

# Free-fall experiments of volcanic ash particles

## using a 2D video disdrometer

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Key Points: Volcanic ash, 2DVD, aerodynamic properties, laboratory experiments

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## Abstract

Information of aerodynamic parameter of volcanic ash particles, such as terminal velocity, axis ratio, and canting angle, which are necessary for quantitative ash-fall estimations with weather radar. In this study, free-fall experiments of volcanic ash particle were accomplished using a two-dimensional video disdrometer under controlled conditions.

Samples containing a rotating symmetric axis were selected and divided into five types according to shape and orientation: oblate spheroid with horizontal rotating axis (OH), oblate spheroid with vertical axis (OV), prolate spheroid with horizontal rotating axis (PH), prolate spheroid with vertical rotating axis (PV), and sphere (Sp). The horizontally (OH and PH) and vertically (OV and PV) oriented particles were present in proportions of 76% and 22%, and oblate and prolate spheroids were in proportions of 76% and 24%, respectively. The most common shape type was OH (57%).

The terminal velocities of OH, OV, PH, PV and Sp were obtained analyzing 2DVD data. The terminal velocities of PV were highest compared to those of other particle types. The lowest terminal velocities were found in OH particles. It is interesting the terminal velocities for OH decreased rapidly in the range  $0.5 < D < 1$  mm corresponding to the decrease in axis ratio (i.e., smaller the particle, the flatter the shape). The axis ratios of all particle types except Sp were found to be converged to 0.94 at  $D > 2$  mm.

The histogram of canting angles followed unimodal and bimodal distributions with respect to horizontally and vertically oriented particles, respectively. The mean values and the standard deviation of entire particle shape types were close to  $0^\circ$  and  $10^\circ$ , respectively under calm atmospheric conditions.

## 1. Introduction

Volcanic eruptions are considered one of the most severe types of natural phenomena, and can lead to human casualties and property damage. Ash consists of very fine-grained fragments, volume-equivalent spherical particle diameter ( $D$  in  $\mu\text{m}$ ) is generally smaller than 2 mm, and these are generally dominated by broken glass shards rather than crystal and lithic fragments. Wilson et al. (2012) overview for the ash effects on critical infrastructures such as ash fall and acid rain. Hilman et al. (2012) investigate the effect ash particles on human health in case of Sakurajima volcanic eruptions. More comprehensive descriptions on the volcanic ash impacts on society is found in Sigurdsson et al. (2015) and 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, accurate description of transport and deposition in the numerical forecasting model of volcanic ash clouds are vitally important (Poulidis et al., 2017).

Terminal fall velocity ( $V_T$ ) of a particle is affected by its shape, density, size, and atmospheric properties. Wilson and Huang (1979), Dellino (2005), and Coltelli et al. (2008) introduced the influence of ash particle shapes on its  $V_T$ . Haider and Levenspile (1989) and Ganser (1993) analyzed  $V_T$  of volcanic ash particle on the drag coefficient ( $C_D$ ) which is dependent on the particle shape and atmospheric condition. 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.

Aerodynamic properties are important for safe aviation, and for studying the effects of volcanic ash on climate change, since these parameters determine the residence time of ash particles in the

71 atmosphere (e.g., Folch et al., 2009). The  $V_T$  of particles vary widely due to their irregular shapes and  
72 material components (e.g., Wilson, 1972; Harris and Rose, 1983; Bonadonna et al., 2011, Maki et al.,  
73 2016). Bonadonna et al. (2011) analyzed  $V_T$  of volcanic ash particle with various particle density ( $\rho_s$ )  
74 from 990 to 2738 kg m<sup>-3</sup> and Maki et al. (2016) summarized the list of various  $V_T$  relationships  
75 suggested by previous studies. Volcanic ash particles have a range of shapes, and this presents a major  
76 challenge when analyzing their characteristics. Recently, the irregularity of volcanic ash particles was  
77 analyzed in detail based on the features of various regular particles, such as cubes, cylinders, and disks  
78 (Bagheri and Bonadonna, 2016), using a computed tomography (CT) scanner (Dioguardi et al., 2017;  
79 Garboczi and Bullard, 2017).

80 There are two approaches to studying these aerodynamic properties. The first approach is  
81 theoretical, where a numerical simulation model is used to calculate terminal velocities, drag force,  
82 and Reynolds number (Re); examples of this approach can be found in Happel and Brenner (2012).  
83 The second approach is related to experimental research, in which the aforementioned relationships  
84 are determined experimentally. For instance, Bagheri et al. (2013) and Bagheri and Bonadonna (2016)  
85 analyzed the aerodynamic features of irregular shaped ash particle from the free fall experiments.  
86 Dioguardi et al. (2018) suggested a new model of fluid drag for irregular shaped particles using various  
87 previous researches. Since the aerodynamic feature depends on atmospheric condition and it can affect  
88 the retrieval of  $V_T$ , it would be suggested that it can be analyzed through the free-fall laboratory  
89 experiments for the following main reasons: 1) there is few chance to measure natural falling ash  
90 particles. 2) it can be possible to control the size of ash particle in the free fall experiments. 3) and can  
91 reduce wind effect in the experiments.

92 The present study applies the second approach (experimental research) to clarify the physical  
93 characteristics of volcanic ash particles analyzing the experimental data. The rest of this paper is  
94 organized as follows. Section 2 describes the free-fall experiments of ash particles and methods of

95 analysis. Section 3 presents the results of the free fall experiments. Section 4 is discussions and section  
96 5 summarizes the results.

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## 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  $\beta$  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 (Joss and Waldvogel, 1967), the Precipitation Occurrence Sensor System (Sheppard, 1990), and Parsivel (Löffler-Mang and Joss 2000). 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 cm<sup>2</sup> 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  $\gamma$  and  $\beta$ . 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). Böhm (1989) analyzed the aerodynamic properties of an irregular hydrometeor and 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**

Volcanic ash particles have various shapes that can be detected by 2DVD (Fig. 1). In the case of raindrops, the DSD is dependent upon the break-up and coalescence processes occurring via up and downdrafts, since the forces of gravity and buoyancy can easily affect raindrop shapes (Rosenfeld and Ulbrich, 2003). However, solid particles do not readily change shape when falling without the influence of forces such as collision. It is thus inferred that many particle shapes would be found in the atmosphere, and that it would be possible to define and classify each particle shape type if we were able to accurately detect a single particle. Thus, the range of  $\gamma$  for solid particles would be expected to be wide compared to that of raindrops, and various values of  $V_T$  and  $\beta$  would likely be observed. The  $\gamma_x$  of a particle is defined as the ratio of height to width for the observation direction  $x$ , and its representative value is calculated using the geometric means of the two  $\gamma$  ( $\gamma_1, \gamma_2$ ) detected by cameras 1 and 2, respectively (Eq. 1):

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

Eq. (1).

The difference in angle between the rotating symmetric axis and vertical axis is defined as  $\beta$ . The counter-clockwise (clockwise) movement of the rotating symmetric axis has a positive (negative) value and the entire range is  $180^\circ$  (from  $-90^\circ$  to  $90^\circ$ ) with  $0^\circ$  as the center.

It is necessary to consider the true axis ratio ( $\gamma_T$ ) to correctly define the particle shape (Fig. 2). The apparent axis ratio ( $\gamma_A$ ) considers the effect of  $\beta$  but the  $\gamma_T$  does not. The 2D coordinates ( $x, z$ ) of the particle shape with  $\beta$  are defined as follows:



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

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

where subscript A is the coordinate of the original data coordinate considering the  $\beta$  and subscript T is the modified data coordinate. The symbol  $r$  refers to the length from the data point to the center and the symbol  $\theta$  represents the degrees of data coordinates from the positive x axis, which range between  $0^\circ$  and  $180^\circ$ . In this paper,  $\gamma$  stands for  $\gamma_A$  for convenience.

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

To define these particle shape types, a strict definition of the  $\gamma_T$  is required, which can be calculated from the  $\beta$ . As with the  $\gamma$ , the two  $\beta$  values are automatically calculated by 2DVD. In the case where the  $\beta$  is assumed as  $0^\circ$ , the rotating symmetric axis for OH and PV can be defined, since it is observed for any observation direction parallel to the ground. However, in the case of OV and PH particles, the rotating symmetric axis cannot be defined when the observation direction is parallel. In the case where

the  $\beta$  is not  $0^\circ$ ,  $\gamma_T$  for all particle shape types would not change when oscillation occurs in a direction orthogonal to the observation direction, but it is difficult to estimate both  $\gamma_T$  and  $\beta$  when particle oscillation appears in a direction parallel to the observation direction. The ability to restore the  $\gamma_T$  and  $\beta$  relative to this observation direction is limited, which is one of the main disadvantages of the 2D observation strategy.

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

Based on the definition of  $\beta$  (Fig. 2), the perfect condition with respect to ellipsoids is satisfied when  $|\beta| = 0^\circ (90^\circ)$  for OH and PV (OV and PH); these values are defined as the center values. However, 2DVD calculated that the  $\beta$  for each particle shape type was concentrated around  $|\beta| = 0^\circ (90^\circ)$  with respect to horizontally (vertically) oriented particles, which correspond to OH and PH (OV and PV). Furthermore, analysis of particles with an orthogonal center angle from  $0^\circ$  is difficult, since they have two center angles ( $\pm 90^\circ$ ). To address observation errors and enhance the convenience of analysis, all center angles were set to  $|\beta| = 0^\circ$  and modified to give the representative canting angle,  $\beta_R$ , using the following equation:

$$\beta_R = \beta - \beta_0$$

where  $\beta_0$  is the orienting angle, defined by the central angle of oscillation. In the case of vertically oriented particles (OV and PV),  $\beta_0$  could be defined as  $\pm 90^\circ$ . The sign of  $\beta_0$  follows that of  $\beta$ .

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

### c. Calculate the terminal velocity for the various particle shape types

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 on atmospheric density ( $\rho_g$  in  $\text{g cm}^{-3}$ ),  $T$ ,  $Re$ ,  $C_D$ ,  $D$ ,  $\rho_s$ , and its shape. Kunii and Levenspiel (1969) developed a theoretical  $V_T$  equation:

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

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 Sommerfeld (2008) introduced a progressed  $C_D$  equation considering two types of sphericity: lengthwise ( $\Phi_{||}$ ) and crosswise ( $\Phi_{\perp}$ ). 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  $\mu$  is the dynamic viscosity ( $\text{kg m}^{-1} \text{s}^{-1}$ ), which we assumed to be  $1.983 \times 10^{-5}$  based on atmospheric conditions at a  $T$  of  $25^\circ\text{C}$ . 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 ( $\text{mm}^2$ ). 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):

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

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 tephra is approximately 60–66 %  $SiO_2$  Peléan-type (Oguchi et al., 2009; Takahashi et al., 2013). The major eruptive events in historic age of Sakurajima were 1471–1476 (Bunmei era), 1779–1782 (An-ei era) and 1914 (Taisho era). Sakurajima is an andesitic volcano with two peaks (Kita-dake and Minami-dake). Volcanic activity at Kita-dake ended around 4,900 years ago then it changed to Minami-dake. Activity has centred on Showa crater from 2006 (Iguchi, 2013). Showa crater is located on the eastern flank approximately 500 m east of Minami-dake (Southern Peak) of Sakurajima volcano. It was appeared in 1939 after one month of eruptions that year (Yokoo and Ishihara, 2007). The Minami-dake summit crater was the only active center of Sakurajima volcano until the recommencement of Showa

crater from 1948 to 2006. The eruptive activity of Showa crater was resumed in June 2006 and vulcanian eruptions gradually increased in the autumn of 2009 (Hotta et al., 2016). The Japan Meteorological Agency (JMA) reported that the eruption frequency of Sakurajima would increase significantly from 2009 and the accumulated ash fall exceeded  $3.5 \text{ kg m}^{-2}$  in Kagoshima city in 2012. The Ministry of Land, Infrastructure, Transport, and Tourism (MLITT) installed an operational X-band radar 10.7 km from the vent, as well as 16 automatic volcanic ash weight measurements, to observe volcanic eruptions in 2011 (Fig. 3).

