Timing and Nature of Volcanic Particle Clusters Based on Field and Numerical Investigations

Gholamhossein Bagheri^{a,b,*}, Eduardo Rossi^a, Sébastien Biass^{a,c}, Costanza Bonadonna^a

^a*Department of Earth Sciences, University of Geneva, Rue des Marachers 13, 1205 Genève, Switzerland*

^b*Max Planck Institute for Dynamics and Self-Organization, Am Faßberg 17, 37077 Göttingen, Germany*

^c*Department of Geology and Geophysics, University of Hawai'i at Mānoa, Honolulu, United States*

Abstract

Aggregation processes are known to play an important role in volcanic particle dispersal and sedimentation. They are also a primary source of uncertainty in ash dispersal forecasting since fundamental questions, such as the timing and deposition dynamics of volcanic aggregates, still remain unanswered. Here, we applied a state-of-the-art combination of field and numerical strategies to characterize volcanic aggregates. We introduce a new category of aggregates observed with high-speed-high-resolution videos, namely cored clusters. Cored clusters are mostly sub-spherical fragile aggregates that have never been observed in the deposits nor on adhesive tape as they typically break at impact with the ground. They consist of a core particle (200-500 μm) fully covered by a thick shell of particles $< 90 \mu\text{m}$. The low preservation potential of cored clusters in ash deposits explains the poor documentation in the literature and the low consideration attributed so far. Cored clusters can also better explain the deposition of fine ash in proximal and medial regions and the polymodality observed in many ash deposits. In addition, numerical inversions show how cored clusters can rapidly form within 175 seconds from eruption onset. Finally, our observations represent the first field-based evidence of the so-called rafting

*Corresponding author

Email address: gholamhossein.bagheri@unige.ch (Gholamhossein Bagheri)

effect, in which the sedimentation of coarse ash in cored clusters is delayed due to aggregation.

Keywords: aggregation, volcanic aggregate, particle cluster, field investigation, numerical, Sakurajima volcano

1. Introduction

Dispersal and deposition of ash during volcanic explosive eruptions strongly affect the surrounding environment and distal atmosphere, with disruptive consequences on local communities and both land and aviation transport (Blong, 1984; Guffanti et al., 2010). Aggregation processes are well known to affect sedimentation of fine ash ($< 63 \mu\text{m}$) by considerably reducing its residence time in the atmosphere (e.g. Lane et al., 1993; Rose and Durant, 2011; Brown et al., 2012; Durant, 2015). If particle aggregation is not considered, volcanic ash transport and dispersal models fail to accurately describe both particle deposition in proximal areas and atmospheric ash concentration in the far field, with important implications for both hazard assessment and real-time ash forecasting (e.g. Folch et al., 2010; Rose and Durant, 2011; Brown et al., 2012).

Volcanic particles mainly collide and cluster because of complex interactions of surface liquid layers, electrostatic forces, turbulence and/or differences in settling velocities. Depending on the water content, particle aggregation results in the formation of *particle clusters* and/or *accretionary pellets* (e.g. Sparks et al., 1997; Brown et al., 2012). Based on the classification introduced by Brown et al. (2012), the category of particle clusters (also known as *dry aggregates* in previous literature) includes both *ash clusters*, noted as PC1, and *coated particles*, noted as PC2 (see Fig. 1). Ash clusters are defined as fragile irregular shaped aggregates composed of particles *typically* $< 1 \mu\text{m} - 40 \mu\text{m}$, whereas coated particles are defined as fragile aggregates comprised of a crystal, crystal fragment, pumice or lithic clast *partially* covered with fine ash particles. The category of accretionary pellets comprises poorly structured pellets, noted as AP1, pellets with concentric structures (also known as *accretionary lapilli*),

noted as AP2, and *liquid pellets* (also known as mud rain), noted as AP3 (Brown et al., 2012).

During the last two decades, several experimental, numerical and field investigations have been carried out to describe aggregation processes in terms of grain-size distribution, terminal velocity, structure, density and porosity (Lane et al., 1993; Schumacher, 1994; Gilbert and Lane, 1994; Schumacher and Schmincke, 1995; James et al., 2002; Taddeucci et al., 2011; Bonadonna et al., 2011; Van Eaton and Wilson, 2013; Telling et al., 2013; Van Eaton et al., 2015; Mueller et al., 2016). However, due to the low preservation potential of particle clusters and most accretionary pellets in deposits, most studies focused on the characterization of the more resistant well-structured pellets (i.e. accretionary lapilli; Gilbert and Lane, 1994; Van Eaton and Wilson, 2013; Van Eaton et al., 2015). In addition, aggregate classifications available in literature are mostly based on both field observations of deposits and Scanning Electron Microscope (SEM) analysis of aggregates sedimented on adhesive tape (e.g. Brown et al., 2012; Sparks et al., 1997; Van Eaton and Wilson, 2013).

Here we introduce new High Speed-High Resolution (HS-HR) observations that reveal additional complexities of particle clusters. In particular, we focus on the relation between ash clusters and coated particles (in the notation of Brown et al., 2012) and we introduce an additional category of aggregates observed with HS-HR videos, namely *cored clusters* (which we define here as PC3 adding on the original terminology of Brown et al., 2012, that firstly introduced PC1 and PC2). Cored clusters are mostly sub-spherical fragile aggregates that have never been observed in the deposits nor on adhesive tape as they typically break at impact with the ground (Fig. 1). They consist of a *core particle* (coarse ash to fine lapilli size) fully covered by a thick *shell* of particles $< 90 \mu\text{m}$.

It is important to note that cored clusters here are different from *poorly-structured pellets*, AP1 (Brown et al., 2012), that include *core-type accretionary lapilli* (Schumacher and Schmincke, 1991), *Type II accretionary lapilli* (Reimer, 1983) and *ash pellets* (Thordarson, 2004; Brown et al., 2012). In fact, the more fragile cored clusters described here break at impact with the ground; as a result,

they more appropriately belong to the particle cluster category. They also differ from the *well-structured accretionary pellets*, *AP2* (Brown et al., 2012), that include *rim-type accretionary lapilli* (Schumacher and Schmincke, 1991), *Type I*
60 *accretionary lapilli* (Reimer, 1983), *armoured lapilli* and *cored lapilli* (with lithic or pumice cores) *accretionary lapilli* and *coated pellets* (Brown et al., 2012), as the cored clusters described here do not present any internal structure.

Field studies of particle clusters (both ash clusters and coated particles) include the investigations of the 1980 eruption of Mount St. Helens, USA (Sorem,
65 1982), the 1990 – 1994 eruptions of Sakurajima volcano, Japan (Gilbert et al., 1991; Sparks et al., 1997), the 1997 eruption phase of Soufrière Hills volcano, Montserrat (Bonadonna et al., 2002b) and the 2010 eruption of Eyjafjallajökull volcano, Iceland (Taddeucci et al., 2011; Bonadonna et al., 2011). Sorem (1982) observed ash clusters with diameters between 250 μm to 500 μm about 390 km
70 from the vent that were composed of ash < 40 μm . He also reported that large ash particles (40 μm to 60 μm) deposited as aggregates far beyond the distance at which they would have settled if traveling independently. This phenomenon is known as *rafting*, which results in the delayed sedimentation of large particles due to coating of fine ash of low bulk density and falling at a lower settling
75 velocity.

Gilbert et al. (1991) and Sparks et al. (1997) reported ash clusters at Sakurajima volcano with diameters < 3 mm, which consisted of particles < 200 μm , whereas coated particles had diameters > 200 μm and were covered with particles < 20 μm . Bonadonna et al. (2002a) observed both types of aggregates (i.e.
80 ash cluster and coated particle) resulting from either dome collapses or Vulcanian explosions at Soufrière Hills volcano. Finally, Bonadonna et al. (2011) observed both types of aggregates at distances between 10 km and 55 km from vent during the 2010 long-lasting eruption of Eyjafjallajökull volcano. Ash clusters had diameters up to 600 μm and consisted of particles < 90 μm , while coated
85 particles were composed of large particles up to 760 μm that were coated with particles < 100 μm . For the same eruption (but different days compared to Bonadonna et al. (2011)), High-Speed (HS) videos recorded by Taddeucci et al.

