A Review of Volcanic Ash Aggregation

R.J. Brown[1], C. Bonadonna[2], A.J. Durant[3,4]

[1]Department of Earth Sciences, Durham University, Science Labs, Durham, DH1 3LE, UK.

[2]Section des sciences de la Terre et de l'environnement, Université de Genève, Rue des Maraîchers, 13, CH-1205, Genève, Switzerland

[3]Norwegian Institute for Air Research, Atmospheric and Climate Department, Instituttveien 18, P.O. Box 100, NO-2027 Kjeller, Norway

[4]Geological and Mining Engineering and Sciences, Michigan Technological University, 1400 Townsend Drive, Houghton, MI 49931, USA

Corresponding author: richard.brown3@durham.ac.uk

Abstract

Most volcanic ash particles with diameters < 63 μm settle from eruption clouds as particle aggregates that cumulatively have larger sizes, lower densities, and higher terminal fall velocities than individual constituent particles. Particle aggregation reduces the atmospheric residence time of fine ash, which results in a proportional increase in fine ash fallout within 10s km to 100s km from the volcano and a reduction in airborne fine ash mass concentrations 1000s km from the volcano. Aggregate characteristics vary with distance from the volcano: proximal aggregates are typically larger (up to cm size) with concentric structures, while distal aggregates are typically smaller (sub-millimetre size). Particles comprising ash aggregates are bound through hydro-bonds (liquid and ice water) and electrostatic forces, and the rate of particle aggregation correlates with cloud liquid water availability. Eruption source parameters (including initial particle size distribution, erupted mass, eruption column height, cloud water content and temperature) and the eruption plume temperature lapse rate, coupled with the environmental parameters, determines the type and spatiotemporal distribution of aggregates. Field studies, lab experiments and modelling investigations have already provided important insights on the process of particle aggregation. However, new integrated observations that combine remote sensing studies of

ash clouds with field measurement and sampling, and lab experiments are required to fill current gaps in knowledge surrounding the theory of ash aggregate formation.

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1. Introduction

Explosive volcanic eruptions generate large amounts (>50 % of total erupted mass) of fine ash particles (here defined as particles with diameter < 63 μm) which are dispersed into the atmosphere by buoyant plumes above volcanic vents and pyroclastic density currents (PDCs; Carey and Sigurdsson, 1982; Hildreth and Drake, 1992; Rose and Durant, 2009). Most volcanic ash finer than 125 μm settles out of the atmosphere as particle aggregates that have higher settling velocities than individual constituent particles (Carey and Sigurdsson, 1982; Sorem, 1982; Lane et al., 1993). While aggregation exerts a first order control on the dispersal of fine ash within eruption clouds, the physical and chemical processes involved are not completely understood despite significant progress over the past twenty years (Schumacher and Schmincke, 1995; Gilbert and Lane, 1994; James et al., 2002, 2003; Textor et al., 2006a and b; Durant et al., 2009; Costa et al., 2010). Fine airborne particles adhere to each other as a result of electrostatic attraction, moist adhesion between particles (e.g., Sorem, 1982; Gilbert and Lane, 1994; Schumacher and Schmincke, 1991, 1995; James et al., 2002) and hydrometeor formation (e.g., Veitch and Woods, 2001; Textor et al., 2006a; Durant et al., 2009). The atmospheric residence time of fine ash determines the hazard to aviation (Casadevall, 1994), and ash fallout impacts local environments and infrastructure (Stewart et al., 2006; Spence et al., 2005; Wardman et al., 2011) and may present a health hazard over an extended period of exposure (Horwell and Baxter, 2006).

Over the past few decades, models of varying complexity have been developed for the dispersal and sedimentation of volcanic particles. These models include analytical solutions, used widely for investigations of particle sedimentation and for hazard assessments (e.g., Armienti et al., 1988; Bonadonna et al., 2005a; Bursik et al., 1992a and b; Connor et al., 2001; Connor and Connor, 2006; Glaze and Self, 1991; Hurst and Turner, 1999;

Koyaguchi and Ohno, 2001a; Macedonio et al., 2005; Suzuki, 1983), and numerical models for real-time forecast of plume evolution and sedimentation (e.g., Barsotti and Neri, 2008; Barsotti et al., 2008; Costa et al., 2006; Searcy et al. 1998). Both types have been validated with field data and are now used regularly for both these purposes. However, the majority of these models do not account for ash aggregation and as a consequence tend to underestimate proximal fallout and overestimate ash concentrations in the atmosphere far from source, in particular in case of ash-rich volcanic plumes.