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#### 269 **e. Free fall experiments**

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

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

284 range of Type1 data set was proportional to its phi-scale, since small particles may be contained by  
285 screening.

286 Figure 7 shows the distribution of raw data (the number of data: 274,215) for  $V_T$  and  $\gamma$  with  $D$ .  
287 There were various  $\gamma$  from 0 to 2 when  $D < 2$  mm and most of the data were concentrated near  $\gamma = 0$ .  
288 The  $\gamma$  values converged around 1 and their distributional range decreased with  $D$ . The median value  
289 with a 0.25 mm  $D$  interval corresponded well to the center of the data contour. The median line  
290 converged around  $\gamma = 0.935$  based on the correlation coefficient value (CC). When this was higher  
291 than 0.95 for each  $D$  interval, the data converged. According to this condition, the range of  $2 < D < 5$   
292 mm was satisfied and the mean value was calculated using these data. The  $V_T$  had a wider range when  
293  $D < 2$  mm but the median line corresponded well to the center of the data (Fig. 7b). The line  
294 representing the largest amount of data is lower than the volcanic ash discussed by Bonadonna et al.  
295 (2011). To select a available range of  $D$ , a theoretical terminal velocity equation ( $V_{T,Ref}$ ) for a non-  
296 sphere corresponding to Eqs. 2–7 was used as the reference. The particle density associated with the  
297 eruption of Sakurajima volcano is between 2.43 and 2.59 g cm<sup>-3</sup> (Oguchi et al., 2009), but the actual  
298 particles contain air vacuoles (Van Eaton et al., 2012). This means that  $\rho_s$ , including vacuoles, is  
299 smaller than  $\rho_s$ . Therefore, the minimum  $\rho_s$  was considered to be 2.43 g cm<sup>-3</sup>, and this was used as an  
300 input parameter. The atmospheric conditions of  $T$  and  $P$  were considered from an automatic weather  
301 station (AWS), supported by the JMA. The falling height of a particle which follows the  
302 aforementioned conditions ( $\gamma = 0.935$ ,  $\rho_s = 2.43$  g cm<sup>-3</sup>) when  $D = 4$  was lower than that under the  
303 condition of free fall experiment (17 m) when it reached 90% of  $V_T$  (13.9 m); therefore, the available  
304 data range is considered to be  $D \leq 4$  mm, and this would satisfy the terminal fall velocity. The detailed  
305 equations used in the present study are shown in Appendix A.

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## **f. Quality control procedures**

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

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

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

iii) To consider volcanic ash particle, the sample satisfying in the certain range of terminal velocity relationship was selected. If we consider a single  $V_T$  QC measure for the entire particle shape types, a number of available data will be removed since  $V_T$  critically depends on particle shape. Therefore, we applied a 60%  $V_T$  QC threshold (Jaffrain et al., 2011) for each particle shape type. It could be applied once  $V_T$  relationship of volcanic ash particles is obtained. After selecting the particle shape types and applying these two QC procedures (i and ii), 19.31% of the data (62,953) remained (Table 1).



### 3. Results

#### a. Ash particle shape

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

There was large variation in shape among particles  $0.25 < D < 0.5$  mm, but the variation decreased with increasing  $D$ . All of the particle shape types had the largest number of particle at  $D > 1.0$  mm and the next were shown at  $0.5 < D < 1.0$  mm, except for OH. In total, 95.80% of OH particles had  $D < 1$  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 and OV, 93.63% and 93.87% of these particles, respectively, had  $D < 1$  mm. Beyond  $D > 1$  mm, the differences in the number of particles for each particle shape type were considerably decreased.

#### b. Terminal velocity

The  $V_T$  for the entire particle shape types well follows a polynomial regression analysis which was applied to define the nonlinear relationship between  $V_T$  and  $D$  (Fig. 9a). It corresponds to that obtained by Miwa et al (2015), who analyzed Parsivel data using the same laboratory experiments. There was the inflection point of  $V_T$  at  $D < 1.3$  mm and it came from an increase in the number of OH and an decrease in their  $V_T$  when  $D < 1$  mm. It can be seen the  $V_T$  relationship for each particle shape type.

The observed values of  $V_T$  were well classified by particle shapes (Fig. 9b). The highest values of  $V_T$  were recorded in the order of prolate, sphere, and oblate. Vertically oriented particles had higher  $V_T$  values than horizontal ones. The  $V_T$  for every particle type can be expressed in a power-law form, except for OH. OH particles followed the regression line relatively closely, and showed the highest CC and root mean square error (RMSE) values, of 0.94 and 0.46 m s<sup>-1</sup>, respectively. Horizontally orientated particles had relatively higher correlations (OH: 0.94, PH: 0.87) compared to those with a vertical orientation (OV: 0.75, PV: 0.71). The  $V_T$  relationships and those of statistical parameters are summarized in Table 3.

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

### **c. Aerodynamic properties**

Particle densities were estimated using the  $V_{T,Ref}$ , converged to 2.37 g cm<sup>-3</sup> when  $D < 1.5$  mm (Fig. 10a). This value corresponds well to that of the minimum  $\rho_s$  (2.43 g cm<sup>-3</sup>) reported by Oguchi et al. (2009). The slight difference is likely due to observation errors and the presence of vesicles (e.g., Seligman et al., 2016). The median value of  $\rho_s$  is changed to  $0.5 < D < 1.5$  mm, and this range corresponds to that of  $V_T$ . Horizontally oriented particles (OH and PH) have relatively smaller  $\rho_s$  and vertically oriented particles (OV and PV) have higher density (Fig. 10b); spheres have particle densities that accord best with  $D$ . The median  $\rho_s$  values for all particle shapes converged when  $D < 2$  mm, and the converged  $\rho_s$  values ranged from 2.35 to 2.50 g cm<sup>-3</sup>.

The  $Re$  and  $C_D$  for the all particle shapes ranged from 10 to 4,000 and 0.6 to 20, respectively (Fig. 11a). Higher values of  $C_D$  were observed when  $\log(Re) < 1.845$  ( $Re < 70$ ); above this threshold,  $C_D$  dramatically decreased. These results were derived according to the number of OH particles, which mainly had  $D < 0.5$  mm, and had higher  $C_D$  and lower  $Re$ . Particle shape was divided into two types: OH and others (Fig. 11b). OH particles had higher  $C_D$  compared to the other particle shapes, which in turn showed few differences among themselves. The OH particles experience strong drag forces under the same flow conditions, leading to lower  $V_T$ . The differences between OH and other particles diminished with  $Re < 1,000$ . The other particle shapes had relatively higher  $C_D$  in the range  $10 < Re < 3,000$ . Rong et al. (2015) analyzed the relationship between  $Re$  and  $C_D$  for oblate and prolate particles, and showed that OH particles had higher  $C_D$  compared to the reference line (Clift and Gauven, 1971) in the range  $0 < Re < 400$ . Each relationship is summarized in Fig. 11.

#### d. Axis ratio

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

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

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

Eq. (8)

The particles are more easily classified by shape than by  $V_T$  (Fig. 12b). Every particle shape type was independent of  $D$ , except for OH. The  $\gamma$  of each particle types are expressed the following relationships (Eqs. 9–12):

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

Eq. (9)

$$\gamma_{OV} = 1.15$$

Eq. (10)

$$\gamma_{PH} = 0.88$$

Eq. (11)

$$\gamma_{PV} = 1.24$$

Eq. (12)

The parameter  $\gamma(D)_{OH}$  was calculated using the hyperbolic tangent ( $\tanh$ ) for the following reasons: i) its range of values was wider than those of other particle types; ii) the data distribution changed continuously with  $D$ ; and iii) it was present in a higher proportion (30.44%) compared to the other parameters. We found that variations in  $\gamma$  decreased with  $D$  and the proportion of Sp shapes increased when  $D > 2$  mm. The particle types OV and PH showed a wide distribution over  $1 < \gamma < 1.5$  and  $0.4 < \gamma < 1.0$ , respectively, when  $D < 2$  mm, but the variability in median values was relatively low. The relationships of  $\gamma$  for each particle shape type could be expressed by constant values at  $\gamma = 0.75$  (OH), 0.88 (PH), 1.15 (OV), and 1.24 (PV), respectively, and these differences are around  $\gamma = 0.12$ .

#### e. Canting angle

Statistical analysis of  $\beta$  is required to understand the aerodynamic properties of volcanic ash particles, and the input parameters of T-matrix scattering simulations, to verify the observed radar variables. The histogram of  $\beta_R$  can be shown in Fig. 13

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

The values of  $|\overline{\beta_R}|$  and  $|\sigma_\beta|$  for OH and PV (OV, PH) were  $0^\circ$  and  $3.5^\circ$  ( $0.4^\circ$ ,  $13.1^\circ$ ), and  $1.3^\circ$  and  $12.7^\circ$  ( $0.2^\circ$ ,  $10.9^\circ$ ), respectively. OV (OH) had the highest (lowest) value of  $|\sigma_\beta|$ . This validates the assumption of Marzano et al. (2012) under stable conditions ( $|\sigma_\beta| = 10^\circ$ ). Therefore, we believe that the tumbling phenomenon ( $|\sigma_\beta| > 30^\circ$ ) of the particles under calm atmospheric conditions was likely to be minor.

To analyze the correlation between particle D and  $\beta$ , quartiles for each particle D interval were calculated (Fig. 14). The particles were concentrated at  $|\beta_R| < 30^\circ$  regardless of D, and median values were stable when  $D < 1$  mm for the entire particle shape types; however, fluctuation increased with D (Fig. 14a). The  $|\sigma_\beta|$  values gradually increased from  $10^\circ$  to  $13^\circ$  when  $0.3 < D < 1.3$  mm and variability was greatest around the center ( $13^\circ$ ). This increase in  $|\sigma_\beta|$  would not be expected in the case of a

447 relatively small number of particles (Fig. 8a), since their standard deviation is largely maintained at  
448 about  $13^\circ$  regardless of dataset size.

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

453

#### 4. Discussions

The results of present study can be extended in viewpoint of the radar meteorology: i) the radar observation and ii) ashfall rate ( $R_A$ ) estimations. i) Weather radar operates for a similar purpose to that of meteorological satellites, and provides information for determining the volume, mass, and echo top height of weather systems. Short-duration eruptions, i.e., less than 1 hour, can be detected at high spatio-temporal resolution, especially in the early period of an eruption. The temporal resolution of weather radar is a few minutes for a single volume scan, and depends on the observation strategy and radar band. The spatial resolution of weather radar is a few hundred meters and is proportional to the radar frequency. A number of ash cloud detections were reported in several observational cases in the US and Japan (Maki and Doviak, 2001; Maki et al., 2012). Marzano et al. (2013) summarized 28 major explosive volcanic eruptions detected by weather radars from 1970 to 2011. Harris and Rose (1983) attempted to analyze volcanic ash particle size and total mass using a C-band weather radar. Maki and Doviak (2001) proposed the method to retrieve PSD from radar measurements of volcanic ash and Donnadieu et al. (2012) detected volcanic eruptions using an L-band fixed radar. Marzano et al. (2006, 2012) and Maki et al. (2012, 2014) detected and analyzed volcanic eruptions using weather radars, from theoretical (physical) and experimental (engineering) perspectives. Thus, observability of volcanic ash clouds using weather radar can be confirmed from previous researches. To verify radar-based volcanic ash cloud observations, a scattering simulation can be considered. The basic parameters: axis ratio, canting angle which are the results presented in this study are the input information of scattering simulation to simulate the theoretical radar variables. In particular, the T-matrix scattering simulation developed by Waterman (1965) is useful for calculation of the theoretical backscattering power of non-spherical particles.