(2011) show how most aggregates fell with terminal velocities between $1 - 4 \text{ m s}^{-1}$.

90 The first laboratory experiment on the clustering of volcanic particles was conducted by Schumacher (1994), who found that electrically charged volcanic ash from the 1980 eruption of Mount St. Helens could cluster when they are transported within an electrostatic field. The results also indicated how the first aggregates to sediment were coated particles that included a large particle
95 $> 63 \mu\text{m}$ and some accreted fines, whereas the last to sediment were ash clusters composed of particles $< 45 \mu\text{m}$. In addition, Schumacher (1994) observed that large particles between $125 - 180 \mu\text{m}$ can be rafted to long distances when covered with finer ash (i.e. producing a coated particle). Finally, the comparison with grain-size data from other volcanic eruptions suggested that the change in
100 structures of proximal to distal aggregates is not unique to Mount St. Helens.

Almost a decade later, James et al. (2002) conducted a similar experiment to study the role of electrostatic charge on the aggregation of volcanic particles freely falling in dry conditions (relative humidity $\sim 25 - 30\%$). By charging pumices from the 1980 eruption of Mount St. Helens through fragmentation,
105 they found that ash particles $< 70 \mu\text{m}$ could cluster together to create aggregates up to $800 \mu\text{m}$ with density of $\sim 100 - 200 \text{ kg m}^{-3}$ and settling velocity of $< 1 \text{ m s}^{-1}$. They also investigated the possibility of particle rafting through numerical methods. However, they concluded that the rafting of large particles ($< 200 \mu\text{m}$) due to aggregation is unlikely since the window of possibility is very
110 narrow (i.e. the aggregate diameter should not be greater than 3 – 4 times of the diameter of the core particle for rafting to occur).

Despite numerous studies on volcanic aggregates, fundamental questions remain unanswered, such as the structural and aerodynamic characteristics of aggregates, the time required to form, their evolution in time and space, and
115 the modification they impose on sedimentation patterns. The objective of this study is to answer these questions through a multi-technique approach applied to a specific volcanic explosion of Sakurajima volcano (Japan), which includes field HS-HR imaging, analyses of in-situ collected aggregates and numerical in-

versions combined with published data for additional volcanic eruptions.

120 **2. Methods**

2.1. Field investigations

Our experiment was designed to allow for a comparison of pre- and post-depositional features and was conducted at Sakurajima volcano, Japan, which has been continuously active since 1955. The associated ash-rich Vulcanian
125 plumes occurring on an almost daily basis show clear evidence of aggregation (Gilbert et al., 1991; Sparks et al., 1997). Detailed measurements were acquired for one particular explosion that started at 17:47 Japanese Standard Time (JST) on the 3rd of August 2013 (Figure 2A). The observed explosion was comprised of instantaneous bursts of ash-rich plumes, which all were initiated in < 60 s. Such
130 Vulcanian explosions are typical of Sakurajima volcano and can occur several times within a day. The explosion was recorded with a normal High-Definition (HD) camera, from which a maximum plume height of 2.8 km a.s.l. reached within 100 s after the onset is inferred. The rising plume was bent over by the wind and started spreading horizontally ~ 900 m away from the vent (i.e. plume
135 corner) and spread toward south-east with a velocity of $\sim 5.5 \text{ m s}^{-1}$. From the HD movie the cloud base was estimated to be at a height of 2.3 km a.s.l. and constant both in time and distance from the vent (in the considered spatial and temporal window of observation).

Our ground-based observation site was located about 3.7 km downwind from
140 the vent along the dispersal axis (see Figure 2). The total sampling period was relatively short (~ 10 min) since Vulcanian explosions at Sakurajima are inherently short. The sampling was started at 18:02 JST that coincides with the start of the fallout and ended at 18:12 JST, when afterwards no more fallout was observed at our location. During the sampling period, tephra samples were
145 collected in dedicated trays at two sequential time steps in order to monitor accumulation rates and time variations in grain-size distribution: Phase I) between 18:02 and 18:07 JST, associated with individual particles followed immediately

(< 1 min) by sub-spherical aggregates, and Phase II) between 18:07 and 18:12 JST mostly associated with sub-spherical aggregates. Note that the fallout was
150 continuous and the collection in separated trays was carried out only to study the evolution of deposition and aggregation processes in time (i.e. the choice of sampling intervals was arbitrary).

A schematic diagram of the field setup is shown in Figure 3: a High-Speed (HS) camera on a tripod aimed at a thin-glass support covered with a specific
155 double-sided tape allowing for further analyses with a SEM. The HS camera with a resolution of 1200×800 pixels at 800 fps mounted with a Nikon 60 mm f/2.8D AF Micro-Nikkor lens was used at the ground-based observation site in order to capture HS-HR movies with a pixel size of $40 \mu\text{m}$. In order to cross correlate the deposits on the adhesive tapes between the HS-HR movies and
160 SEM analyses, the thin-glass support was kept at an angle during the movie recording. In some cases, however, we had also placed the support horizontally in order to prevent the core-particle from rebound and preserve it for further lab analysis, but our attempts were not successful.

Aggregates were observed while impacting the thin-glass support. HS-HR videos were taken at 18:05 and 18:09 JST and each lasted 10 s, since the camera memory was limited to 8 Gb of storage and videos had to be transferred before the next recording. The movies were then converted frame-by-frame to 8-bit Tiff format images and analysed by the ImageJ software to measure aggregate diameter, position and velocity. Aggregates were mostly sub-spherical and consisted of a core particle that at impact with the thin-glass support separated from the outer fine-ash-rich shell (from here the definition of cored clusters). Both aggregate diameter, d_{agg} , and core-particle diameter, d_{core} , are measured from the HS-HR movies and assumed to be equal to the diameter of a circle with the same area as the aggregate or core-particle image. Terminal velocity measurements were performed within the distance from the top of the videos to the plates, which is in the range of $\sim 10 - 45$ times the aggregate diameter. Aggregate density, ρ_{agg} , was calculated by solving the particle equation of motion

(considering gravity, drag and buoyancy forces) (Bagheri et al., 2013):

$$\rho_{agg} = \rho_{air} \left(1 + \frac{3 C_D v_t^2}{4 g d_{agg}} \right) \quad (1)$$

where ρ_{air} is the air density, C_D is the drag coefficient, v_t is the terminal velocity
 165 of aggregate measured from HS-HR movies and g is the gravitational acceleration. Given that HS-HR videos indicate that aggregates have sub-spherical shapes, their drag coefficient was calculated with the standard drag coefficient model of Clift and Gauvin (1971) for spherical particles.

Tephra collected in dedicated trays (Fig. 3) was manually sieved at half ϕ
 170 down to 500 μm and the fine fraction ($< 500 \mu\text{m}$) was analysed with the laser-diffraction CILAS 1180 instrument (<http://www.cilas.com/>). Particle density, measured by a water pycnometer (with an uncertainty of 3%). Density of core particles, ρ_{core} , observed in HS-HR videos was assumed to be the same as the density of the particles measured with the water pycnometer. SEM images were
 175 processed with the ImageJ software, and particles were measured in the range 1 – 250 μm ($2 - 10 \phi$). Areas of particles were converted to equivalent diameters (assuming spherical shapes) and then to volumes and ϕ values. To obtain the grainsize distribution of aggregates from SEM images, particles in the SEM images were segmented manually or based on edge-detection algorithms.