This paper reviews the current understanding of ash aggregation, summarizes observations of aggregate fallout, the structure and morphology of ash aggregates, and reviews the effects of aggregation on the dispersal of tephra. We draw together observations of eruptions, field studies of deposits, experimental studies and numerical modelling. Observations of recent eruptions indicate that the type of aggregate falling from ash clouds changes with distance from the volcano (Rosenbaum and Waitt, 1981; Hobbs et al., 1981; Sorem, 1982): proximal aggregates are larger and can contain water on deposition (liquid or frozen); distal aggregates are much smaller, fragile, and often reach the surface without direct evidence for the involvement of water in the process (we loosely use the term proximal for regions within the plume corner, i.e. <15km depending on plume height; Bonadonna and Phillips, 2003, and distal for regions beyond the plume corner). We support improved observation and documentation of ash aggregates both during eruptions and within deposits in order to advance knowledge of this important topic.

1.1 A note on ash aggregate terminology

Ash aggregates present a range of sizes, textures and shapes, from fragile sub-millimetre-size clusters of ash to centimeter-size concentric-laminated aggregates displaying variously sharp and graded laminations (e.g., Moore et al. 1966; Fisher and Schmincke 1984; Reimer, 1983; Scolamacchia et al., 2005). An early line of studies on particle aggregation focused on proximal aggregates often associated with phreatomagmatic activity (e.g., Lorenz 1974; Rosi 1992; Gilbert and Lane, 1994). Later studies of aggregation processes brought to light the complexities of particle aggregation and the greater variety of particle aggregates from other types of eruptions, and also included more focus on laboratory study (e.g., Sorem 1982; James et al. 2002; James et al. 2003; Durant et al. 2009). As a consequence,

aggregation terminology has evolved and a number of different classification schemes have been proposed (Reimer, 1983; Schumacher and Schmincke, 1991; Thordarson, 2004; Brown et al., 2010). No classification scheme has yet been widely adopted and, over the past twenty years, the term 'accretionary lapilli' has been used to describe unstructured aggregates (e.g., Hayakawa, 1990; Rosi, 1992; Sisson, 1995; Watanabe, 1999; Trusdell et al., 2005), multiple concentric-laminated aggregates (e.g., Cole and Scarpati, 1993; Junqueira-Brod et al., 2005; Edgar et al., 2007), aggregates with a single fine-grained coating around a massive ash core (Branney, 1991) and ash-coated lithic clasts (Bednarz and Schmincke, 1990; Palladino et al., 2001). There are other examples in the literature where the term accretionary lapilli is used without an accompanying description of the aggregates. Usage of terms is inconsistent and confusion exists particularly around the term 'accretionary lapilli', which includes all lapilli-sized ash aggregates, but not aggregates smaller than 2 mm. Consistent use of terminology is important for clear communication of ideas. In recognition that the term 'lapilli' is a particle size denominator and that many aggregates commonly referred to as 'accretionary lapilli' are < 2 mm, we propose to replace this term with 'accretionary pellet', which avoids particle size connotations. Accretionary pellets may be divided into three subcategories (AP1, AP2 and AP3) based only on internal structure, which avoids any implications regarding formation mechanisms. We define a second group of aggregates called particle clusters, which include 'ash clusters' (PC1; e.g., Sorem, 1982) and coated particles (PC2). Revised definitions for aggregate types are provided in Table 1 and illustrated in Figure 1.

2. Evidence for aggregation of volcanic particles

2.1. Visual observations

Ash aggregates have been observed falling during many historic explosive eruptions (Table 1; see examples in Fig. 1). Fallout of fine ash >100s km downwind appears exclusively dominated by millimetre-scale loosely-bound ash clusters (PC1) that rarely survive impact, whereas aggregate fallout from the column may include ash cluster formation, but is dominated by denser, typically spherical or subspherical aggregates (e.g., AP1, AP2 and AP3). As a result of aggregation, many distal ash fall deposits are poorly sorted and exhibit

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polymodal particle size distributions (e.g., Varekamp et al., 1984; Carey and Sigurdsson, 1982; Scott and McGimsey, 1994; Watt et al., 2009; Durant et al. 2009).

There are many observations of both liquid and frozen ash-rich hydrometeors formed in volcanic clouds (Williams and McNutt, 2004; Sparks et al., 1997; Table 2). Liquid and frozen water provides the binding mechanism to hold ash particles together in aggregates. The growth of ice on ash in volcanic clouds also increases particle size and mass, which in turn increases particle terminal fall velocity and enhances collision rate and sedimentation. Observations during the 18 May 1980 eruption of Mount St. Helens, USA, indicate a close association between the formation of volcanogenic hydrometeors (rain, graupel/hailstones and snow) and fine ash fallout within ~20-30 km of the volcano (Waitt and Dzurisin, 1981; Rosenbaum and Waitt, 1981). The timescales of aggregate formation are short: for example, AP1 aggregates were deposited within five minutes of the onset of the 1990 eruption of Sakurajima volcano, Japan (Gilbert and Lane, 1994) and frozen AP2 aggregates reached the ground in less than 10 minutes during the March 2009 eruption of Redoubt volcano, Alaska (K. Wallace, personal communication, 2010). A distinct volcanic hail layer (analogous to AP2) was also present in the deposit formed by the May 2011 eruption of Grimsvötn, Iceland (preserved in the deposit on the Vatnajökull glacier; author's own observations).