Seconds, the  $V_T$  is one of the main parameters for  $R_A$  ( $\text{kg m}^{-2} \text{hr}^{-1}$ ) and it is defined in terms of PSD and  $V_T$  as follows:

$$R_A = V_r \frac{3.6}{10^3} \int_{D_1}^{D_2} V_T(D) N(D) D^3 dD$$

Eq. (13)

where  $V_r$  is the particle volume ratio. In case of sphere, it is considered as  $\pi/6$ .

Marzano et al. (2012b) proposed Z- $C_A$  relationships using the ashfall concentration ( $C_A$  in  $\text{g m}^{-3}$ ) and z. Maki et al. (2016) introduced  $R_A$ -z relationship for Sakurajima eruption case (18. Aug. 2013) using the time integration of  $R_A$  ( $S_A$  in  $\text{kg m}^{-2}$ ) obtained by automatic volcanic ash weight measurements. However, once we use the disdrometer such as 2DVD, it will be possible to estimate  $R_A$  directly since 2DVD can measure PSD,  $V_T$  and  $\gamma$ . Therefore, basic parameters could help to simulate radar variables and estimate  $R_A$  which are necessary to develop the quantitative ash fall estimation (QAE) method.



## 5. Summary and Conclusions

The basic parameters ( $V_T$ ,  $\gamma$  and  $\beta$ ) of volcanic ash particles were analyzed from the free fall experiments with 2DVD. Data were collected with 18 automatic volcanic ash weight measurements performed on Sakurajima volcano, Japan (31.58° N, 130.65° E). To identify the aerodynamic properties of the volcanic ash particles in the samples, a free-fall experiment was conducted in the large-scale rainfall simulator of the NIED, and 274,215 samples were analyzed.

Radar variables are highly dependent on the  $|K|^2$ , size, and shape of particles. Particle types with rotating symmetric axes were assumed to represent volcanic ash particles which have a wide variety of irregular shape. Their orientation was also considered with respect to horizontally (OH, PH) and vertically (OV, PV) oriented oblate and prolate spheroids were studied, respectively.

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

The  $V_T$  of the particles were classified in the order of PV, OV, Sp, PH and OH. These results are consistent with the  $V_{T, Ref}$ , which suggests that 2DVD is a reliable for observing volcanic ash particles under stable weather conditions. A noticeable increase in  $V_T$  for OH in the range  $0.5 < D < 1$  mm occurred through an increase in  $\gamma$ ; this was not observed for other particle types.

The estimated  $\rho_s$  converged to  $2.37 \text{ g cm}^{-3}$  when  $D > 1.5$  mm, and the median value changed over the range  $0.5 < D < 1.5$  mm. The converged value of  $\rho_s$  is consistent with the value for the Sakurajima volcano reported by Oguchi et al. (2009). The relationship of  $C_D$  and  $Re$  were divided into two particle type categories (OH and the others) and  $C_D(\text{OH})$  was dramatically increased in the range of  $Re < 70$ . These results were derived from the particle concentration of OH, which was highest when  $D < 0.5$

515 mm; at this threshold,  $C_D$  was higher and  $Re$  was lower. The range of  $V_T$  over  $0.7 < D < 1.3$  mm was  
516 informed by both  $\gamma$  and  $\rho_s$ .

517 The  $\sigma_\gamma$  decreased in the range of  $D > 0.75$  mm, to 0.15, and converged to  $\gamma = 0.94$ . Maki et al.  
518 (2014) introduced the radar variables and found that  $Z_{DR}$  gradually increased with time; the dominant  
519 values at 10 and 18 min after the eruption were close to 1 and 2 dB, respectively. The results presented  
520 in this study corresponded to volcanic eruption clouds with positive  $Z_{DR}$  since it is a function of  $|K|^2$   
521 and  $\gamma$  (Herzogh and Jameson, 1992). In addition, it can be explained that the size sorting of ash particle  
522 (e.g., Beckett et al., 2015; Stevenson et al., 2015) will affect the increase of  $Z_{DR}$  by the results of  
523 present study.

524 The  $|\sigma_\beta|$  of OV particles with  $|\beta_0| = 90^\circ$  was largest ( $13.1^\circ$ ) among all particle types and OH particles  
525 had the lowest  $|\sigma_\beta|$  at  $3.5^\circ$ . Based on the  $|\sigma_\beta|$  results, the tumbling phenomenon would not be dominant  
526 under calm atmospheric conditions. The quartiles were stable when  $D < 1$  mm for the entire particle  
527 shape types, but increased with  $D$ . The value of  $\sigma_\beta$  was higher when  $D < 1.3$  mm and started to converge  
528 around  $13^\circ$  due to a decrease in the number of OH particles.

529 These results could be the essential information to develop the new approaches for detecting non-  
530 hydrometeors and numerical model. The axis ratio and canting angle of ash particles obtained from  
531 the present study are necessary for scattering simulations.  $V_T$  obtained by the present study suggests  
532 that smaller particles can be transported longer distance. Therefore, it will be useful for scattering  
533 simulation of ash particles to develop QAE and help to improve a numerical model using  $V_T$  obtained  
534 by the present study.

535

## **Author contributions**

Masato Iguchi and Masayuki Maki designed the study. Akihiko Yamaji and Tatsuya Momotani collected the samples and performed the free-fall experiment. Sung-Ho Suh modified the original study theme and performed the study. Masayuki Maki and Sung-Ho Suh performed research, obtained the results and prepared the manuscript along with contributions from all of the co-authors. Dong-In Lee examined the results and checked the manuscript.

## **Acknowledgments**

This work was supported by a Grant-in Aid for JSPS KAKENHI (grant number JP16H03145) and partially supported by a DPRI collaborative research grant (Kyoto Univ. 25G-11). The authors acknowledge the provision of ashfall data and funding for this work from Kyoto and Kagoshima Universities. The 2DVD data were provided by MEXT, Japan. We also thank the NIED for use of their large-rainfall simulator.

## Appendix A

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

$$F = ma = F_g - F_D$$

Eq. (A1)

$$F_g = mg$$

Eq. (A2)

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

Eq. (A3)

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

In Eq. A2,  $g$  is the acceleration due to gravity, considered to be  $9.81 \text{ m s}^{-2}$ , and  $V$  and  $C_D$  in Eq. A3 correspond to the values in Eq. 2–7. The symbols  $\mu$  and  $\rho_g$  are the dynamic viscosity and density of 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.

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

To ensure accuracy, we considered the surface roughness effect of a volcanic ash particle ( $1.07^{-1}$ ) on the fall velocity, as suggested by Bagheri and Bonadonna (2016), and the results for  $D = 4 \text{ mm}$  are shown in Fig. A1.

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Figures

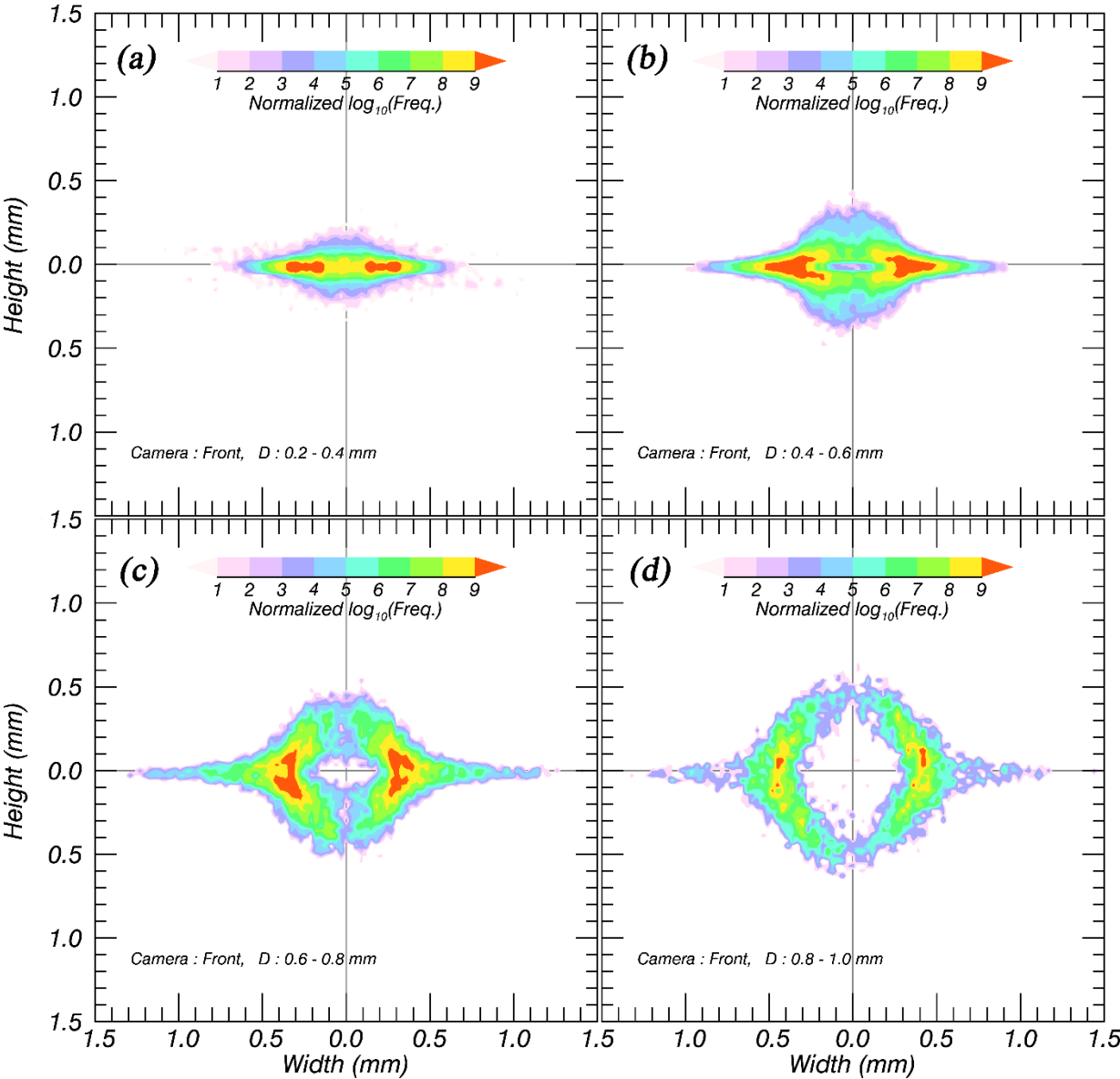


Figure 1. Accumulated contoured images of volcanic ash particles with D measured by a two-dimensional video disdrometer (2DVD).