180 2.1.1. Experimental errors and error analysis

The main error in measuring the velocity of falling aggregates is due to the uncertainty in the aggregate centroid position. Considering the exposure time of movies (150 μs) and the falling velocity of aggregates ($1.2 - 2.9 \text{ m s}^{-1}$), the uncertainty in the aggregate position δy is between 180 – 435 μm (i.e.
 185 exposure time multiplied by the falling velocity). Thus, given that the vertical displacement of aggregates in the image H is $\sim 400 \text{ px} \times 40 \mu\text{m/px}$ ($\sim$ half of the image height), the error on the measured *average velocity* (i.e. averaged over the total falling distance observed in the movies) is between 1.1 – 2.7% ($= 100 \times \delta y / H$). Effects of the thin-glass support (end-effects) on the measured
 190 average velocity is negligible since the total traveled vertical distance is > 5

times the aggregate diameter Chhabra (2006). Moreover, based on the Lorentz formulation (Hoper and Grant, 1948), the instantaneous falling velocity when the aggregate is at its closest distance (i.e. here the last frame in the movie before impacting the support) is at most 2 – 9% higher than measurements in
195 unbounded conditions. Considering all these points and since we are using the average velocity as the aggregate terminal velocity, the end-effects are negligible and not considered in our calculations.

The average error on the measurement of d_{agg} from HS-HR movies is about 5% since in some frames they are not completely in focus. Converting 2D
200 measurements from images to volume was done through correlations presented by Bagheri et al. (2015) that can be associated with an average error of 3% and uncertainty of < 10%. This can result in 15 – 25% error on the aggregate density. Uncertainty on d_{core} is higher since core particles are more irregular than aggregates. To reduce this uncertainty, we assumed a shell density, ρ_{shell} ,
205 of 60 – 220 kg m⁻³ (James et al., 2002; Brown et al., 2012) for the aggregate and as a result the diameter range of core particles is further constrained based on the measured ρ_{agg} and the mass conservation law. Through this procedure the uncertainty on the core diameter reduces to $\pm 10\%$. This uncertainty, however, can be higher if our assumption of the shell density is not valid.

210 Finally, some of the fine particles in the trays might have been lost when we collected samples into plastic bags. In addition, some re-suspended fine particles might have been transported in the trays and contaminated our samples. Nonetheless, we did not observe any strong wind at the ground level and, therefore, we consider this effect negligible. Finally, grainsize distribution made by
215 image analysis of SEM images can be biased by particle overlapping.

2.2. Numerical investigations

Field observations were inverted using a two-dimensional Lagrangian particle tracking code to constrain the trajectory and the timing of aggregates observed at the ground-based observation site. The purpose of the numerical inversions is to have a rough estimation of the duration that aggregates spent in the plume,

spreading cloud and atmosphere in order to be able to estimate an upper-bound for the time needed for particles to form aggregates. Numerical inversions were performed for the aggregates sedimented during both fallout phases (i.e. Phase I and Phase II). Forces considered in the particle equation of motion are the drag, buoyancy (although negligible) and gravity:

$$\frac{d\mathbf{v}_{\text{agg}}}{dt} = \left(\frac{\rho_{\text{agg}} - \rho_{\text{air}}}{\rho_{\text{air}}} \right) \mathbf{g} - \left(\frac{3\rho_{\text{air}}}{4\rho_{\text{agg}}d_{\text{agg}}} \right) C_D |\mathbf{v}_{\text{agg}} - \mathbf{v}_{\text{air}}| (\mathbf{v}_{\text{agg}} - \mathbf{v}_{\text{air}}) \quad (2)$$

where $\mathbf{v}_{\text{agg}}$, $\mathbf{v}_{\text{air}}$ and $\mathbf{g}$ are vectors of aggregate velocity, air (wind) velocity and gravitational acceleration, respectively. The drag coefficient, C_D , was calculated with the model of Clift and Gauvin (1971) for spherical particles. If we consider sub-spherical aggregates with flatness and elongation of 0.80, the terminal velocities by using model of Bagheri and Bonadonna (2016) is 10% lower than those of spherical aggregates. However, we considered spherical models in order to decrease the complexity of the numerical simulations and enhance the generality of our results. ρ_{agg} and d_{agg} are obtained from velocity measurements (through HS-HR movies) at the ground-based observation site. Atmospheric properties (i.e. air density and viscosity) were calculated based on atmospheric sounding measurements available from Wyoming University database (<http://weather.uwyo.edu/upperair/sounding.html>). Air density and viscosity vary in the atmosphere with altitude and, therefore, they were interpolated and updated at the particle position as it falls.

We assumed that aggregates formed either in the rising plume or in the horizontal cloud due to high concentration of particles (Veitch and Woods, 2001; Costa et al., 2010), and that they do not grow/shrink due to aggregation/disaggregation after they are released. We have neglected effects of the convection within the plume and atmospheric turbulence. These assumptions are valid given that we are only interested in the average time that aggregates spend in the plume or atmosphere and not in the individual trajectory for each aggregate.

Four different wind profiles were used since no direct measurement was available: i) a profile that linearly increases the wind speed from 0 m s^{-1} at the

ground to 5.5 m s^{-1} at the height of cloud base (2.3 km a.s.l.), ii) a two-segment linear profile based on experimental measurements of turbulent flow behind pyramids that linearly increases the wind speed from 0 m s^{-1} at the ground to 3.8 m s^{-1} at the vent height ($\sim 870 \text{ m a.s.l.}$) and then to 5.5 m s^{-1} at the height of the cloud base (Ikhwan, 2005), iii) reanalysis data from ERA-Interim database (ECMWF, European Center for Medium range Weather Forecasting), iv) interpolated data from atmospheric sounding measurements available at the Wyoming University database. It should be noted that the plume spread is measured directly from the HD movie and is independent of the wind profile assumptions.

In the simulations, fifty aggregates were released along the plume margins at increasing heights/distances from the vent with time (independent of the wind profile and with the same rate as the plume rise/spread) and at 0.05 s intervals for a total duration of 10 minutes. As a result, a total of 10^4 aggregates were tracked for each wind profile and fallout phase. Only particles matching the sampling location (i.e. $3.7 \pm 0.25 \text{ km}$) and timing (i.e. total ejection to sedimentation time of 15 min-20 min for Phase I and 20 min-25 min for Phase II) were considered. The range of $\pm 0.25 \text{ km}$ is an arbitrary value, which we chose to increase the number of deposited particles during each phase for a reliable statistical analysis.

3. Results

3.1. Field Observations

Accumulation rate for Phase I (the first 5 minutes of fallout) was $2.9 \pm 0.3 \text{ g m}^{-2} \text{ min}^{-1}$, which decreased down to $2.2 \pm 0.2 \text{ g m}^{-2} \text{ min}^{-1}$ for Phase II (the second 5 minutes of fallout). Individual particles were recognized as mostly non-vesiculated fragments with diameters between $\sim 300 - 1200 \text{ }\mu\text{m}$ and density of $2300 - 2700 \text{ kg m}^{-3}$ (measured by water pycnometer). The variation of the density of particles between $63 - 1000 \text{ }\mu\text{m}$ ($0 - 4 \phi$) was found to be a linear function of the diameter (Figure 4).

270 HS-HR videos revealed that all aggregates of both phases are composed of
a single or multiple coarse-ash particles acting as cores that rebounded upon
impact with the adhesive tape. As a result of the impact, only an ensemble of
ash $< 90 \mu\text{m}$ (i.e. shell particles) was preserved on the adhesive tape (Figure
5A, B). Traditional SEM images of particle clusters deposited on adhesive tape
275 (e.g. Sparks et al., 1997; Bonadonna et al., 2011) can only show the occurrence
of partially coated particles and broken ash clusters that appear to be separate
objects, while HS-HR videos indicate they can sediment together as a single
cored cluster (Fig. 6).