In long-lived ash clouds (>1 day), cloud water concentrations fall close to background levels due to mixing and sedimentation (e.g., Schumann et al., 2011), and electrostatic forces may then play an important role in particle binding (James et al., 2003). Aggregation played an important role in the settling of ash from eruption clouds generated during the 2010 eruption of Eyjafjallajökull volcano, Iceland (Taddeucci et al., 2011). Ground observations indicate that in May 2010 fine ash reached the surface as both particle clusters and accretionary pellets (e.g., Fig. 1) (Bonadonna et al. submitted). Particle clusters were ubiquitous in the collected samples and were observed as ash clusters and coated particles (PC1 and PC2), whereas accretionary pellets were collected in only a few localities > 20km from vent (depending on local availability of water) and were observed as poorly structured pellets and liquid pellets (AP1 and AP3). Nonetheless, all these aggregates were not very well preserved in the resulting deposit and could only be observed due to dedicated acquisition strategies at the time of fallout. In contrast, AP2 aggregates produced during the first phase of the eruption (14 April 2011) reached sizes up to a few mm and were well preserved in the

deposit (in proximal areas they fell as hail). It follows that aggregation-enhanced fallout reduced the airborne fine ash mass concentrations in distal parts of the Eyjafjallajökull ash cloud (Bonadonna et al., submitted).

Particle aggregation acts to increase fine ash accumulation rates at the surface. As an example, peak accumulation rates of $0.5\text{-}1.2\times10^{-3}$ kg m^{-2} s^{-1} during two Vulcanian explosions at Soufriere Hills volcano, Montserrat, in 1997 associated with column collapse (plume height of 3-5 km altitude) coincided with fallout of AP1 aggregates (up to 4 mm in diameter; Fig. 1D); accumulation rates for similar eruptions in the absence of aggregation were an order of magnitude less and ranged between $0.01\text{-}0.5\times10^{-3}$ kg m^{-2} s^{-1} (Bonadonna et al., 2002b). Sedimentation of AP1 aggregates at these localities (~7 km from vent) initiated ~15 minutes after the onset of the explosions, lasted 10-30 minutes, and was followed by heavy precipitation (volcanogenic water drops started falling up to an hour after the fallout event). Some AP1 aggregates also aggregated during sedimentation and formed clusters up to 6 mm in diameter. The deposit at these sites was polymodal (due to both aggregation and the combination of co-PDC and vertical plumes fallout) with a fraction of fine ash of about 50-90 wt% and a fine mode around 5 phi.

Most historic ash fall deposits from eruption columns that reach the tropopause have a distal region of increased fallout, 100s km from the volcano (Fig. 2). This is known as a distal mass accumulation maximum (or 'secondary thickening'), and results from aggregation in the drifting ash cloud (Carey and Sigurdsson, 1982; Sarna-Wojcicki et al., 1981; Rose et al., 1982; Sorem, 1982; Durant et al., 2009). Distal mass accumulation maxima have been recognised in a number of recent ash deposits e.g., 1932 Quizapu eruption (Hildreth and Drake, 1992); May-Aug, 1980 eruptions of Mount St Helens, USA (Sarna-Wojcicki et al., 1981); 1991 eruptions of Unzen, Japan (Watanabe et al., 1999); 1991 eruption of Mt Hudson (Scasso et al., 1994); 1991 Pinatubo eruption (Wiesner et al., 2004); June-Sept1992 eruptions of Crater Peak, USA, (McGimsey et al., 2001), 2008 eruption of Chaiten, Chile, (Watt et al., 2009) as well as in ancient deposits (e.g., Lerbekmo, 2002).

