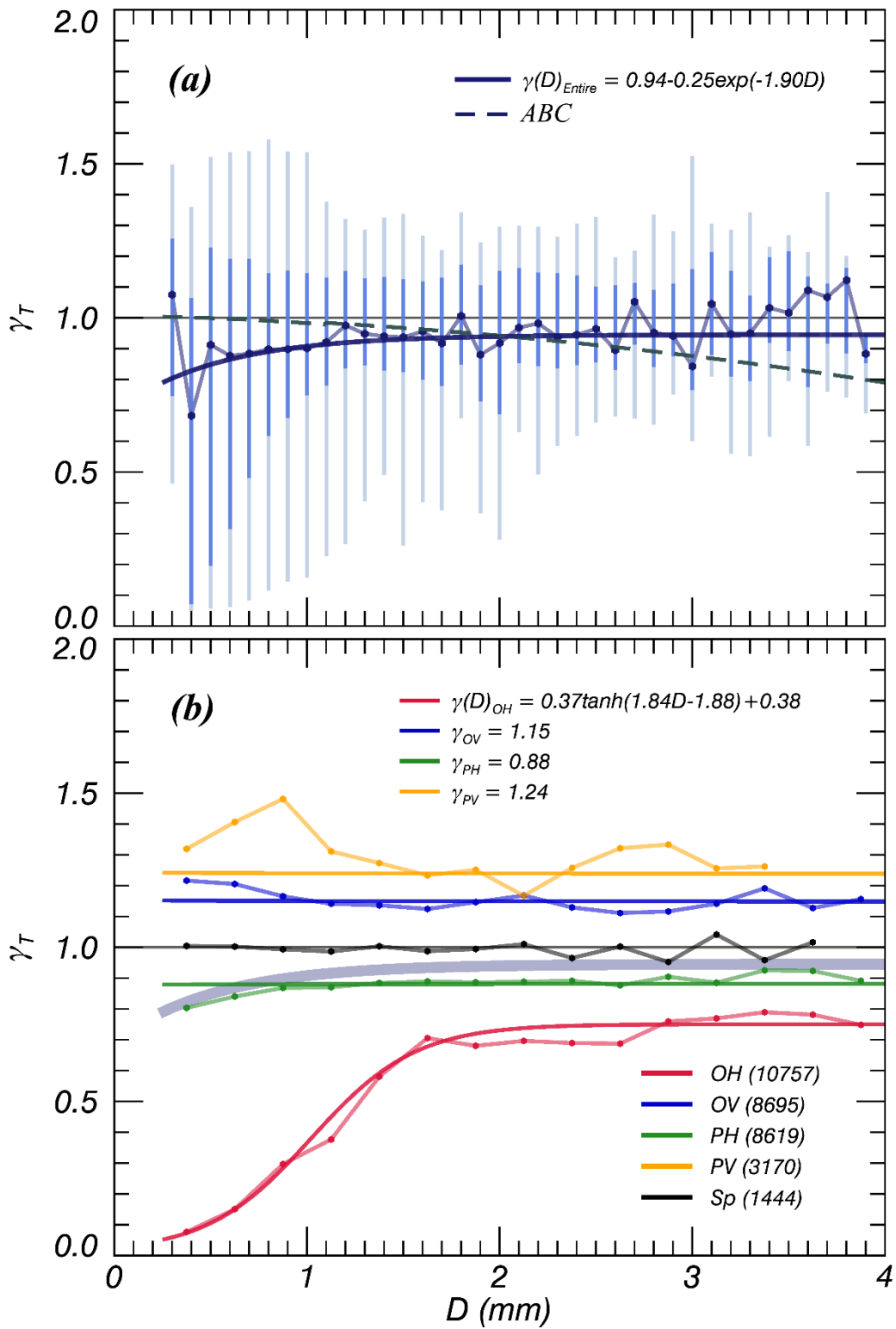
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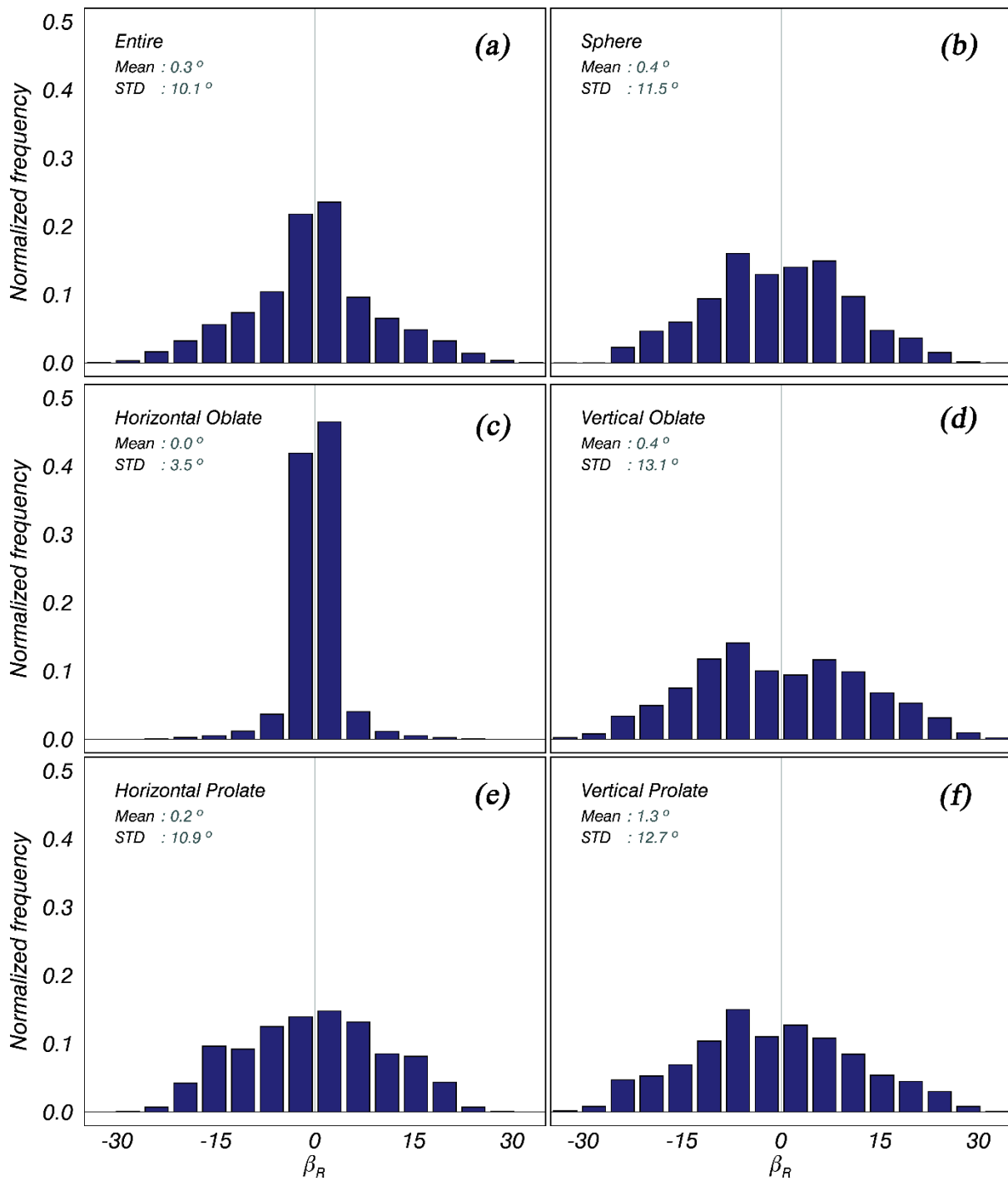
Figure 11. Same as Fig. 10 but for Reynolds number (Re) and drag coefficient (C_D). The grey solid and broken lines in (a) are the relationships of spheres suggested by Clift and Gauvin (1971) and Stokes (1851), respectively.