The size of the entire aggregate before the impact ranged between ~ 400
280 and $800 \mu\text{m}$, while the core particles ranged between ~ 200 and $500 \mu\text{m}$. The
size of core particles corresponds well to the mode of the coarse population
of ground deposit (samples of trays) for both fallout phases (i.e. $500 \mu\text{m}$ and
 $250 \mu\text{m}$, respectively; red arrows in Figure 5C,E). HS-HR movies were also used
to derive size, terminal velocity and density of aggregates that range between
285 $1.2 - 2.9 \text{ m s}^{-1}$ and $350 - 1000 \text{ kg m}^{-3}$, respectively (see Table 1). Besides the
videos described here in detail, an additional 19 videos of falling aggregates at
Sakurajima from other *independent* explosions between 29th July-2nd August
2013 were also recorded, which indicate that 90% of aggregates were cored
clusters.

The mass fraction of particles $< 90 \mu\text{m}$ in trays (which must have deposited
as shell particles, since no individual fine ash was observed on adhesive tapes
during SEM analysis) is about 4% and 12% for Phase I and Phase II, respec-
tively. The mass fraction of shell particles (i.e. particles $< 90 \mu\text{m}$) in each
aggregate, f_{agg} , is quantified as follows:

$$f_{agg} = \frac{m_{shell,a}}{m_{shell,a} + m_{core,a}} \quad (3)$$

290 where $m_{shell,a}$ ($= \pi \rho_{agg} d_{agg}^3 / 6 - m_{core,a}$) and $m_{core,a}$ ($= \pi \rho_{core} d_{core}^3 / 6$)
are the masses of shell particles and of the core particle in the cored cluster,
respectively. Based on Eq. (3) and given that ρ_{agg} , d_{agg} and d_{core} are known
from HS-HR movies and ρ_{core} is approximated from the water-pycnometer mea-

surements, f_{agg} is found to be on average 10% and 17% for Phase I and Phase
 295 II aggregates, respectively. In other words, the fraction of shell particles within
 each aggregate phase increased from Phase I to II.

We also demonstrate that the wt% of particles $> 90 \mu\text{m}$ in the tray that acted
 as core particles (i.e. involved in the aggregation process) for Phase I is 38%
 and increased to 67% for Phase II fallout, respectively (see F_{tray} in Appendix
 300 A for more details). These ratios (i.e. f_{agg} and F_{tray}) indicate that both the
 amount of shell particles and the rate of aggregation increased from Phase I to
 Phase II.

3.2. Numerical investigations

Figure 7A shows all possible trajectories of Phase I aggregates that could
 305 reach our sampling location within the observed deposition time. It should
 be reminded that the effect of each specific wind profile on sedimentation is
 not important here since we are only interested in the end-members of possible
 release distance and time for each aggregate phase. The resulting position in the
 plume at which particles were released for Phase I aggregates ranges between
 310 250 m and 2300 m from the vent depending on the wind profile. Thus, the 95th
 percentile of Phase I aggregates must have formed within 300 s from eruption
 onset. In contrast, the 95th percentile of Phase II aggregates should have been
 formed within 175 s (Figure 7B).

Based on our Lagrangian model, the maximum distances from the vent at
 315 which the observed Phase I and Phase II aggregates were released are 2300 m
 and 1750 m, respectively. This is compatible with the sedimentation model of
 Bursik et al. (1992), which predicts that aggregates of Phase I (terminal velocity
 of $\sim 3 \text{ m s}^{-1}$) and Phase II (terminal velocity of $\sim 2 \text{ m s}^{-1}$) represent $\sim 30\%$
 and $\sim 60\%$ of the respective initial masses at the corner of the plume ($\sim 900\text{m}$)
 320 at the distances of 2300 m and 1750 m from the vent (Figure 7). Such a model
 has already been validated with a range of eruptive styles and intensities (e.g.
 Bonadonna and Phillips, 2003; Bonadonna and Costa, 2013; Bursik et al., 1992;
 Sparks et al., 1992; Koyaguchi and Ohno, 2001) and predicts the abundance in

the umbrella cloud with distance from vent of a certain object (either individual
325 particle or aggregate) with a given terminal velocity based on the turbulent
sedimentation equation of Martin and Nokes (1988).

3.3. *Simultaneous premature and delayed sedimentation of ash particles*

Our observations show how particle aggregation can simultaneously induce
both premature and delayed sedimentation (i.e. rafting) (Fig. 8). In fact, we
330 have seen how fine ash particles constituting the aggregate shell increase their
terminal velocity up to several orders of magnitude. Nonetheless, the coarse-
ash particles acting as aggregate core ($\sim 200 - 500\mu\text{m}$) decrease their terminal
velocity by 9–33% (Fig. 8). Such decrease in terminal velocity can translate in
an increase of $\sim 10 - 50\%$ in the sedimentation distance.

335 4. Discussion

4.1. *Importance of cored clusters for ash sedimentation*

Coated particles have often been considered not to play an important role in
particle aggregation because they have always been observed as only partially
covered by fine ash (e.g. Sparks et al., 1997; Bonadonna et al., 2002b, 2011;
340 James et al., 2002; Brown et al., 2012). As a result, fine ash has always been
assumed to sediment prematurely mostly as fragile ash clusters and accretionary
lapilli. However, our new observations suggest that ash clusters observed previ-
ously are likely to represent the shell of cored clusters lost at impact with the
sample collector, and that ash clusters and coated particles often represent frag-
345 ments of the same cored cluster (Fig. 1). Thus, fragile cored clusters that are
typically not preserved in the deposit, nor on adhesive tape, play a significant
role in the premature sedimentation of fine ash. In addition, the syn-deposition
of ash clusters and coated particles (that is normally difficult to explain due
to the very different terminal velocities of the two types of aggregates) can
350 be finally reconciled if ash clusters and coated particles are considered to fall
together as part of the same cored cluster. Given the limited number of observa-
tions of cored clusters, the difference between coated particles and cored clusters

is difficult to quantify. Previous studies have described coated particles as only partially covered by fine ash (e.g. Brown et al., 2012), while our results suggest
355 that cored clusters are as fragile as coated particles but they are characterized by a minimum coating that is $\sim 10 - 20$ wt% of the total aggregate.

By reinterpreting the observations of Taddeucci et al. (2011) based on settling velocity measurements, we suggest that the proximal tephra fallout associated with the 2010 eruption of Eyjafjallajökull was similarly dominated by
360 cored clusters. In fact, they reported sedimentation of aggregates with settling velocity mostly $> 1 \text{ m s}^{-1}$, which is indicative of cored-cluster sedimentation based on our observations and on the experiments of James et al. (2002) (also see "VIDEO DR2" in Taddeucci et al., 2011, in which the core particle can be clearly seen). We suggest that most ash clusters described in literature at
365 proximal and medial distances were likely deposited as cored clusters that broke with impact with the ground.

Cored clusters can also better explain the polymodal grainsize distribution of proximal to medial tephra deposits. In fact, grainsize polymodality has often been explained by particle aggregation because of the simultaneous sedimen-
370 tation of individual lapilli and/or coarse-ash particles together with fine ash particles aggregated as ash clusters (e.g. Carey and Sigurdsson, 1982; Brazier et al., 1983). This theory can account for the deposition distance of individual particles, which in some cases matches the deposition distance of aggregate core particles. However, it fails to explain the proximal deposition of aggregates
375 only composed of fine ash, since ash clusters typically have settling velocities $< 1 \text{ m s}^{-1}$ (James et al., 2002). In fact, it is the high settling velocity of coated particles and cored clusters (here $1.2 - 2.9 \text{ m s}^{-1}$ and $\sim 1 - 4.0 \text{ m s}^{-1}$ for 2010 eruption of Eyjafjallajökull; Taddeucci et al., 2011), which can explain the premature fallout of fine ash at proximal to medial distances. This is supported
380 by the fact that the mode of the grain size distribution for coarse-ash particles deposited at our location is very close to the size of core particles measured from the videos (Fig. 5).