2.3.1. Stratigraphic context of accretionary pellets (AP) in tephra fallout layers

AP-bearing fall layers derived from eruption clouds or co-PDC clouds ('co-PDC deposit clouds') are common in the geological record and recent past (e.g., Self, 1983; Hayakawa, 1990; Schumacher and Schmincke, 1991; De Rita et al., 2002; Branney et al., 2007; Brown et al., 2010; Ritchie et al., 2002). Typically these deposits contain AP1 aggregates or, less commonly, AP2 aggregates (Table 1). Ash fall layers comprised of AP1 aggregates commonly exhibit well-sorted, framework supported textures (Fig. 3A) although coalescence may occur post-deposition after rainfall or melting of originally frozen aggregates causing the texture to disintegrate (e.g., Rosi, 1992). In the latter case, evidence that an ash layer originally comprised aggregates or ash clusters may come from poor sorting and polymodal grainsize distributions (e.g., Carey and Sigurdsson, 1982; Durant et al., 2009). Occurrences of large AP2 aggregates within widespread ash fall layers derived from eruption clouds or co-PDC clouds associated with some phreatomagmatic and phreatoplinian eruptions may be clast- or matrix-supported (Self, 1983; Giordano, 1998; Wilson, 2001; De Rita et al., 2002). Layers of well-sorted, clast-supported AP2 aggregates with lithic or pumice cores have been interpreted as proximal fall deposits at several volcanoes (e.g., Lorenz, 1974; Bednarz and Schmincke, 1990; Houghton and Smith, 1993; Cole et al., 2001).

There are few studies examining how aggregates change in size with distance from source within a fall deposit. Accretionary pellets in phreatoplinian fall deposits in New Zealand decreased in diameter from 20 mm to 5 mm over distances of ~50 km (Self, 1983) and aggregates from the 11 ka eruptions of Laacher See volcano, Germany, decreased from 10 mm to 4 mm over a distance of ~10 km (Schumacher and Schmincke, 1991). Aggregates present in the ash fall deposit of the 1980 Mount St. Helens eruption associated with the initial blast increased in size away from the volcano in an extremely non-systematic manner (Figure 3 of Sisson, 1995). Ritchie et al. (2002) demonstrated that the size of aggregates within the co-PDC ash layer increased away from source.

Preservation of aggregates in distal ash-fall layers is often poor: delicate ash clusters tend to disaggregate on impact or on sedimentation through a water column, although their presence can be inferred by polymodal grainsize populations. Features such as distal mass deposition maxima are more difficult to identify in the geological record due to rapid compaction and erosion (e.g., Hildreth and Drake, 1992).

2.3.2. Stratigraphic context of accretionary pellets (AP) in pyroclastic density current deposits

Ash aggregates have been documented in the flow deposits of PDCs (excluding co-PDC deposit fall deposits). Most occurrences are of AP2 aggregates (e.g., McPhie, 1986; Schumacher and Schmincke, 1991; Ui et al., 1992; Sohn, 1996; Baer et al., 1997; Colella and Hiscott, 1997; De Rita et al., 2002; Palladino et al., 2001; Scolamacchia et al., 2005; Branney et al., 2007; Brown et al., 2007; Andrews et al., 2008; Brown et al., 2010; Ellis and Branney, 2010). AP2 aggregates are commonly found within the matrices of PDC deposits or clast-supported in discontinuous layers or lenses within bedded PDC deposit (Fig. 3A). Broken fragments of AP2 aggregates are common alongside whole ones (Brown et al., 2010). Other types of ash aggregates in PDC deposits are less commonly reported (e.g., ash pellets and clusters). Scolamacchia et al. (2005) documented a range of types (AP1-3 aggregates) in PDC deposits from the 1982 eruption of El Chichón volcano, Mexico. Fragile aggregates may not survive transport within a vigorously turbulent current or may be indistinguishable in the matrix. In a study of ash aggregates in PDC deposits at Laacher See, Schumacher and Schmincke (1991) found AP1 and AP2 aggregates distributed at various levels throughout PDC deposits. Brown et al. (2010) documented a pattern in the distribution of ash aggregate types within numerous PDC deposits emplaced during Plinian eruptions on Tenerife. In outcrops 15-20 km from the vent, the lower parts of PDC deposits lack ash aggregates, but upper parts contain matrix-supported whole and broken AP2 aggregates in low abundances. Co-PDC ash fall layers that mantle the tops of the PDC deposits and extend beyond their parent PDC deposit sheet consist exclusively of clast-supported AP1 aggregates (Fig 3B). The unstructured cores of the AP2 aggregates in the PDC deposits are closely similar to the AP1 aggregates in the overlying fall layer suggesting a genetic link. Brown et al. (2010) concluded that the AP1 aggregates grew in buoyant co-PDC plumes during the passage of the PDCs and that these subsequently accreted layers—evolving into AP2 aggregates—as they passed through lower parts of the buoyant plume above a PDC.