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936 Figure 12. Same as Fig. 10 but for γ . The grey broken line (ABC) in (a) shows the relationships of
 937 γ for raindrops suggested by Beard and Chuang (1987) and Andsager et al. (1999).

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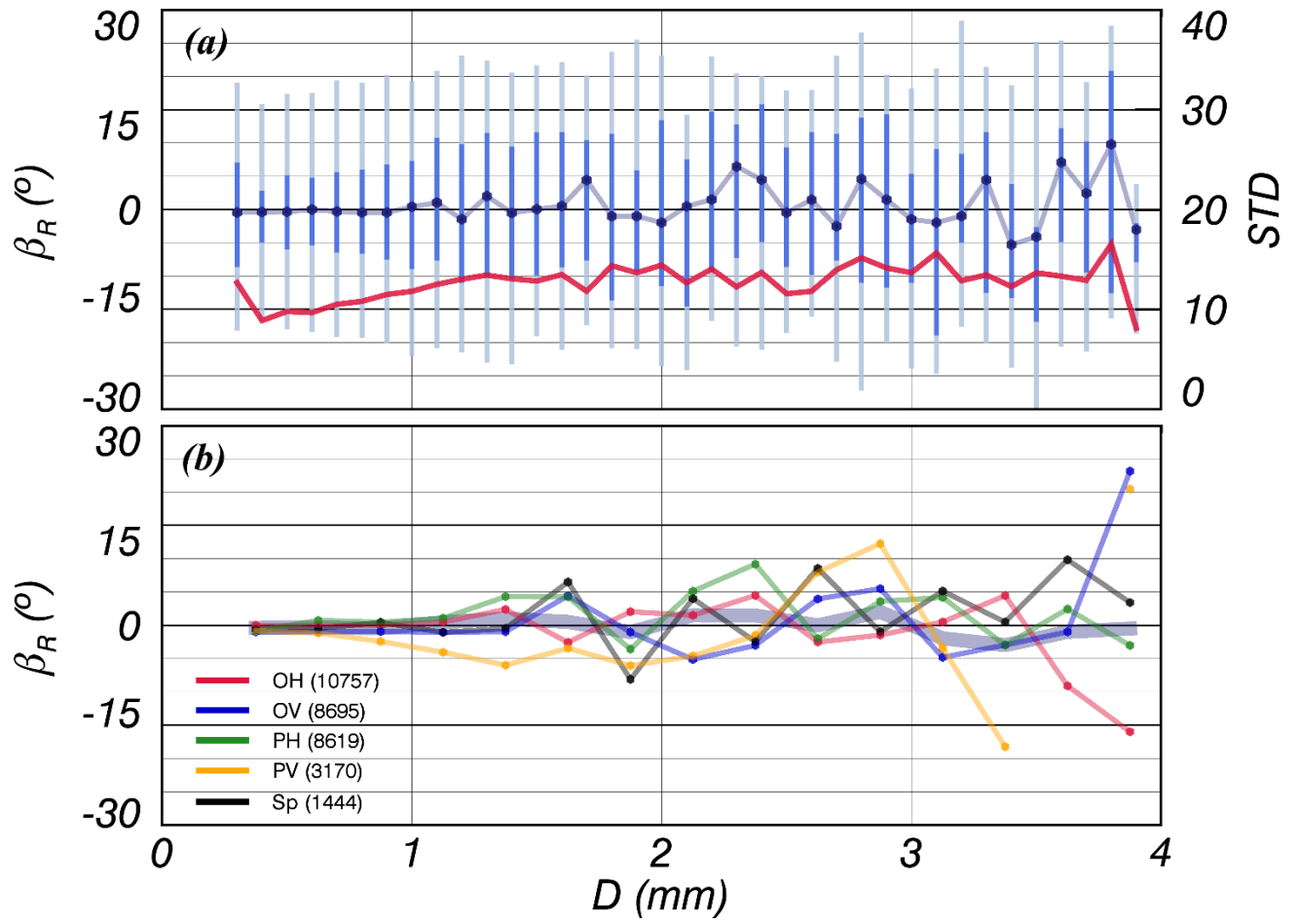
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Figure 13. Histograms of representative canting angle (β_R) for each particle shape type, including the data for all particles.