It should also be noted that the ability of forming coated particles and cored

clusters in volcanic fallout is dependent on the availability of coarse ash and, thus, decreases with distance from vent. This indicates that strong polymodal grainsize distributions are more likely caused by a dominance of cored clusters and/or coated particles than by a syn deposition of ash clusters and large individual particles. Given that coarse ash concentration decreases with distance from vent, ash clusters become dominant and polymodal grainsize distributions gradually disappear in the deposit (e.g. Bonadonna et al., 2011). Through this transition from dominant fallout of cored clusters to ash clusters, the size of core particles in aggregates gradually decreases up to a point that all particles are comparable in size (i.e. sedimentation of ash clusters).

Besides their high efficiency in removing fine ash from volcanic clouds, the dominant role of cored clusters in the volcanic deposits has also important implications for our understanding and modelling of particle aggregation (i.e. collision and sticking). First, at given concentration and environmental conditions, cored clusters can form faster and more easily than ash clusters. In fact, their collision mechanisms (i.e. ballistic and sedimentation) are several orders of magnitude more efficient than the collision mechanisms that typically control the formation of ash clusters (i.e. Brownian motion and turbulence) (by using models provided in Elimelech et al., 1998; Costa et al., 2010; Pruppacher and Klett, 2012). In addition, theoretical descriptions (Costa et al., 2010) show how the increase of sticking efficiency of fine ash is not sufficient to counterbalance the strong decrease in collision efficiency. As a result, whenever both fine and coarse ash are available, aggregation is more likely to result in cored clusters than ash clusters. Coated particles are also more efficient than ash clusters to form as they simply represent less developed cored clusters (i.e. cored clusters with thinner outer shell).

4.2. *Timing of aggregation*

Given that Phase II aggregates are characterized by thicker shells and most likely needed more time to form than Phase I aggregates, it can be concluded that cored clusters can be formed within 175 s after the onset of the eruption. As

a comparison, accretionary lapilli and frozen accretionary pellets were reported
 415 to be formed within 300 and 600 seconds after the onset of 1990 eruption of
 Sakurajima volcano (Japan) and the March 2009 eruption of Redoubt volcano
 (Alaska), respectively (Gilbert and Lane, 1994; Brown et al., 2012). It cannot
 be excluded that in our case aggregation continued to occur beyond 175 s simply
 because our observation is just based on a single proximal location in the field.
 420 Nonetheless, the presence of unexpected large particles observed in distal regions
 (e.g. Sorem, 1982) show that volcanic aggregates form in the vicinity of the
 vent, where the ash concentration is high and large particles are still present
 in the cloud. However, to be able to draw general and quantitative conclusions
 on aggregation timing, further field studies of volcanic eruptions of different
 425 magnitudes and environmental conditions need to be carried out.

4.3. Temporal and spatial evolution of coated particles

Aggregation processes associated with the selected explosion evolved in time
 with significant differences in observed aggregates. First, Phase I aggregates
 have a higher settling velocity compared to those of Phase II. Second, aggre-
 430 gates of Phase I are characterized by larger core particles than those of Phase
 II (Figure 5 and Table 2). Third, shells of Phase I aggregates are thinner than
 shells of Phase II compared to their core particle diameter. Finally, the wt%
 of coarse particles that sedimented individually (and not as core particles in-
 side aggregates) decreased from Phase I to Phase II. Given that the terminal
 435 velocity of aggregates decreased with time at our location, a similar variation
 is expected to occur also in space along the dispersal axis. As a result, the
 expected trends with distance from the vent are i) an increase of the ratio of
 the wt% of aggregates to individual particles, ii) a decrease of the size of core
 particles and iii) an increase of the wt% of fine ash in aggregates. The fine-ash
 440 population of fallout deposits is then expected to increase with distance from
 vent with the associated grainsize distribution showing a weak polymodality
 in proximal region, a stronger polymodality in medial region and unimodality
 in distal region where sedimentation is expected to be characterized mostly by

ash clusters (Schumacher, 1994; Bonadonna et al., 2011). However, given that
445 we sampled only a single location along the dispersal axis for a relatively short
duration, these need to be confirmed with further field studies.

4.4. Importance of rafting on coarse-ash sedimentation

We have provided the first field-based evidence for the rafting effect, which
has only been discussed theoretically and its occurrence considered not to be
450 likely (Lane et al., 1993; James et al., 2002). Our measurements indicate that
the terminal velocity of core particles is reduced up to 9 – 33% due to aggre-
gation. The decrease in the settling velocity due to rafting effect allows for
core particles to deposit at larger distances than expected. This contradicts the
common idea that aggregates result uniquely in higher terminal velocities than
455 individual constituent particles and always induce a premature sedimentation of
volcanic particles (Lane et al., 1993; Rose and Durant, 2011; Brown et al., 2012).
As an example, rafting can potentially explain the sedimentation of 90 μm par-
ticles at about 1300 km from the vent during the 2010 Eyjafjallajökull eruption
(Stevenson et al., 2015), which is double the sedimentation distance expected
460 for this particle size (Beckett et al., 2015).

Rafting is caused by the non-linear relation between the aggregate drag
coefficient and the increase in the aggregate mass and size, which can increase
or decrease the aggregate settling velocity. Thus, rafting is really dependent
on the density, size and even shape of aggregates. Given that the terminal
465 velocity measured from HS-HR videos and the derived aggregate densities, size
and shape of core particles are all associated with some degree of uncertainty, in
order to draw general conclusions more field and analytical investigations should
be preformed that are beyond the scope of this paper and should be addressed
in future studies. In particular, future field studies should be performed with
470 even higher camera resolution (i.e. smaller pixel size), lower exposure time (to
capture better the aggregate/core-particle shape) and at multiple points along
the dispersal axis. Such studies should be able to provide detailed information on
the shape, density and structure of aggregates, and capture a complete picture

of aggregation evolution in time and space.

475 5. Conclusions

Although our study is based on the field and numerical investigation of a single Vulcanian explosion at Sakurajima volcano, important aspects of volcanic aggregates have been revealed. In particular, our new HS-HR observations helped reconcile the syn-deposition of ash clusters and coated particles (characterized by different terminal velocities) based on the introduction of a new category of aggregate that has never been observed in the deposit or adhesive tape due to its very low preservation potential, namely cored clusters (PC3). In fact, the abundance of coated particles and bimodal grain-size distributions in proximal regions reflect the sedimentation of cored clusters composed of a core particle ($\sim 200 - 500 \mu\text{m}$) enclosed in a thick shell of finer particles ($< 90 \mu\text{m}$). By reinterpreting published data, this was found to be also valid for the 2010 eruption of Eyjafjallajökull. We suspect that most aggregates sedimented up to medial regions and classified in literature as ash clusters and coated particles were likely sedimented together as cored clusters. Based on our field and numerical investigations the following conclusions can be drawn:

- By using an in-situ HS-HR imaging technique, a new category of aggregates, namely cored clusters (PC3), is introduced. Cored clusters found to be comprised of a large core particle $> 90 \mu\text{m}$ covered by a thick shell of finer particles $< 90 \mu\text{m}$.
- Post-processing of the field data showed that the shell of cored clusters comprised $\sim 10-17 \text{ wt\%}$ of the aggregates sedimented during the observed explosion (f_{agg} in Eq. 3). Such a significant portion of shell particles indicates the importance of cored clusters in enhancing sedimentation of fine ash as opposed to coated particle, which are considered not to play an important role in the aggregation processes. In fact, observed coated particles in the literature can simply represent core particles of cored clusters broken at impact with the ground (or adhesive tape).