3. Aggregation within ash plumes: conditions and downwind changes

Large variations in particle aggregate morphology as a function of distance from source suggests that there are multiple aggregation pathways, which implies formation processes evolve with time during transport. The availability and abundance of water in the eruption cloud exerts a dominant control on aggregation (Gilbert and Lane, 1994; Veitch and Woods, 2001; Durant et al., 2009; Costa et al., 2010; Folch et al., 2010; Textor et al., 2006a and b). Initial fragmentation, particle collisions, mixed phase clouds and particle separation all lead to particle charging (Mather and Harrison, 2006) which influences particle collisions and may provide binding forces in the absence of water (more relevant for clouds 1000s km from source or >24 hours in age). Water in ash clouds originates from volatiles in the pre-eruptive magma (up to ~8 wt.%; Wallace, 2005), entrainment of moist lower tropospheric air (e.g., Woods, 1993; Ernst et al., 1996) and through interaction with external water bodies, (e.g., phreatomagmatic eruptions or from the passage of PDCs across water, e.g., Carey et al., 1996; Edmonds and Herd, 2005). The importance of cloud water content is supported by the presence of large ash aggregates (AP1 and AP2) generated in eruption clouds from both small and large phreatomagmatic eruptions in which a large proportion of external water is incorporated into the eruption column. However, these eruptions also tend to produce larger quantities of fine ash and aggregation may play a greater role in fine ash removal in hydro-volcanic clouds relative to eruption clouds containing less water.

The presence of aggregates >4 mm diameter (diameter of raindrop break-up; Houze, 1994) requires a mixed-phase cloud containing supercooled liquid water and ice: growth to sizes above this threshold occurs through riming and recycling through the cloud. Fallout of centimetre-sized aggregates occurs in close association with the eruption column usually found within 10s km of the vent. Aggregate sedimentation results in the progressive removal of fine ash and water, such that a given eruption cloud will become depleted in water and fine ash with distance from the volcano. Thus, the rate of aggregation directly effects airborne particle mass in distal regions of the cloud.

During the 18 May 1980 eruption of Mount St. Helens, medial-distal particle aggregation (i.e. > 200 km from the vent) was mainly characterized by formation of ash clusters (PC1, Table 1) of a few hundreds of microns dominantly composed of ash <45 μm (Carey and Sigurdsson, 1982; Schumacher, 1994; Sorem, 1982), whereas particle aggregation within tens of kilometres from the vent was characterized by deposition of accretionary pellets between 2-10 mm in diameter (Rosenbaum and Waitt, 1981; Carey and Sigurdsson,

1982). Hobbs et al. (1981) reported "fist-sized dry and wet clumps of loosely aggregated material bombarding their aircraft at the edge of the dark ash plume about 37 km downwind and east of the volcano" while attempting airborne measurements. Ash aggregation in distal regions of volcanic clouds emplaced in the upper troposphere (and higher) occurs much like snowflake growth (Durant et al., 2009). Ice is routinely detected in eruption clouds (Rose et al., 1995; Rose et al., 2004) and fine ash particles act as ice nuclei for the growth of ice/ash-hydrometeors (Durant et al., 2008). Throughout the May 18 eruption there were two regions of radar reflectivity at fixed location (Harris et al., 1981); one over the volcano and the other centred approximately 200-300 km downwind, which correlated to observations of aggregate fallout (Fig. 4). These observations indicated: (1) the particles in the cloud increased in size either as a result of ash particle aggregation, growth of hydrometeors on ash particles, or as a combination of both; and (2) a "bright band" in the vertical structure of radar echoes due to the ice to liquid water phase change (Fig. 5). Ash-hydrometeor growth combined with cloud base sublimation results in mammatus cloud formation (Fig. 6; Schultz et al., 2006). Aggregation efficiency increases in the presence of liquid water (Lawson et al., 1998) which allows fine ash aggregates to rapidly grow and sediment *en masse*. Liquid water should be present at temperatures to -23 °C during plume ascent (Durant et al. 2008) and at T > 0 °C during descent. Therefore the distance at which hydrometeor-enhanced aggregation occurs in the distal part of the cloud is determined by the point at which the cloud passes through the 0 °C isotherm. During fallout through regions of the atmosphere with low humidity, evaporation/sublimation may leave only fragile aggregates held together by adsorbed water molecules and electrostatic forces on particle surfaces.

In small, lower tropospheric eruption plumes, ash aggregation may be partly influenced by the ambient meteorological conditions. Gilbert et al. (1994) noticed that damp ash pellets at Sakurajima volcano fell during periods of high relative humidity (>80 % relative humidity; cloudy days following a period of regional rainfall), while apparently dry, delicate AP1 aggregates fell on days with lower relative humidity. Watanabe et al. (1999) noted that AP1 aggregates at Unzen volcano were generated in plumes above block-and-ash flows (which generally have a low water content due to the lack of magmatic water input) on cloudy days (i.e., water required to drive aggregation was derived from moist entrained air). During the 1979 eruption of Soufriere St. Vincent, fallout of 'dry' AP1 aggregates (which broke on impact) and wet AP1 and AP3 aggregates (which splashed or flattened on

deposition) occurred on separate occasions in proximal regions (< 9 km) from plumes that reached ~18 km height (Brazier et al., 1982). More observations of aggregate morphology alongside quantitative measurement of environmental parameters are needed to discriminate the effect on aggregation rate.