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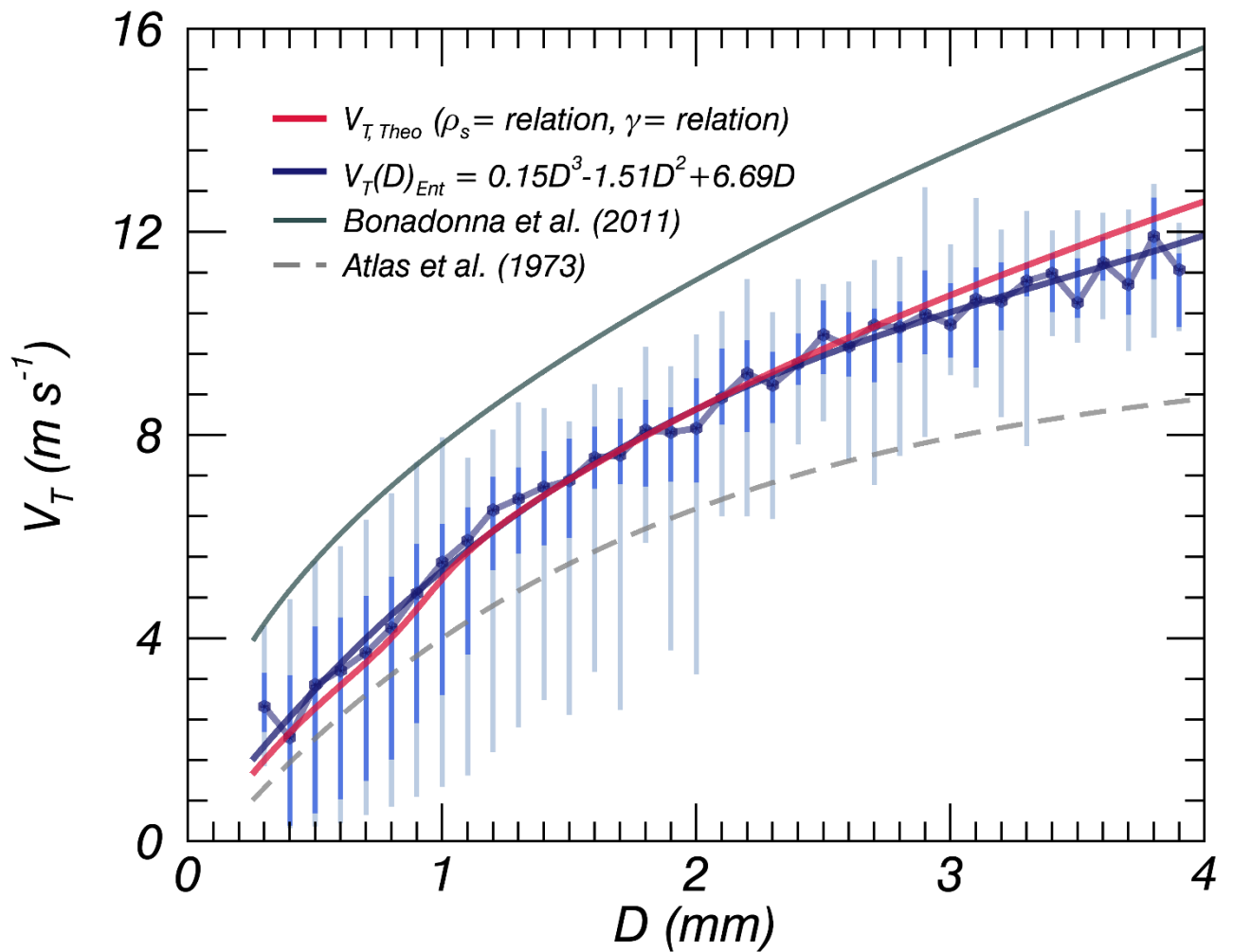
944 Figure 14. Distribution of β_R with D for each particle shape type including the data for all particles.