- 505 • Various evidences, including high terminal velocity of aggregates observed in this study and Taddeucci et al. (2011), and strong polymodal grainsize distribution of proximal to medial tephra deposits, suggest that where coarse and fine ash are both present, cored clusters are the most likely aggregate to deposit. Cored clusters are also most likely to form than ash clusters due to higher collision efficiency of particles, and, therefore, can be more efficient at scavenging fine ash.
- 510 • Cored clusters with relatively thick shells can also better explain the fine ash deposition and grainsize polymodality observed in proximal–medial regions, which are not compatible with slowly falling ash clusters.
- 515 • We have introduced a new methodology (Appendix A) that allows for the calculation of the mass fraction of core particles involved in aggregation from the deposit. Such a methodology helps characterize the variation of aggregation conditions in time. As an example, for our case study, the wt% of particles that acted as aggregate cores ($> 90 \mu\text{m}$) increases from 38% for Phase I to 67% for Phase II.
- 520 • By conducting numerical inversions of field observations, it was also found that cored clusters can be formed very rapidly and within order of seconds after the eruption onset.
- 525 • Finally, the first field-based evidence for the rafting effect is presented and it was shown that due to the rafting, aggregates can simultaneously promote premature fallout of fine ash and delay fallout of coarse ash (acting as aggregate core). This can potentially explain the sedimentation of coarse ash at larger distance from the volcano than expected (e.g. Sorem, 1982; Stevenson et al., 2015).

7. Acknowledgments

The research leading to these results has received funding from the Swiss National Science Foundation (SNSF, Grant No. 200020-125024) and the People Programme (Marie Curie Actions) of the European Union's Seventh Framework Programme (FP7/2007-2013) under the project NEMOH, REA grant agreement
535 n 289976. Authors are grateful to M. Ichiara, A. Kurokawa, L. Dominguez, L. Pioli, I. Manzella and F. Brogi for their enormous support in the field, to M. Pistolesi and R. Cioni for their insights into SEM analysis and to Mr. Frederic Arlaud for his help to design and construct the field setup. The authors are also
540 grateful to M. Mangan and R. Cioni for their constructive reviews that helped us to improve the manuscript.

References

- Bagheri, G., Bonadonna, C., 2016. On the drag of freely falling non-spherical particles. Powder Technology 301, 526–544.
545 URL: <http://dx.doi.org/10.1016/j.powtec.2016.06.015><http://linkinghub.elsevier.com/retrieve/pii/S0032591016303539>,
doi:10.1016/j.powtec.2016.06.015.
- Bagheri, G., Bonadonna, C., Manzella, I., Vonlanthen, P., 2015. On the characterization of size and shape of irregular particles. Powder Technology 270,
550 141–153.
- Bagheri, G.H., Bonadonna, C., Manzella, I., Pontelandolfo, P., Haas, P., 2013. Dedicated vertical wind tunnel for the study of sedimentation of non-spherical particles. Review of Scientific Instruments 84, 054501.

- Beckett, F.M., Witham, C., Hort, M., Stevenson, J., Bonadonna, C., Millington,
555 S., 2015. Sensitivity of dispersion model forecasts of volcanic ash clouds to
the physical characteristics of the particles. *Journal of Geophysical Research:
Atmospheres*, n/a–n/a/doi:10.1002/2015JD023609.
- Blong, R., 1984. *Volcanic hazards. A sourcebook on the effects of eruptions.*
Academic Press, Inc., Orlando, FL.
- 560 Bonadonna, C., Costa, A., 2013. Modeling of tephra sedimentation from vol-
canic plumes, in: Fagents, S.A., Gregg, T.K.P., Lopes, R.M.C. (Eds.), *Model-
ing volcanic processes: The physics and mathematics of volcanism.* Cambridge
University Press, pp. 173–202.
- Bonadonna, C., Genco, R., Gouhier, M., Pistolesi, M., Cioni, R., Alfano, F.,
565 Hoskuldsson, A., Ripepe, M., 2011. Tephra sedimentation during the 2010
Eyjafjallajökull eruption (Iceland) from deposit, radar, and satellite observa-
tions. *Journal of Geophysical Research* 116, B12202.
- Bonadonna, C., Macedonio, G., Sparks, R.S.J., 2002a. Numerical modelling
of tephra fallout associated with dome collapses and Vulcanian explosions:
570 application to hazard assessment on Montserrat. *Geological Society, London,
Memoirs* 21, 517–537.
- Bonadonna, C., Mayberry, G.C., Calder, E.S., Sparks, R.S.J., Choux, C., Jack-
son, P., Lejeune, A.M., Loughlin, S.C., Norton, G.E., Rose, W.I., Ryan, G.,
Young, S.R., 2002b. Tephra fallout in the eruption of Soufriere Hills Volcano,
575 Montserrat. *Geological Society, London, Memoirs* 21, 483–516.
- Bonadonna, C., Phillips, J.C., 2003. Sedimentation from strong volcanic plumes.
Journal of Geophysical Research: Solid Earth 108, n/a–n/a.
- Brazier, S., Sparks, R.S.J., Carey, S.N., Sigurdsson, H., Westgate, J.A., 1983.
Bimodal grain size distribution and secondary thickening in air-fall ash layers.
580 *Nature* 301, 115–119.

- Brown, R., Bonadonna, C., Durant, A., 2012. A review of volcanic ash aggregation. *Physics and Chemistry of the Earth, Parts A/B/C* 45-46, 65–78.
- Bursik, M.I., Sparks, R.S.J., Gilbert, J.S., Carey, S.N., 1992. Sedimentation of tephra by volcanic plumes: I. Theory and its comparison with a study of the Fogo A plinian deposit, Sao Miguel (Azores). *Bulletin of Volcanology* 54, 329–344.
- Carey, S., Sigurdsson, H., 1982. Influence of particle aggregation on deposition of distal tephra from the May 18, 1980, eruption of Mount St. Helens volcano. *Journal of geophysical research* 87, 7061–7072.
- Chhabra, R.P., 2006. Bubbles, drops, and particles in non-Newtonian fluids. CRC press.
- Clift, R., Gauvin, W.H., 1971. Motion of entrained particles in gas streams. *The Canadian Journal of Chemical Engineering* 49, 439–448.
- Costa, A., Folch, A., Macedonio, G., 2010. A model for wet aggregation of ash particles in volcanic plumes and clouds: 1. Theoretical formulation. *Journal of Geophysical Research* 115, B09201.
- Durant, A.J., 2015. Toward a realistic formulation of fine-ash lifetime in volcanic clouds. *Geology* 43, 271–272.
- Elimelech, M., Gregory, J., Jia, X., Williams, R., 1998. Particle deposition and aggregation: measurement, modelling and simulation. Butterworth-Heinemann.
- Folch, A., Costa, A., Durant, A., Macedonio, G., 2010. A model for wet aggregation of ash particles in volcanic plumes and clouds: 2. Model application. *Journal of Geophysical Research* 115, B09202. doi:10.1029/2009JB007176.
- Gilbert, J., Lane, S., Sparks, R., Koyaguchi, T., 1991. Charge measurements on particle fallout from a volcanic plume. *Nature* 349, 598 – 600.

- Gilbert, J.S., Lane, S.J., 1994. The origin of accretionary lapilli. *Bulletin of Volcanology* 56, 398–411.
- Guffanti, M., Casadevall, T.J., Budding, K., 2010. Encounters of Aircraft with
610 Volcanic Ash Clouds: A Compilation of Known Incidents, 1953–2009. Technical Report.
- Hoper, V., Grant, A., 1948. The Influence of a Horizontal Wall on the Motion of a Falling Oil Drop. *Australian Journal of Chemistry* 1, 28.
- Ikhwan, M., 2005. Investigation of Flow and Pressure Characteristics around
615 Pyramidal Buildings. Ph.D. thesis. Karlsruher Institut für Technologie.
- James, M.R., Gilbert, J.S., Lane, S.J., 2002. Experimental investigation of volcanic particle aggregation in the absence of a liquid phase. *Journal of Geophysical Research* 107, 2191.
- Koyaguchi, T., Ohno, M., 2001. Reconstruction of eruption column dynamics
620 on the basis of grain size of tephra fall deposits: 1. methods. *Journal of Geophysical Research: Solid Earth* 106, 6499–6512. doi:10.1029/2000JB900426.
- Lane, S.J., Gilbert, J.S., Hilton, M., 1993. The aerodynamic behaviour of volcanic aggregates. *Bulletin of Volcanology* 55, 481–488.
- Martin, D., Nokes, R., 1988. Crystal settling in a vigorously converting magma
625 chamber. *Nature* 332, 534–536.
- Mueller, S.B., Kueppers, U., Ayris, P.M., Jacob, M., Dingwell, D.B., 2016. Experimental volcanic ash aggregation: Internal structuring of accretionary lapilli and the role of liquid bonding. *Earth and Planetary Science Letters* 433, 232–240. doi:10.1016/j.epsl.2015.11.007.
- Pruppacher, H.R., Klett, J.D., 2012. *Microphysics of Clouds and Precipitation*:
630 Reprinted 1980. Springer Science & Business Media.