The development and growth of droplet and ice crystal populations has been observed as air passes over mountains in lenticular (or lee wave) clouds. Air forced up over a mountain top experiences a steady expansion and a decrease in temperature that results in cloud droplet formation followed by rapid glaciation (Field et al., 2001; Baker et al. 2006). Once over the peak, the air descends and adiabatic compression causes temperature and pressure to increase, which results in hydrometeor sublimation or evaporation. Particle aggregates were observed falling from lee waves formed in the volcanic cloud from the 3 April 1988 eruption of Mount Augustine, USA (Rose et al., 1988; W. Rose, personal communication, 2010). In this scenario, aggregation may be closely related to hydrometeor formation, which was enhanced in the upwards propagating part of the lee wave cloud.

The proportion of fine ash involved in aggregation may be inferred from satellite observations of volcanic ash clouds (e.g., 1992 eruptions of Crater Peak, USA, Rose et al., 2001). In the first 30 minutes, >70 wt.% of erupted material falls out within 10s km of the volcano while in the next 24 hours another ~25 wt.% of total erupted mass falls out within 100s km to 1000s km dominantly through hydrometeor-enhanced aggregation of fine ash. The final <5 wt.% remains airborne in detectable quantities for another 3-5 days in low concentrations. At this stage the cloud has lost most of the erupted water through sedimentation of ash-hydrometeors and mixing with dry ambient atmosphere, and electrostatic aggregation most likely dominates (e.g., see ash clusters, Fig. 1a, b).

The particle size distribution of aggregates is less well known, though may be inferred from sedimentological analysis of fallout known to be influenced by aggregation (Fig. 7) through deconvolution of measured bulk sample polymodal particle size distributions (Wohletz et al. 1989). Aggregate fallout during the 18 May 1980 Mount St. Helens eruption was linked to the formation of a distal mass deposition maximum centred on ~ 330 km from the volcano (Carey and Sigurdsson, 1982). The ash deposit beyond 125 km consisted of 4 subpopulations (Durant et al., 2009): the size and abundance of the coarsest subpopulation decreased with distance and was associated with gravitational sorting and settling of large (>100 µm) individual ash particles; the remaining 3 subpopulations were directly involved in

the ash aggregation process. One of these subpopulations with a mode at ~20 μm that dominated the deposit mass fraction over the distal maximum in mass deposition and correlated closely to the locations of observed aggregate fallout. The particle size distribution of fallout at distances >300 km was remarkably similar (mean particle diameter 18.6 μm; standard deviation 0.7 μm) and represents the size characteristics of ash involved exclusively in ash aggregation (Fig. 7). There is a growing body of evidence that indicates the particle size characteristics of aggregates do not vary between different eruptive styles (Bonadonna et al., 2002b; Durant et al., 2009; Durant et al., 2011) as it is a particle size-selective process, thus, identification and measurement of aggregation particle size subpopulations in fallout deposits provides a tool to evaluate the propensity of aggregation in historic and ancient ash deposits

Aggregation processes are not expected to be linked to eruptive style but on the combination of critical parameters (e.g. particle number distribution, temperature and availability of water). As an example, Bonadonna et al. (2002b) illustrated how aggregates derived from Vulcanian explosions and dome collapse activity show very similar characteristics with most particles <50μm sedimenting as PC1, PC2, AP1, AP2 and AP3 aggregates. The largest aggregates (3-11 mm) were produced as a result of a large dome-collapse PDC entering the ocean (26 December 1997, Ritchie et al., 2002; Mayberry et al., 2002). These aggregates also showed concentric structures (AP2 aggregates) unlike the smaller AP1 aggregates associated with the Vulcanian explosions and smaller dome collapses. Nonetheless, all aggregates observed in Montserrat were characterized by similar unimodal grainsize distribution regardless of internal structure, with the dominant fine mode in the restricted range 27-22μm (5.2-5.5φ) and representing 87-93 wt% of the samples (Bonadonna et al. 2002b). This compares closely to fallout beyond 300 km from the 18 May 1980 Mount St. Helens eruption.