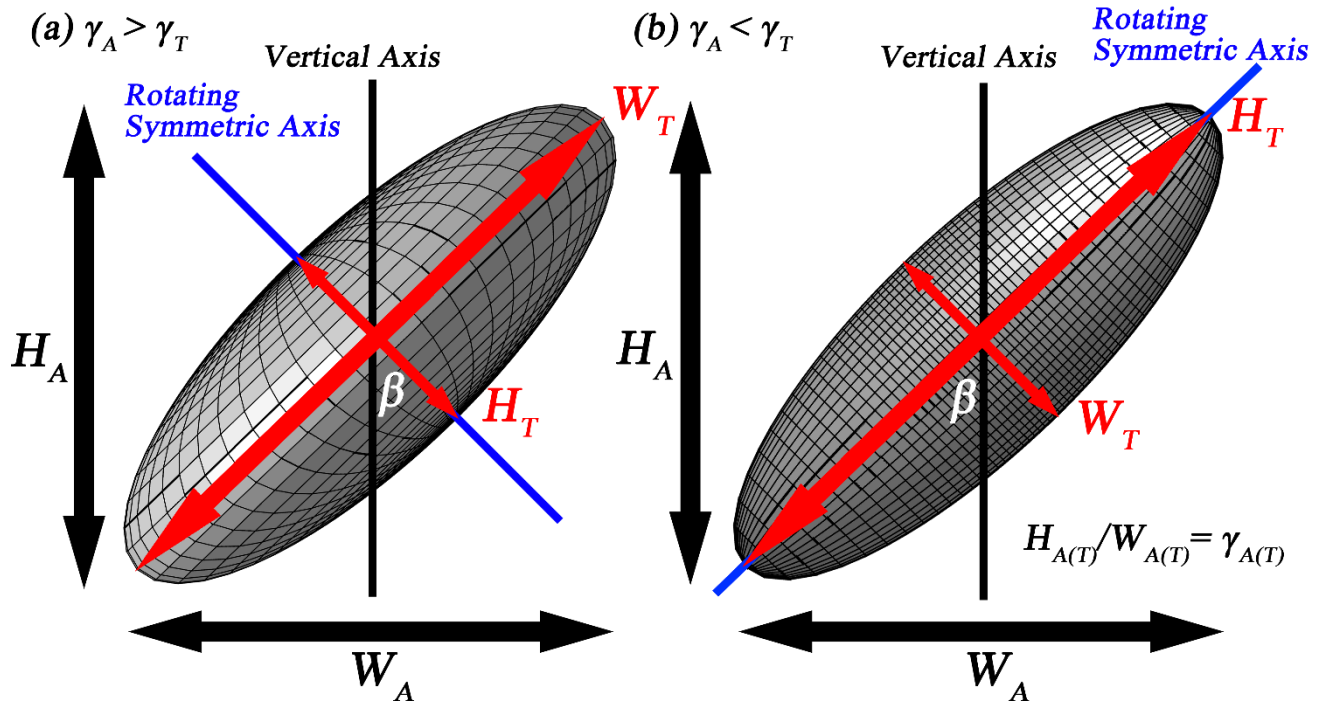


Figure 2. Conceptual model of an (a) oblate and (b) prolate spheroid with the same canting angle ( $\beta$ ).  $W_{A(T)}$  and  $H_{A(T)}$  are the apparent (true) width and height of the particle, respectively.  $\gamma_{A(T)}$  is the apparent (true) axis ratio.

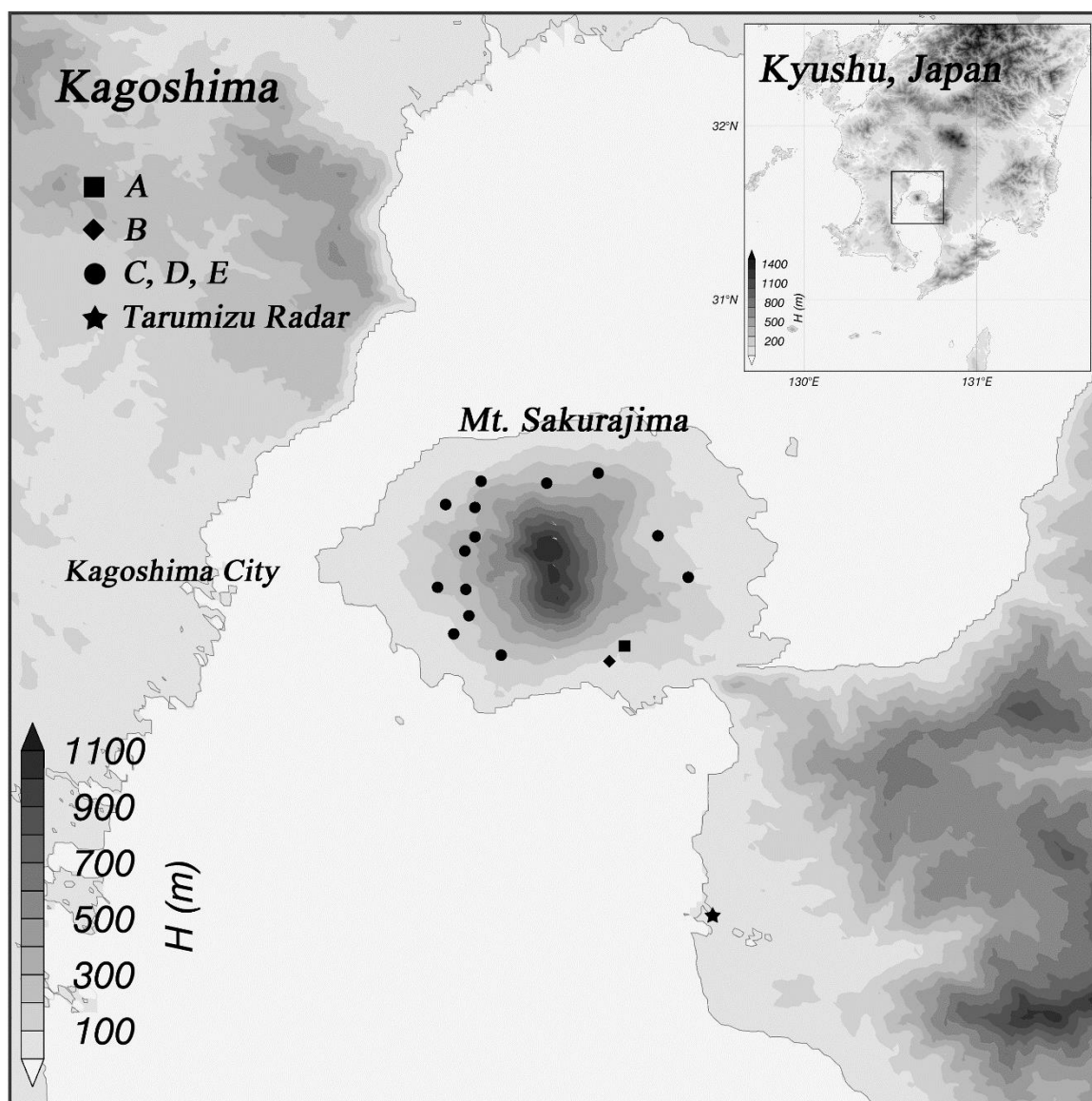


Figure 3. The locations of tephrometers and Showa crater on Sakurajima volcano, Japan. Black symbols indicate the locations of tephrometers and the star, square, and circle symbols correspond to datasets A, B and C–E, respectively. The white circle symbol represents the location of Showa crater.

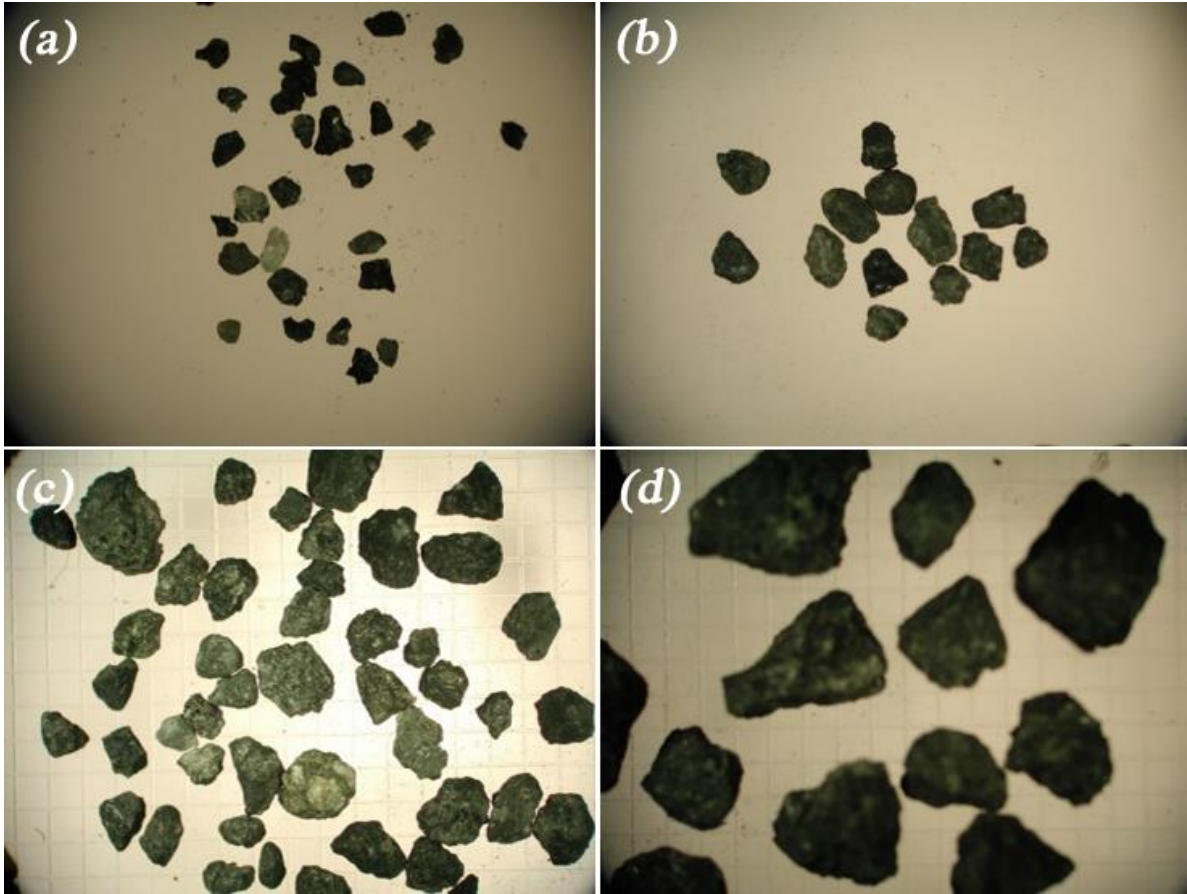


Figure 4. Real images of volcanic ash particles used in the present study. The particles were classified as (a)  $0.125 \text{ mm} < D \leq 0.25 \text{ mm}$ , (b)  $0.25 \text{ mm} < D \leq 1 \text{ mm}$ , (c)  $1 \text{ mm} < D < 2 \text{ mm}$ , and (d)  $2 < D \leq 4 \text{ mm}$ .