- Reimer, T.O., 1983. Accretionary Lapilli in Volcanic Ash Falls: Physical Factors Governing Their Formation. Springer Berlin Heidelberg, Berlin, Heidelberg. pp. 56–68. doi:10.1007/978-3-642-68869-0_6.
- 635 Rose, W.I., Durant, a.J., 2011. Fate of volcanic ash: Aggregation and fallout. *Geology* 39, 895–896.
- Schumacher, R., 1994. A reappraisal of Mount St. Helens’ ash clusters-depositional model from experimental observation. *Journal of volcanology and geothermal research* 59, 253–260.
- 640 Schumacher, R., Schmincke, H., 1991. Internal structure and occurrence of accretionary lapillia case study at Laacher See Volcano. *Bulletin of Volcanology* 53, 612–634.
- Schumacher, R., Schmincke, H.U., 1995. Models for the origin of accretionary lapilli. *Bulletin of Volcanology* 56, 626–639.
- 645 Sorem, R.K., 1982. Volcanic ash clusters: Tephra rafts and scavengers. *Journal of Volcanology and Geothermal Research* 13, 63–71.
- Sparks, R.S.J., Bursik, M.I., Ablay, G.J., Thomas, R.M.E., Carey, S.N., 1992. Sedimentation of tephra by volcanic plumes. part 2: controls on thickness and grain-size variations of tephra fall deposits. *Bulletin of Volcanology* 54, 685–695. doi:10.1007/BF00430779.
- 650 Sparks, R.S.J., Burski, M.I., Carey, S.N., Gilbert, J.S., Glaze, L.S., Sigurdsson, H., Woods, A.W., 1997. Volcanic plumes. 1st editio ed., John Wiley & Sons.
- Stevenson, J.a., Millington, S.C., Beckett, F.M., Swindles, G.T., Thordarson, T., 2015. Big grains go far: reconciling tephrochronology with atmospheric measurements of volcanic ash. *Atmospheric Measurement Techniques Discussions* 8, 65–120.
- 655 Taddeucci, J., Scarlato, P., Montanaro, C., Cimarelli, C., Del Bello, E., Freda, C., Andronico, D., Gudmundsson, M.T., Dingwell, D.B., 2011. Aggregation-

- dominated ash settling from the Eyjafjallajökull volcanic cloud illuminated
660 by field and laboratory high-speed imaging. *Geology* 39, 891–894.
- Telling, J., Dufek, J., Shaikh, A., 2013. Ash aggregation in explosive volcanic
eruptions. *Geophysical Research Letters* 40, 2355–2360.
- Thordarson, T., 2004. Accretionary-lapilli-bearing pyroclastic rocks at odp leg
192 site 1184: a record of subaerial phreatomagmatic eruptions on the ontong
665 java plateau. *Geological Society, London, Special Publications* 229, 275–306.
doi:10.1144/GSL.SP.2004.229.01.16.
- Van Eaton, A.R., Mastin, L.G., Herzog, M., Schwaiger, H.F., Schneider, D.J.,
Wallace, K.L., Clarke, A.B., 2015. Hail formation triggers rapid ash aggre-
gation in volcanic plumes. *Nature Communications* 6, 7860. doi:10.1038/
670 ncomms8860.
- Van Eaton, A.R., Wilson, C.J., 2013. The nature, origins and distribution of
ash aggregates in a large-scale wet eruption deposit: Oruanui, New Zealand.
Journal of Volcanology and Geothermal Research 250, 129–154.
- Veitch, G., Woods, A., 2001. Particle aggregation in volcanic eruption columns.
675 *Journal of Geophysical Research: Solid Earth* 106, 26425–26441.

Tables

Table 1: Characteristics of observed aggregates (i.e. cored clusters) and core particles.

Fallout Phase ^a	Sed. Time	Aggregates				Core Particles	
		Diameter	Density	Velocity	Porosity	Diameter	Density
		[μm]	[kg m^{-3}]	[m s^{-1}]	%	[μm]	[kg m^{-3}]
I	18:02 – 18:07	718 – 807	806 – 1009	2.7 – 2.9	60 – 68	500 – 525	2500 – 2700
II	18:07 – 18:12	440 – 630	357 – 864	1.2 – 1.8	67 – 83	200 – 330 ^b	2500 – 2700

^a Phase I and II indicate aggregates sedimented during the first and second sampling interval, respectively.

^b Special case of Phase II aggregates for which the core consisted of three particles with a minimum diameter of $\sim 200 \mu\text{m}$

Figures

Figure 1: Schematics and images of various types of Particle Clusters' (PC) family including ash clusters (PC1), coated particles (PC2) (following the notation of Brown et al., 2012) and the newly introduced category of cored clusters (PC3). Images of PC1 and PC2 are from Eyjafjallajökull 2010 eruption (Bonadonna et al., 2011), while images of PC3 are obtained from a HS-HR video of a Sakurajima 2013 explosion (this work); also see Fig. 5A and supplementary videos for more details on PC3.

Figure 2: (A) Map of Sakurajima, Japan, showing the location of the vent and our ground-based observation site. (B) Photo of the Vulcanian explosions occurred at 17:47 Japanese Standard Time (JST) on the 3rd of August 2013. The * symbol on the map shows the location where the photo was taken.

Figure 3: Sketch of the setup used in the field.

Figure 4: Logarithmic plot of particle diameter versus density measured with the water pycnometer for samples collected in the trays.

Figure 5: Selection of observed aggregates. A: Image sequences of an HS-HR movie recorded for a Phase I aggregate (time difference between each sequence is 2.5 ms). The HS-HR video shows that the aggregate consists of a large core that rebounded the adhesive tape upon impact. B: SEM image of shell particles shown in Figure 5A (deposited at 18:05 JST). Particle size distributions in weight% obtained from C: laser diffraction measurements for the ash accumulated in the tray during Phase I fallout (18:00 – 18:07 JST). D: SEM analysis of Figure 5B (Phase I aggregate). E: laser diffraction measurements for the ash accumulated in the tray during Phase II fallout (18:07 – 18:12 JST). F: SEM analysis of a Phase II aggregate deposited at 18:09 JST. Red arrows show the size of core particle derived from HS-HR videos.

Figure 6: SEM images of aggregates collected on thin-glass supports during our field investigations in Sakurajima. (a) Coated particles in the nomenclature of Brown et al. (2012), that is here interpreted as the core particle of a cored cluster; (b) ash cluster in the nomenclature of Brown et al. (2012), that is here interpreted as the shell of a cored cluster left on adhesive tape after impact.

Figure 7: Lagrangian simulations of aggregate trajectories based on four wind profiles. Aggregates are released from the plume margins and cloud base, but only those that sedimented at our location (3.7 ± 0.25 km from vent) within the observed deposition time are considered. In order to account for the uncertainty in the wind, four different wind profiles are used based on various sources and assumptions (see Methods). A: Phase I aggregates. B: Phase II aggregates. Normalized mass concentration of aggregates in the cloud (M/M_0 , with M the total mass for each aggregate phase and M_0 the initial mass at plume corner for each aggregate phase) are calculated for both fallout phases based on the sedimentation model (Bursik et al., 1992); aggregate velocities are calculated based on atmospheric properties at the cloud height (3 and 2 m s^{-1} for Phase I and II, respectively). Red vertical dashed lines indicate the maximum distance of release.