Aggregate growth rates as a function of plume/cloud humidity and the effect of residence time within an ash cloud are not known. There is compelling observational evidence that some simple forms (AP1 aggregates) grow into more complex ones (concentrically structured AP2 aggregates; e.g., Brown et al., 2010), but controls on the transition from the accretion of ash as massive, structure-less aggregates to the accretion of thin concentric laminations in which each layer differs markedly from its neighbour in grainsize, or in which each layer exhibits strong grainsize grading are not constrained. In

historic eruption deposits large aggregates (>0.5 cm) are commonly confined to regions 10s km from source, although evidence in the geological record suggests that during some extremely large eruptions they can be deposited > 100 km from source (Self, 1983). Large aggregates may form preferentially within vertical columns where liquid water contents are high and where rapid vertical changes in physical conditions (e.g., temperature, phase conditions of water) allow more complex forms to develop, for example through re-entrainment of falling aggregates at the column margins or for vertical cycling through different parts of a column or proximal eruption cloud in a manner similar to hailstone formation. These conditions may also be met or approximated within co-PDC plumes above sustained PDCs.

Once ash aggregates have formed within an ash cloud, they can be further modified by passage through ambient conditions that differ from those in which they grew (e.g., in the troposphere). Whilst the effect this may have on morphology is poorly constrained, the effects on binding mechanisms can be more readily investigated. Volcanic ash plumes contain abundant aerosol and gas phases which can be scavenged by silicate ash particles and can cause rapid surface acid dissolution (Rose, 1977; Varekamp et al., 1984; Oskarsson, 1980; Delmelle et al., 2005, 2007). This can alter the particle surface chemistry and result in the precipitation of sulphate and halide salts at the ash-liquid interface (Delmelle et al., 2007). Numerous authors have documented microlitic crystals of $NaCl$ and $CaSO_4$, and FeO films that act to cement ash aggregates together (Varekamp et al., 1984; Tomita et al., 1985; Gilbert et al., 1994; Scolamacchia et al., 2005). These minerals precipitate from solution as the aggregates pass through less humid regions of the ash cloud or the atmosphere (Gilbert and Lane, 1994) and aggregates can be strongly indurated by the time they are deposited (e.g., Varekamp et al., 1984; Brown et al., 2010). Studies of amorphous cements within aggregates can help in deductions about ash cloud vapor chemistry in ash clouds (e.g., Scolamacchia et al., 2005).

4. Experimental studies on aggregation

Experimental studies over the last couple of decades have provided fundamental insight on both accretionary pellet and ash cluster formation (Gilbert and Lane, 1994; James et al., 2002; James et al., 2003; Schumacher, 1994; Schumacher and Schmincke, 1995;

Kueppers et al., 2011). As an example, even though ash clusters (PC1, Table 1) are difficult to document and analyze due to their low preservation potential, drag coefficients, aggregation coefficients and particle size distribution have been parameterized through dedicated laboratory experiments that show good agreement with field data (Gilbert and Lane, 1994; James et al., 2002; James et al. 2003; Schumacher, 1994). Material used in experiments range from natural to artificial samples with variable particle size (e.g. ash produced by breaking cm-sized pumices (James et al., 2002, 2003); volcanic ash <250 microns from the Laacher See tephra (PF deposit) (Schumacher, 1994); andesitic particles <300 microns from Sakurajima; ballottini™, silica carbide and fused alumina (Gilbert and Lane, 1994). More than 60% of PC1 aggregates observed in lab experiments are characterized by fall velocities between 0.2-0.4 m s^{-1} which is in agreement with some numerical results (Carey and Sigurdsson, 1982; Cornell et al., 1983; James et al., 2002). James et al. (2003) produced a curve-fitting model to describe the variation of aggregation coefficients with particle sizes (i.e. ratio between aggregated particles and total mass of injected particles) and compared it with the aggregation coefficient resulted from wet-aggregation experiments (Gilbert and Lane, 1994; Fig. 8). The coefficient represents a combination of collision and sticking efficiency for different particle sizes. Dry aggregates (formed in the absence of a liquid phase) up to 700 ⬚m in diameter are believed to have a typical density of 200 kg m^{-3} and are characterized by exponential cumulative distributions of particle sizes with most particles <70 ⬚m (James et al., 2002; James et al., 2003). In contrast, wet aggregates are more likely to be preserved in tephra deposits and therefore have been described in more detail (Gilbert and Lane, 1994; Schumacher and Schmincke, 1995). In particular, Gilbert and Lane (1994) have proposed a quantitative model for the formation of accretionary pellets mainly based on the aggregation coefficient, particle concentration, particle characteristics and cloud thickness. Theoretical simulations predict maximum aggregate sizes in the range of 0.7–20 mm for a cloud thickness of 0.5–10 km respectively.

Durant et al. (2008) report heterogeneous ice nucleation experiments that investigated the effect of ash particle ice nucleus composition and surface area on liquid water drop freezing temperature. They conclude that volcanic clouds have potential to be "over-seeded" due to the presence of high number concentrations of ash particles acting as ice nuclei and would contain high ice crystal concentrations with smaller size relative to pure

water clouds. Ice nucleation from the liquid phase occurs over a relatively narrow altitude range that corresponds to a temperature range of ~250-260 K.