945 The red solid line indicates the standard deviation.

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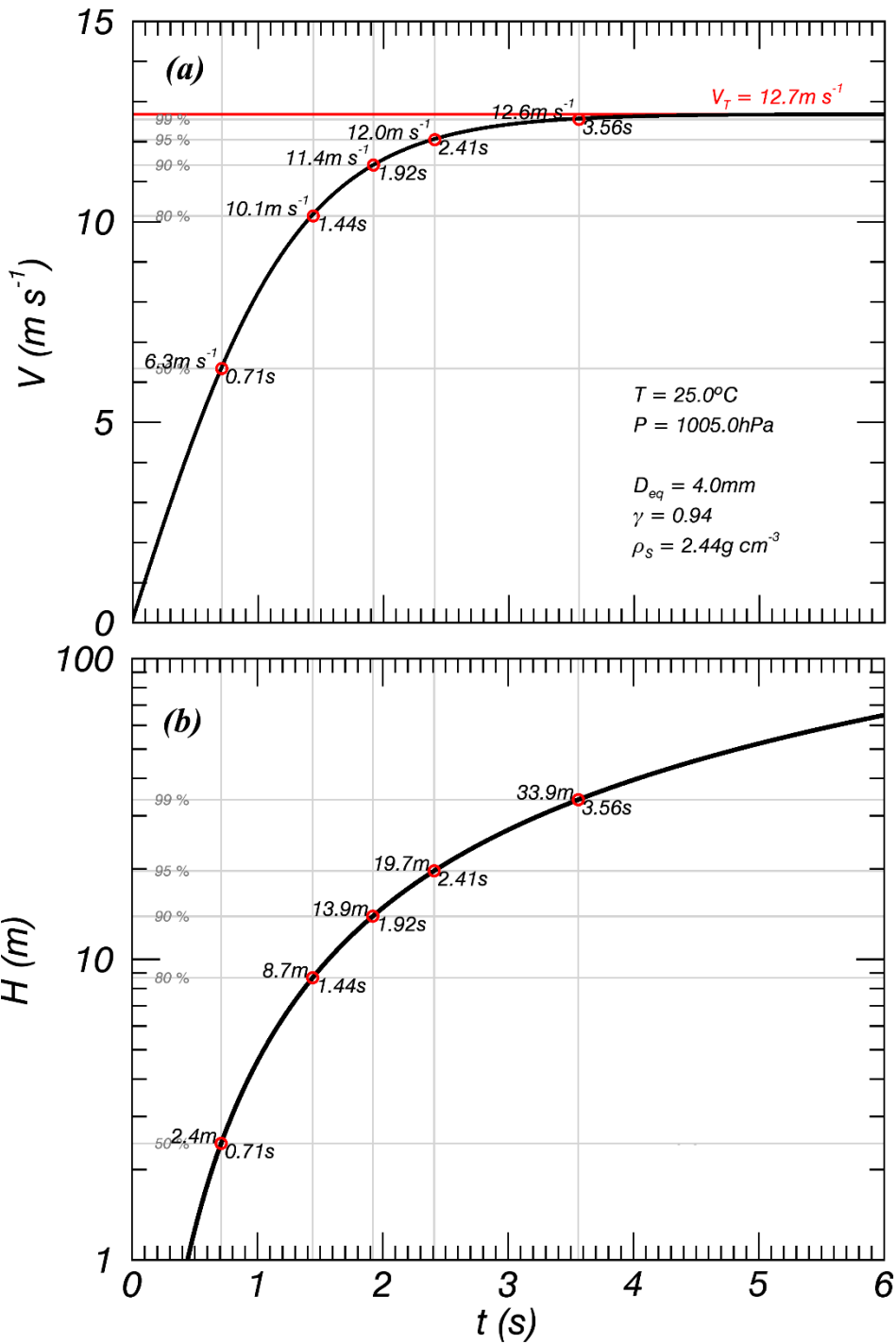
949