Figure 8: Terminal velocity versus diameter of cored clusters and their constituent particles. The terminal velocity of cored clusters are measured by analyzing videos, whereas the terminal velocity of fine ash and core particles are calculated numerically assuming spherical shapes for particles. Curves show the variation of terminal velocity for spheres with different densities; arrows indicate the direction of change in the terminal velocity of fine ash and core-particles as a result of aggregation. Note that aggregate IIE is composed of three core-particles.

Appendix A. Analytical derivation of F_{tray}

In this section we illustrate our methodology to assess the mass fraction
of fallout that is involved in the aggregation process with respect to the total
mass of fallout sedimenting at a given location (and collected in trays). In
our study, such a mass fraction is identified by the mass fraction of particles
 $> 90 \mu\text{m}$ sedimented as cluster core particles in each tray with respect to the
mass fraction of particles $> 90 \mu\text{m}$ that sedimented as individual particles. Here,
we present the derivation of an analytical method to obtain this mass fraction
as a function of measurable variables. Main assumptions are:

1. The threshold of $90 \mu\text{m}$ was chosen because particles $< 90 \mu\text{m}$ were found
to all sediment as the shell of the cored clusters. However, such a threshold
can be adapted to different conditions and studies.
2. All particles with size $< 90 \mu\text{m}$ in the trays are assumed to be sedimented
as a part of cored clusters' shell. This is supported by the fact that no
individual fine ash was observed on adhesive tapes.
3. All aggregates sedimented in the trays are assumed to have the same
average features as those observed with HS-HR videos.

Two sets of parameters are used in our methodology, which are parameters
related to i) trays and ii) falling aggregates (see also Fig. A.1). Parameters
related to trays include:

- $m_{core,t}$: mass of particles $> 90 \mu\text{m}$ that fell inside the tray as the cores of
the cored clusters (not known).
- $m_{indv.,t}$: mass of particles $> 90 \mu\text{m}$ that fell inside the tray as individual
particles and were not involved in the aggregation process (not known).
- $m_{shell,t}$: mass of particles $< 90 \mu\text{m}$ that fell inside the tray as the shell of
cored clusters that can be calculated as (see assumption no. 2):

$$m_{shell,t} = m_{total,t} \sum_{i=1}^n \rho_{p,i} f_{v,i} \quad \text{for } d_{p,i} < 90 \mu\text{m} \quad (\text{A.1})$$

Figure A.1: Simplified sketch illustrating the various parameters used in the methodology.

where $d_{p,i}$ is the diameter of the i -th bin of the grainsize histogram (see Figure 5C,E), $\rho_{p,i}$ is the density of a particle with diameter of $d_{p,i}$ approximated from water pycnometer measurements (see Fig. 4), $f_{v,i}$ is the volume fraction of particles in the i -th bin of the grainsize histogram, n is the number of bins in the grainsize histogram and $m_{total,t}$ is defined below.

- $m_{total,t} = m_{core,t} + m_{indv.,t} + m_{shell,t}$: mass of all particles that fell inside the tray (directly measured with a digital balance).
- f_{tray} : mass fraction of shell particles collected in each tray (i.e. the mass fraction of particles with diameter $< 90 \mu\text{m}$):

$$f_{tray} = \frac{m_{shell,t}}{m_{total,t}} = \frac{m_{shell,t}}{m_{core,t} + m_{indv.,t} + m_{shell,t}} \quad (\text{A.2})$$

- F_{tray} : mass fraction of particles that fell as cluster cores inside the tray

with respect to the whole amount of particles $> 90 \mu\text{m}$:

$$F_{tray} \equiv \frac{m_{core,t}}{m_{core,t} + m_{indv.,t}} \quad (\text{A.3})$$

710 F_{tray} is the parameter of interest that needs to be determined, as it can be used as a proxy for the mass fraction of aggregated particles.

Parameters related to individual cored clusters include (here we used average values for each fallout phase):

- $m_{core,a}$: mass of the particle that fell as core of a cored cluster (measured by HS-HR videos and water pycnometer, see section 2.1).
- 715 • $m_{total,a}$: total mass of individual cored clusters (measured by HS-HR videos: $= \rho_{agg} (\pi d_{agg}^3/6)$, see section 2.1).
- $m_{shell,a} = m_{total,a} - m_{core,a}$: mass of particles that fell as part of the shell of each cored cluster.
- 720 • $f_{agg} = m_{shell,a}/(m_{shell,a} + m_{core,a})$: as already defined in Eq. (3), is the mass fraction of shell particles (i.e. particles $< 90 \mu\text{m}$) in each aggregate.

Here we propose a methodology to extract the value of F_{tray} defined in Eq.(A.3) based on measurable variables, namely f_{tray} and f_{agg} . As mentioned in section 3.1, f_{agg} is between 10% and 17% and following Eq. (A.2), f_{tray} is found to be between 4% and 12% for Phase I and II, respectively.

Based on the third assumption described above, we can deduce that the mass ratio between shell particles, $m_{shell,a}$, and core particles, $m_{core,a}$, within a single cored cluster (or average of that for aggregates of a given phase) should be equal to the mass ratio between all shell particles, $m_{shell,t}$, and all core particles, $m_{core,t}$, deposited in the tray:

$$\frac{m_{shell,a}}{m_{core,a}} = \frac{m_{shell,t}}{m_{core,t}} \quad (\text{A.4})$$

We also need to rewrite Eq. (3), to define f_{agg} as:

$$F_{agg} = \frac{m_{shell,a}}{m_{core,a} + m_{shell,a}} \quad (\text{A.5})$$

With little algebra we can show that:

$$\frac{m_{shell,a}}{m_{core,a}} = \frac{f_{agg}}{1 - f_{agg}} \quad (\text{A.6})$$

Since f_{agg} is different for each aggregate, an average value for each phase can be used (i.e. 10% for Phase I and 17% for Phase II aggregates).

By combining Eqs. (A.4) and (A.6) we can conclude that:

$$m_{shell,t} = \frac{f_{agg}}{1 - f_{agg}} m_{core,t} \quad (\text{A.7})$$

For simplicity of notation we define

$$\lambda = \frac{f_{agg}}{1 - f_{agg}} \quad (\text{A.8})$$

By combining Eqs. (A.2) and (A.7) we obtain:

$$f_{tray} = \frac{\lambda m_{core,t}}{\lambda m_{core,t} + m_{core,t} + m_{indv.,t}} \quad (\text{A.9})$$

Inverting this equation results in:

$$\frac{1}{f_{tray}} = \frac{\lambda m_{core,t}}{\lambda m_{core,t}} + \frac{m_{core,t} + m_{indv.,t}}{\lambda m_{core,t}} = 1 + \frac{1}{\lambda F_{tray}} \quad (\text{A.10})$$

As a result the total fraction of aggregates, F_{tray} , defined as the ratio of coarse-ash particles (i.e. $\text{ash} > 90 \mu\text{m}$) that were involved in the aggregation process to the whole coarse-ash population can be finally derived from Eq. (A.10):

$$F_{tray} \equiv \frac{m_{core,t}}{m_{core,t} + m_{indv.,t}} = \frac{f_{tray} \times (1 - f_{agg})}{f_{agg} \times (1 - f_{tray})} \quad (\text{A.11})$$

From Eq. (A.11) we can finally calculate F_{tray} , which is 38 wt% during Phase I and increases to 67 wt% for Phase II fallout. It should be noted that Eq. A.11

735 is valid for both coated-particle and cored-cluster dominated fallout.