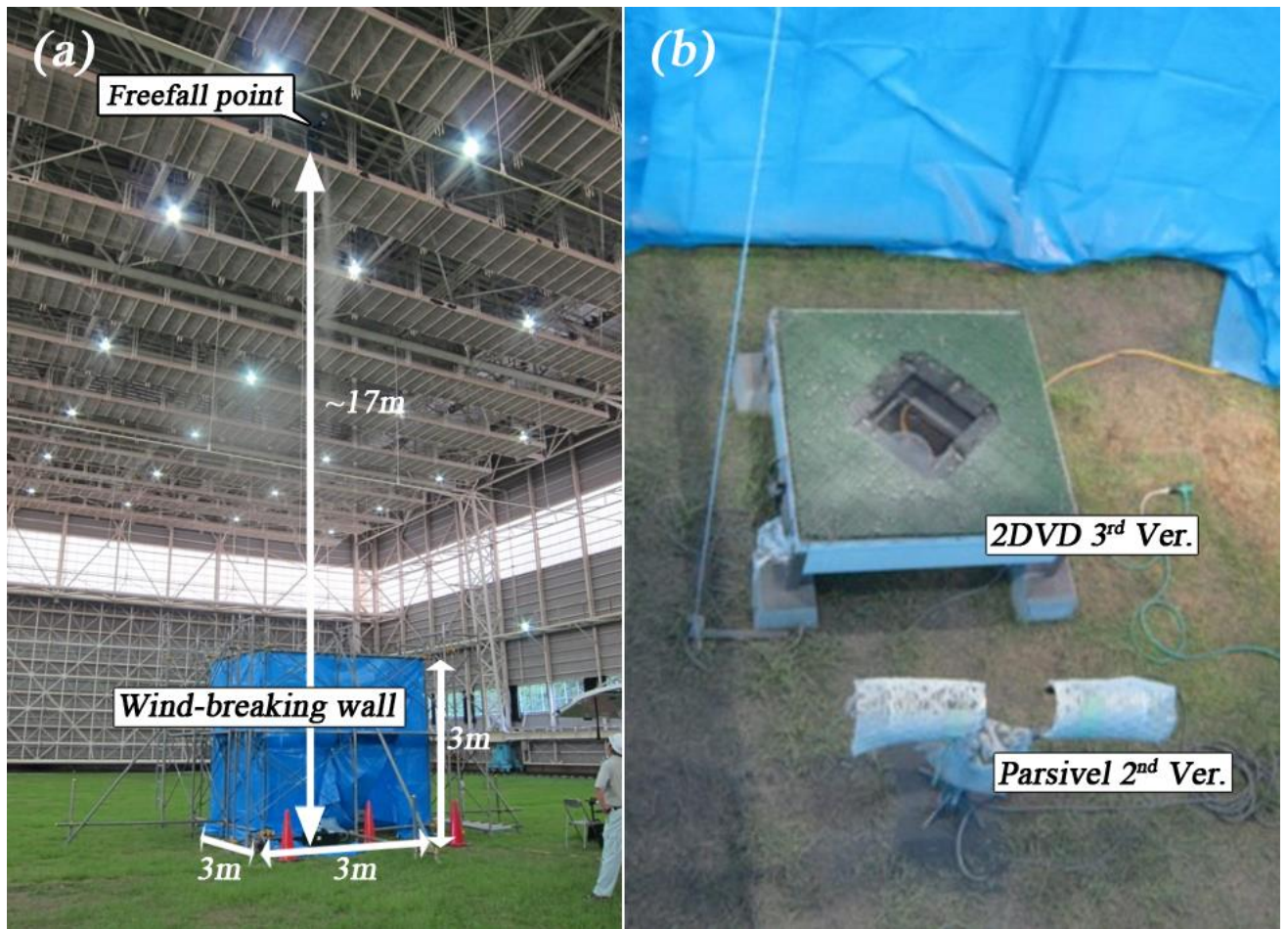


Figure 5. Free-fall experiment conditions of volcanic ash particles on the (a) outside and (b) inside of the wind-breaking wall covering the disdrometers in the large-scale rainfall simulator of the National Research Institute for Earth Science and Disaster Prevention (NIED).

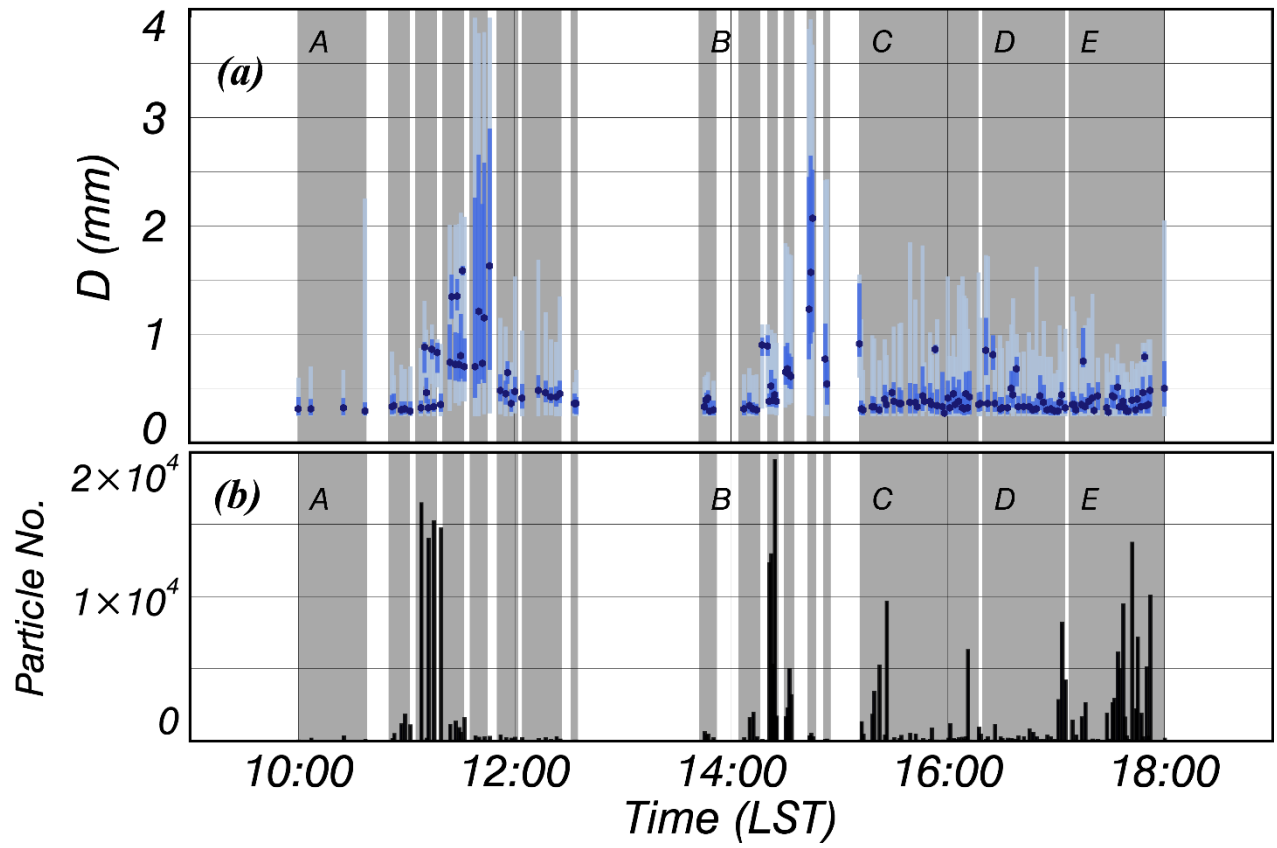


Figure 6. The 1-min interval time series of  $D$  and the number of particles in the free-fall experiment conducted at the NIED.

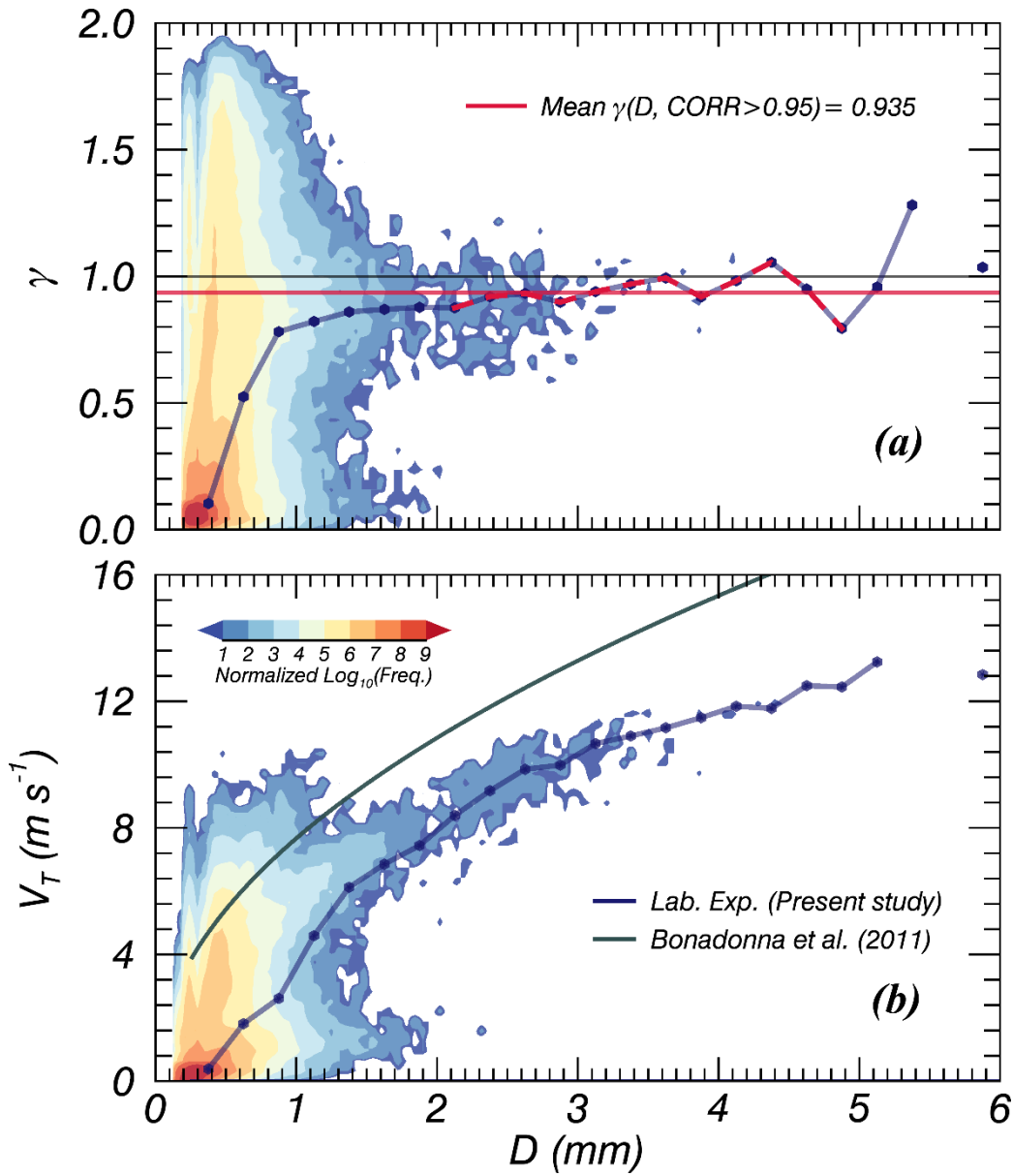


Figure 7. Contour image of volcanic ash particles for (a) the axis ratio ( $\gamma$ ) and (b) terminal velocity ( $V_T$ ) with respect to the sample. The red solid line is the averaged  $\gamma$  satisfying the condition that the correlation coefficient exceeds 0.95. Grey solid line is the relationship of volcanic ash particles suggested by Bonadonna et al. (2011).