950 Figure 15. The V_T quartiles after applying the 60% V_T QC threshold for all, and each individual,
 951 particle shape type, respectively. Grey solid and broken lines in (a) are the relationships of volcanic
 952 ash particles and raindrops suggested by Bonadonna et al. (2011) and Atlas et al. (1971), respectively.
 953 The red solid line indicates the $V_{T, \text{Ref}}$ relationships of γ and particle density suggested by the present
 954 study.

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959 Figure A1. Theoretical (a) fall velocity and (b) falling height for a sphere with $D = 4 \text{ mm}$,
 960 considering the surface roughness coefficient of the volcanic ash particle (1.07^{-1}) relative to its fall
 961 velocity (Bagheri and Bonadonna, 2016).

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963 **Tables**

964

965 Table 1. Particle number by particle shape type after applying quality controls and ash particle
 966 classification criteria.

Type	Description	Classification conditions	Data number (%)
All	All	.	62,953 (100)
OH	Horizontal oblate	$\gamma_{T1(2)} < 0.9$ $ \gamma_{T1} - \gamma_{T2} \leq 0.1\gamma_T$	36,125 (57.38)
OV	Vertical oblate	$0.9 \leq \gamma_{T1(2)} \leq 1.1,$ $\gamma_{T2(1)} > 1.1$	10,000 (15.88)
PH	Horizontal prolate	$ \gamma_{T1} - \gamma_{T2} > 0.1\gamma_T$	11,412 (18.13)
PV	Vertical prolate	$\gamma_{T1(2)} > 1.1$ $ \gamma_{T1} - \gamma_{T2} \leq 0.1\gamma_T$	3,601 (5.72)
Sp	Sphere	$0.9 \leq \gamma_{T1(2)} \leq 1.1$ $ \gamma_{T1} - \gamma_{T2} \leq 0.1\gamma_T$	1,815 (2.88)

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968 Table 2. Information on the collected volcanic ash particles.

Data	Collection date	Re-analysis time (June 18, 2014)	Re-analysis method
A	Dec. 1–31 2008	10:00–12:34 (154 min)	Size by size (phi scale)
B	Mar. 1–31 2010	13:43–14:53 (70 min)	Size by size (phi scale)
C	Feb. 28, 2014	15:11–16:17 (66 min)	Mixed
D	Mar. 31, 2014	16:19–17:05 (46 min)	Mixed
E	Apr. 30 2014	17:07–18:00 (53 min)	Mixed

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971 Table 3. Relationships of terminal velocity with the number of data points, the value of the
 972 correlation coefficient (CC) and the root mean square error (RMSE) after applying the 60% V_T QC
 973 threshold for each particle shape type.

Type	Data number (%)	Relationship ($0.25 < D \text{ (mm)} \leq 4$)	CC	RMSE
All	32685 (100)	$V_T(D) = 0.15D^3 - 1.51D^2 + 6.69D$	0.56	1.22
OH	10757 (34.12)	$V_T(D) = 0.14\exp(2.40D)$ ($0.25 \leq D < 1.6$) $V_T(D) = 4.77D^{0.67}$ ($1.6 \leq D < 4$)	0.94	0.46
OV	8695 (26.60)	$V_T(D) = 5.96D^{0.53}$	0.75	0.85
PH	8619 (26.36)	$V_T(D) = 5.09D^{0.65}$	0.87	0.74
PV	3170 (9.69)	$V_T(D) = 6.47D^{0.49}$	0.71	0.96
Sp	1444 (4.41)	$V_T(D) = 5.61D^{0.56}$	0.91	0.78

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