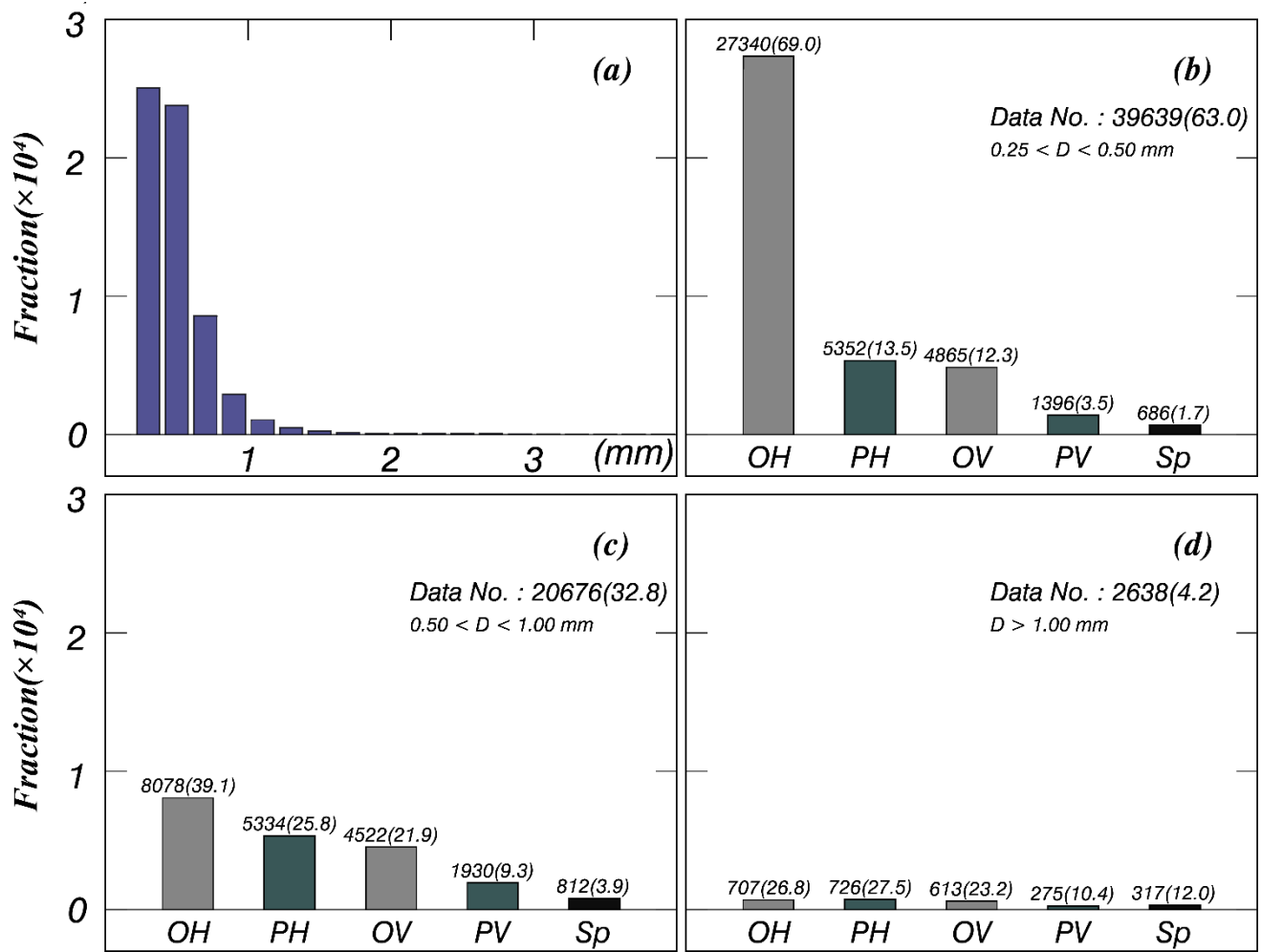


Figure 8. Histograms of volcanic ash particles for (a) all particle types and (b–d) each particle shape type of the phi scale. The grey- and dark grey-shaded (patterned) bars indicate horizontal oblate (OH) (vertical oblate [OV]) and horizontal prolate (PH) (vertical prolate [PV]), respectively. The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

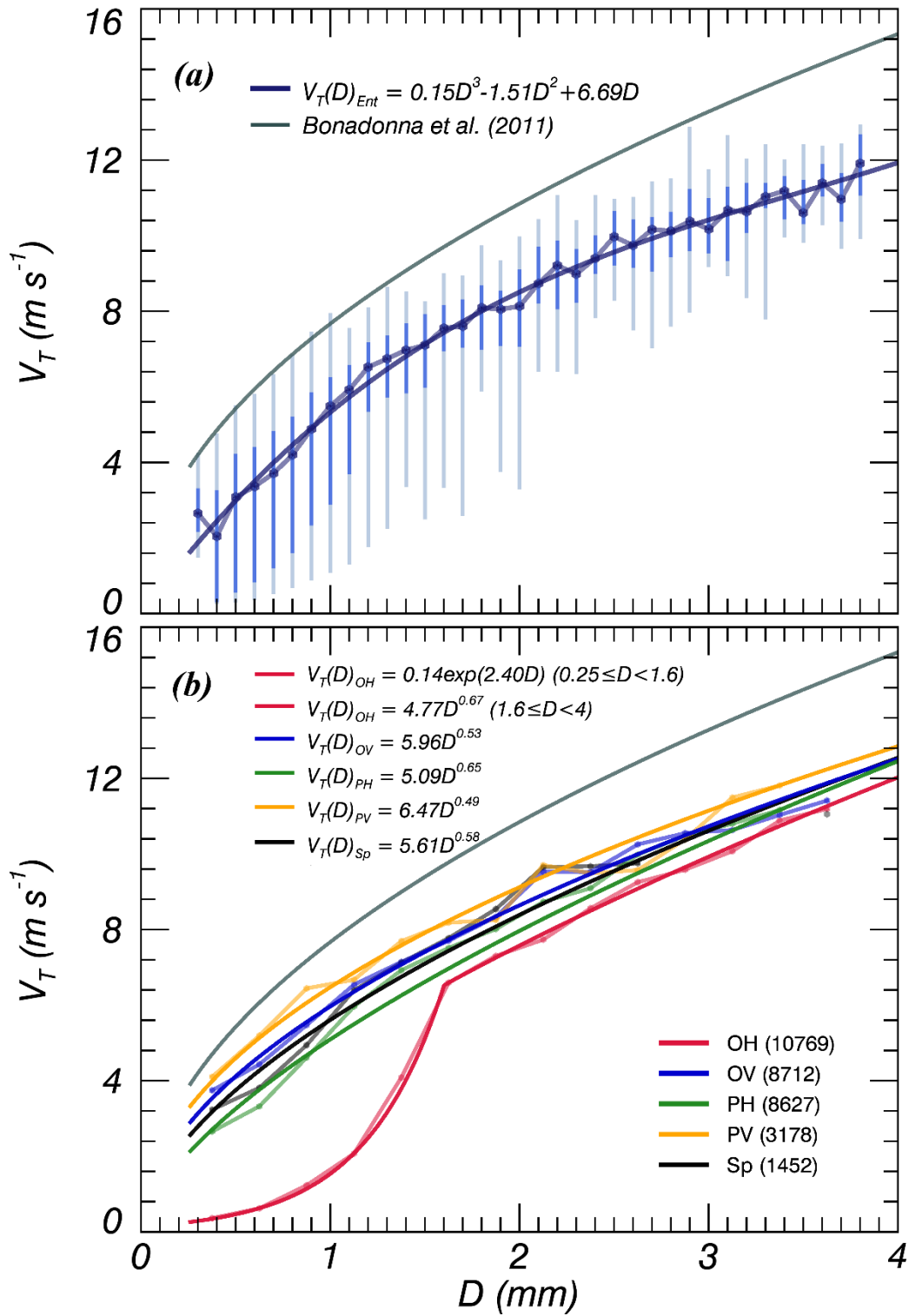
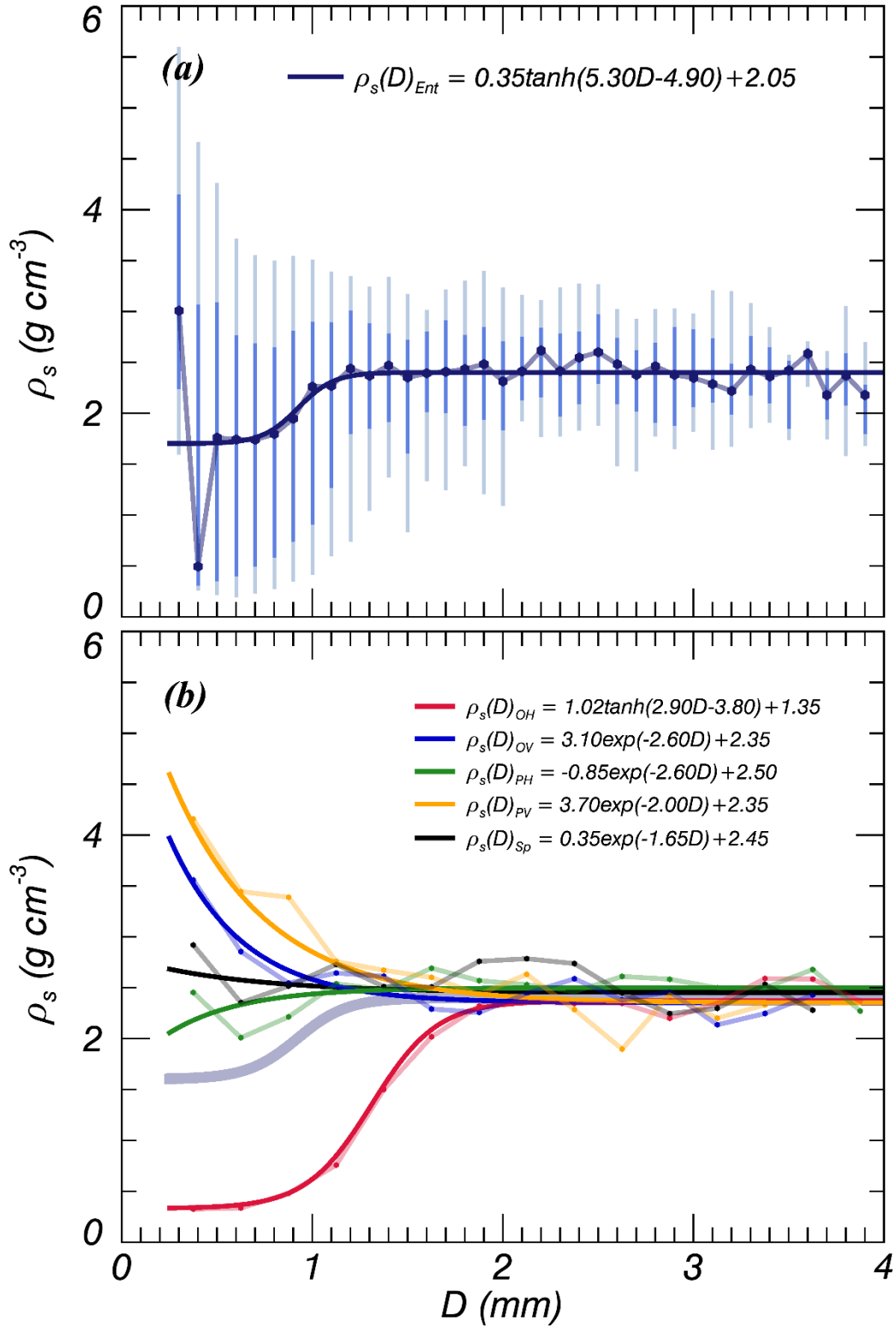


Figure 9. Distribution of (a) quartile and (b) median terminal velocity ( $V_T$ ) values after applying the 60%  $V_T$  QC threshold for all and each individual particle shape type, respectively. The grey solid line shows the relationship of volcanic ash particles suggested by Bonadonna et al. (2011).

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Figure 10. Same as Fig. 9 but for particle density ( $\rho_s$ ).

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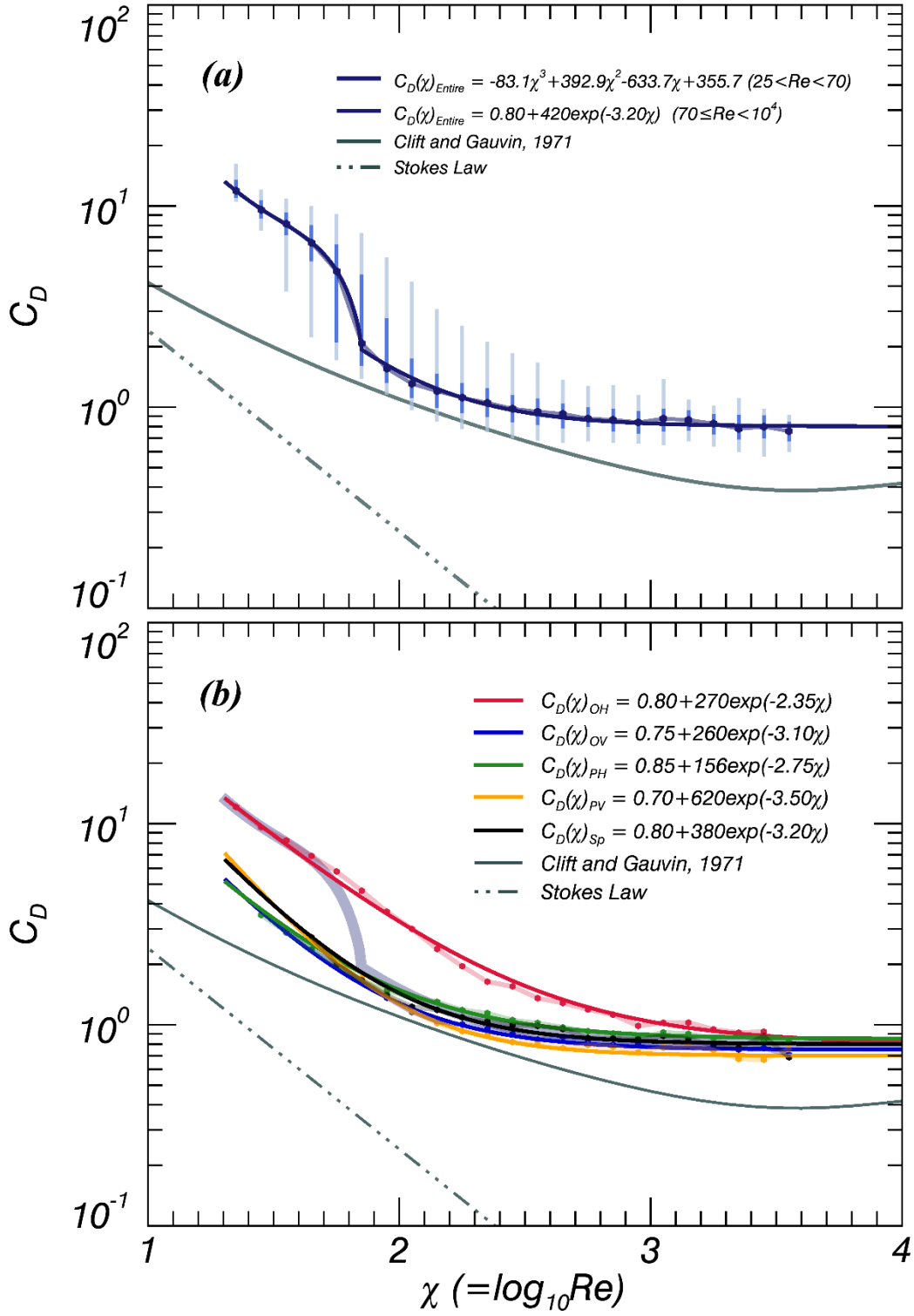


Figure 11. Same as Fig. 9 but for Reynolds number ( $Re$ ) and drag coefficient ( $C_D$ ). The grey solid and broken lines in (a) are the relationships of spheres suggested by Clift and Gauvin (1971) and Stokes (1851), respectively.

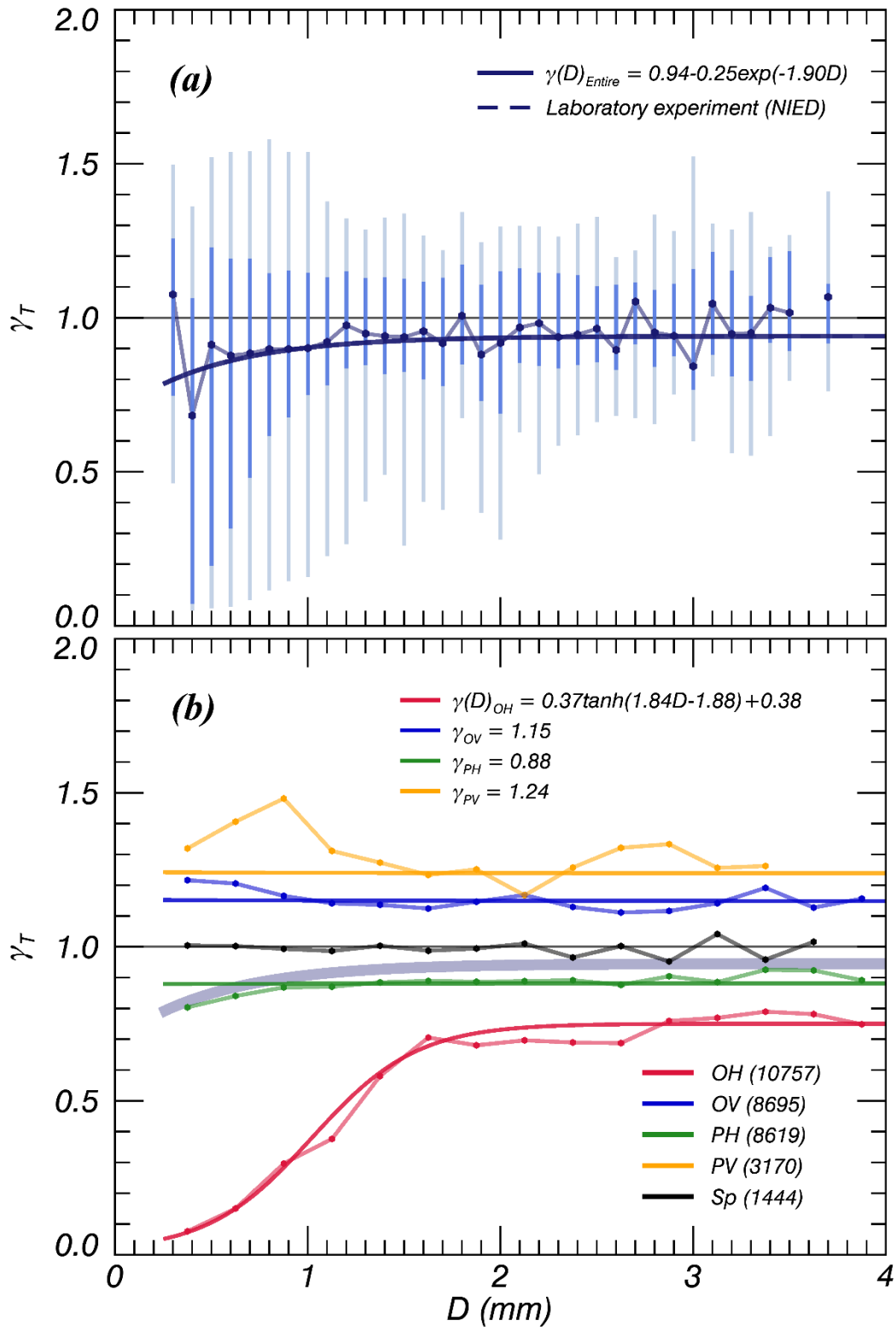


Figure 12. Same as Fig. 9 but for  $\gamma$ .



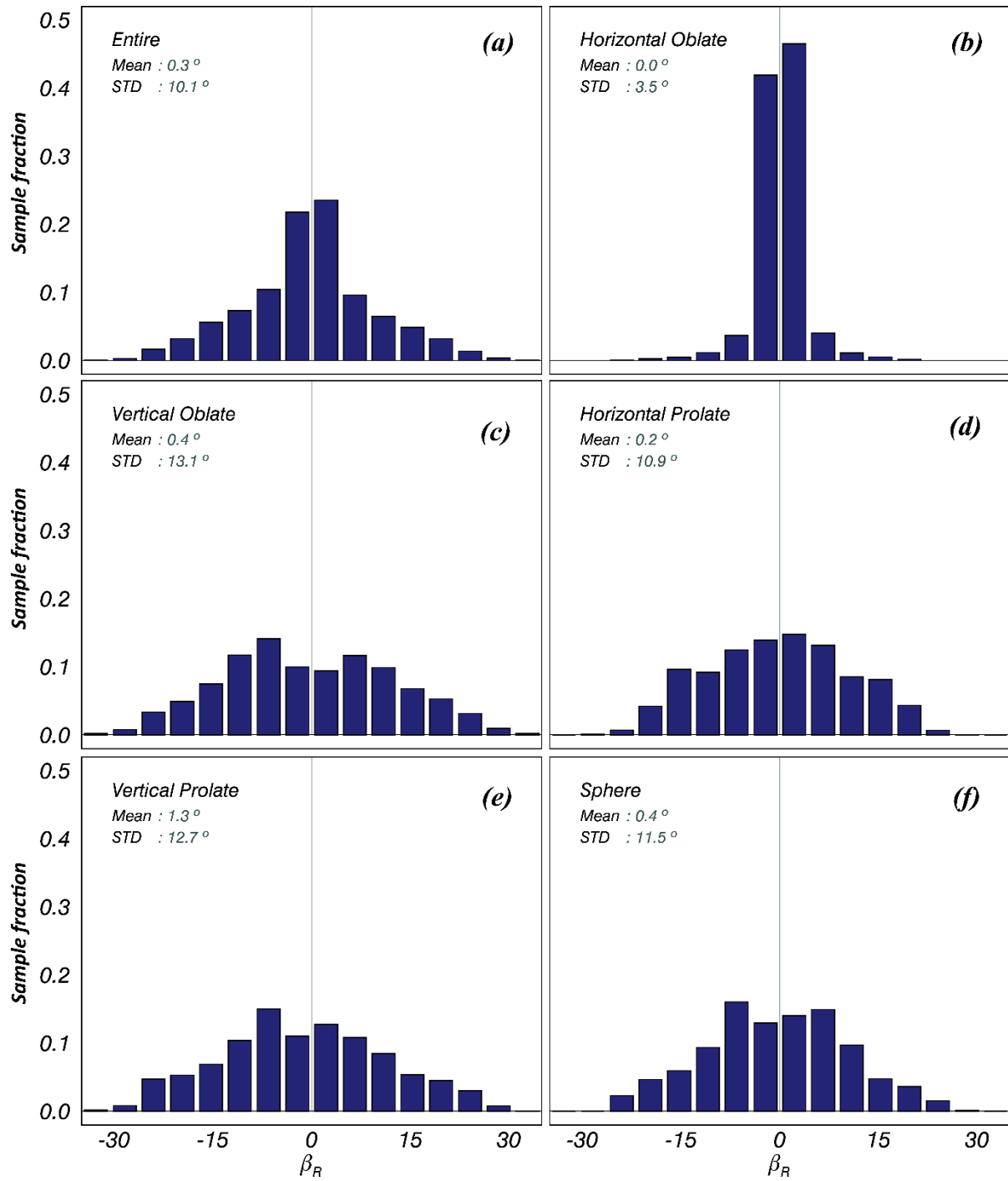


Figure 13. Histograms of representative canting angle ( $\beta_R$ ) for each particle shape type, including the data for all particles.

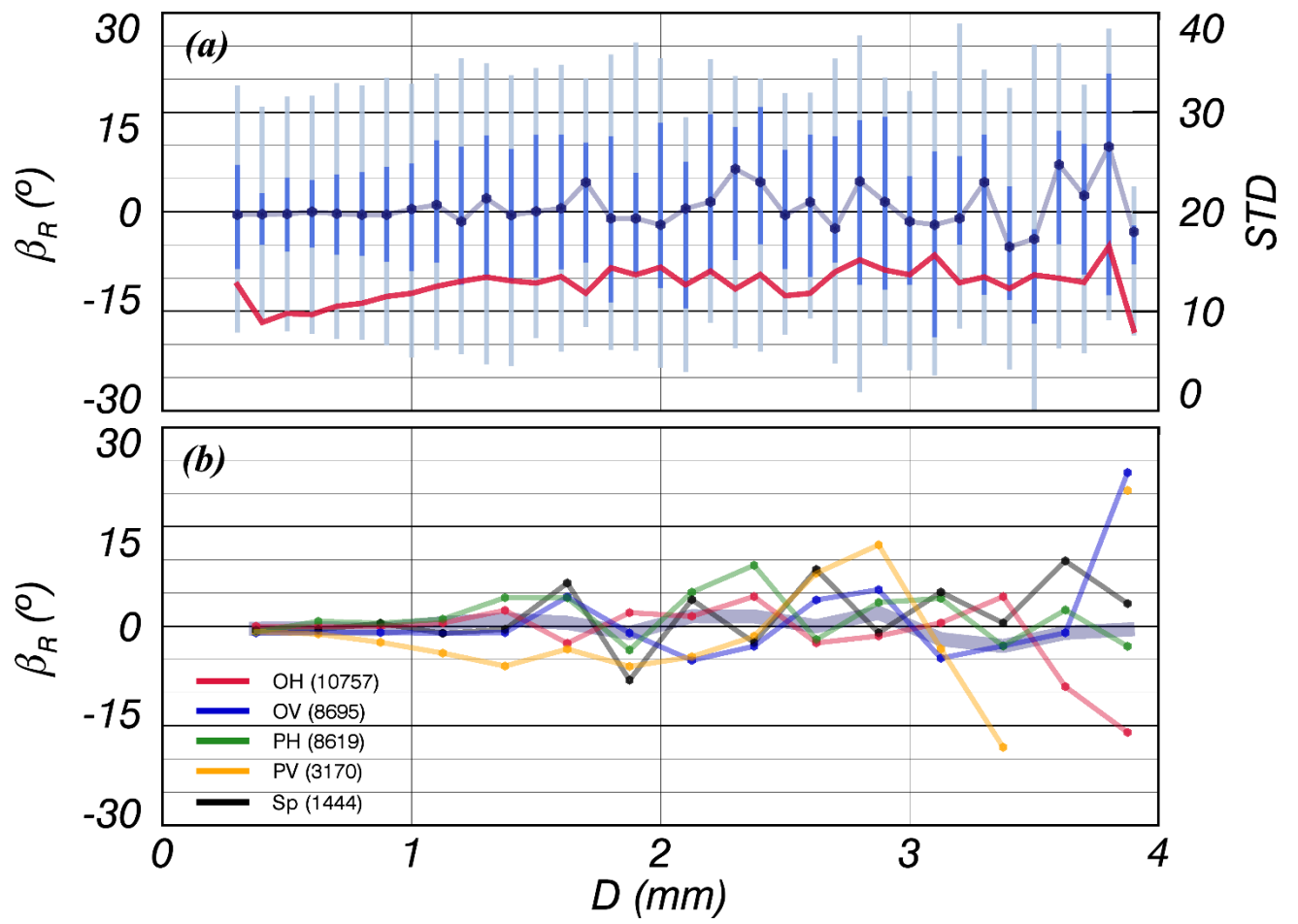


Figure 14. Distribution of  $\beta_R$  with  $D$  for each particle shape type including the data for all particles. The red solid line indicates the standard deviation.

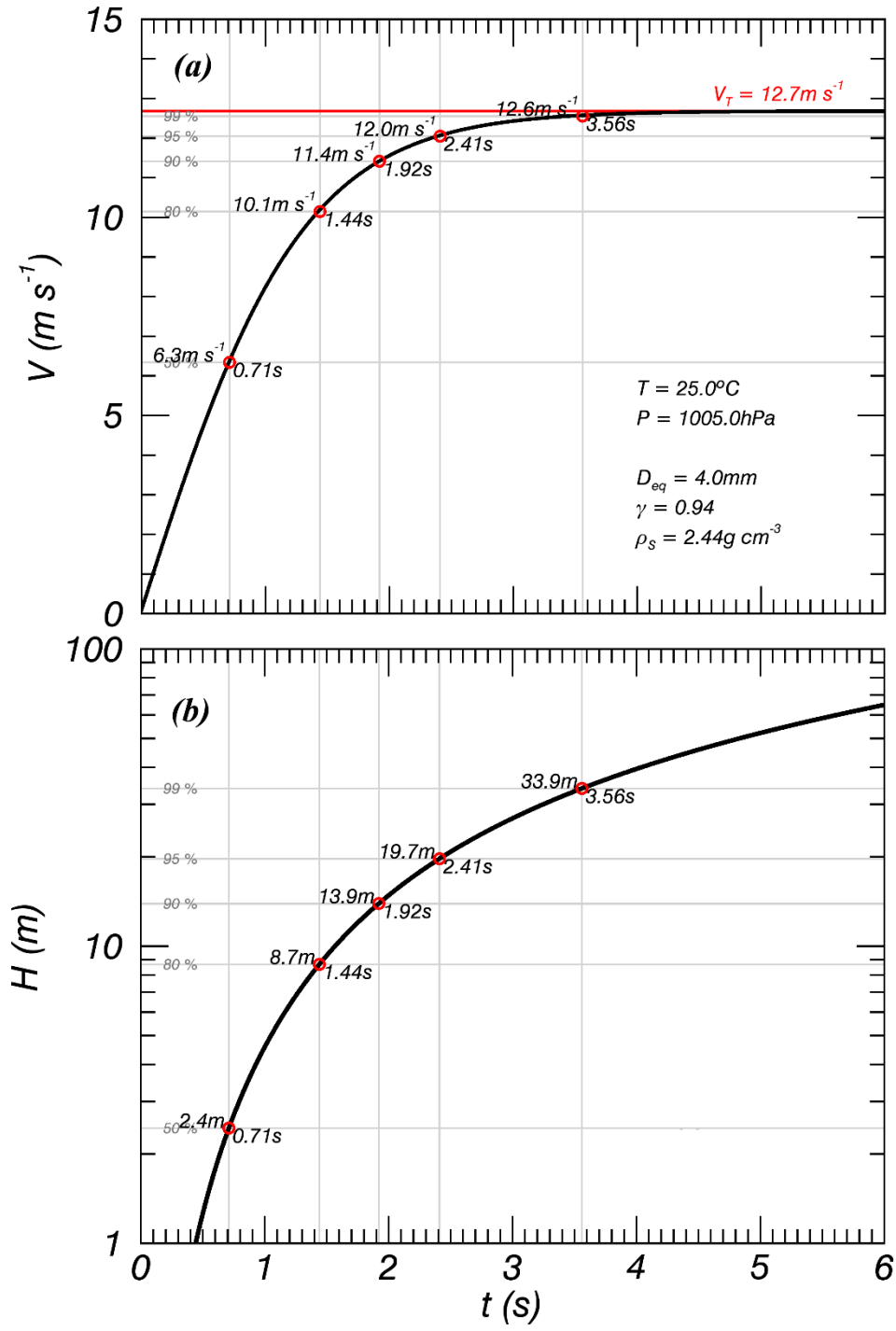


Figure A1. Theoretical (a) fall velocity and (b) falling height for a spheroid with  $D = 4 \text{ mm}$ , considering the surface roughness coefficient of the volcanic ash particle ( $1.07^{-1}$ ) relative to its fall velocity (Bagheri and Bonadonna, 2016).

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820 Table 1. Ash particle classification criteria.

| Type | Description        | Classification conditions  |
|------|--------------------|--|
| OH   | Horizontal oblate  | $\gamma_{T1(2)} < 0.9$<br>$ \gamma_{T1} - \gamma_{T2}  \leq 0.1\gamma_T$             |
| OV   | Vertical oblate    | $0.9 \leq \gamma_{T1(2)} \leq 1.1,$<br>$\gamma_{T2(1)} > 1.1$                        |
| PH   | Horizontal prolate | $ \gamma_{T1} - \gamma_{T2}  > 0.1\gamma_T$  |
| PV   | Vertical prolate   | $\gamma_{T1(2)} > 1.1$<br>$ \gamma_{T1} - \gamma_{T2}  \leq 0.1\gamma_T$             |
| Sp   | Sphere             | $0.9 \leq \gamma_{T1(2)} \leq 1.1$<br>$ \gamma_{T1} - \gamma_{T2}  \leq 0.1\gamma_T$ |

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

| <b>Data</b> | <b>Collection date</b> | <b>Period of free fall<br/>experiment<br/>(June 18, 2014)</b> | <b>Condition of<br/>free fall</b> |
|-------------|------------------------|---|-----------------------------------|
| <b>A</b>    | Dec. 1–31 2008         | 10:00–12:34 (154 min)   | Size by size<br>(phi scale)       |
| <b>B</b>    | Mar. 1–31 2010         | 13:43–14:53 (70 min)  | Size by size<br>(phi scale)       |
| <b>C</b>    | Feb. 28, 2014          | 15:11–16:17 (66 min)  | Mixed                             |
| <b>D</b>    | Mar. 31, 2014          | 16:19–17:05 (46 min)  | Mixed                             |
| <b>E</b>    | Apr. 30 2014           | 17:07–18:00 (53 min)  | Mixed                             |

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

| Type | Data number<br>(%) | 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 (33)         | $V_T(D) = 0.14\exp(2.40D) \text{ (} 0.25 \leq D < 1.6 \text{)}$ | 0.94 | 0.46 |
|      |                    | $V_T(D) = 4.77D^{0.67} \text{ (} 1.6 \leq D < 4 \text{)}$       |      |      |
| OV   | 8695 (27)          | $V_T(D) = 5.96D^{0.53}$   | 0.75 | 0.85 |
| PH   | 8619 (26)          | $V_T(D) = 5.09D^{0.65}$   | 0.87 | 0.74 |
| PV   | 3170 (10)          | $V_T(D) = 6.47D^{0.49}$   | 0.71 | 0.96 |
| Sp   | 1444 (4)           | $V_T(D) = 5.61D^{0.56}$   | 0.91 | 0.78 |

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