5. Empirical and numerical studies on aggregation

Empirical parameterizations have been devised by some authors to understand particle aggregation. In particular, in order to simulate the Campanian Y-5 ash layer, Cornell et al. (1983) assumed aggregation of 50, 75 and 100 wt% of particles in the size range 125-63 μm, 63-31 μm and <31 μm respectively with all aggregates (of unspecified type) having a diameter of 250 μm (ρ = 2) and density of 2000 kg m^{-3}. The distal mass accumulation maximum (the "Ritzville bulge") of the Mount St. Helens 1980 eruption was modelled by Carey and Sigurdsson (1982) simply by allowing ash <63 μm in diameter to fall with a collective terminal velocity of 0.35 m s^{-1}. From this computed terminal velocity and from the observed density of particles deposited in this region (2000 kg m^{-3}) they also deduced an aggregate density of 200-1300 kg m^{-3} (assuming 0.9-0.4 porosity). Armienti et al. (1988) considered the same data and required particles smaller than 90 μm to fall at speed of 0.55 m s^{-1}. However, Carey and Sigurdsson (1982) were not able to reproduce the deposition of fine ash observed in the proximal area (Hobbs et al., 1981; Carey and Sigurdsson, 1982). Schumacher (1994) reproduced the fine-ash deposition in the proximal area by electrostatic adhering of small particles (<45 μm) on larger particles (>63 μm) (i.e. single-grain clusters). Finally, Bonadonna et al. (2002a) and Bonadonna and Phillips (2003) have used a combination of the Cornell et al. (1983) model, the parameterization of Gilbert and Lane (1994), and field observations from the Montserrat eruption (Vulcanian explosions and dome collapses of August-October, 1997; Bonadonna et al., 2002b) to compile the hazard assessment of Montserrat (West Indies) and to investigate the effects of aggregation on the thinning of tephra deposits.

Empirical description of particle aggregation is an important component of volcanic hazard assessment associated with eruptions rich in fine ash, although most operational ash cloud models do not include any description of ash aggregation. Particle aggregation is a complex time-dependent process and is better described by a numerical solution. However, numerical models only exist for wet aggregation (Veitch and Woods, 2001; Textor et al., 2006a,b: Costa et al., 2010; Folch et al., 2010) and wet sedimentation (i.e., removal of ash

due to rain; Stohl et al., 2005; Draxler and Hess, 1998; D'Amours et al., 2010; Jones et al., 2007; Barsotti et al., 2008). Here we will only describe details of wet-aggregation modelling. Veitch and Woods (2001) focused on the process of wet aggregation which may arise when cloud water vapor cools and condenses during rise, thereby providing a binding agent for ash particles within the eruption column. It is assumed that particle aggregation only occurs in presence of liquid water and therefore they focus on wet aggregation in the eruption column (and assume the downwind plume of large Plinian eruptions is always below freezing temperature). As collision and sticking efficiency is decoupled and the relative speed of particles is enhanced by turbulence, the collision rate is high in the eruption column. Veitch and Woods (2001) concluded that aggregation mainly depends on the sticking and collision model used and is not significantly affected by the solid-mass fraction and the plume temperature at the vent. Initial mass flux might exert an influence on the presence of water in the column and therefore on the aggregation efficiency. Veitch and Woods (2001) also show that their model would be consistent with the distal thickening of the deposit from the 18 May 1980 eruption of Mount St. Helens.

According to Textor and Ernst (2004) the plume model considered by Veitch and Woods (2001) provides an overly-simplistic description of the aggregation process as it does not consider microphysical processes crucial for the aggregation of ash in the presence of water. Textor et al. (2006a, b) presented a more sophisticated model of wet aggregation using ATHAM (Active Tracer High-resolution Atmospheric Model, Graf et al. 1999; Herzog et al. 2003; Oberhuber et al. 1998) which accounts for the formation of liquid and solid hydrometeors and the effect on the plume dynamics from the latent heat generated by water phases changes. During rapid rise to high cold altitudes, ice is dominant in typical Plinian plumes relative to liquid water. In addition, the total mass of tephra in Plinian eruptions is about two orders of magnitude greater than that of condensed hydrometeors. As a result, most particles occur in the form of frozen aggregates with low water content (ice fractions usually lower than 10% by mass).

Costa et al. (2010) developed a model of wet aggregation based on the classical theory of Smoluchowski (1916) using a fractal relationship to describe the rate at which particles form aggregates based on three different collision mechanisms (i.e., Brownian motion, ambient fluid shear, and differential sedimentation). Both the effects of magmatic water and atmospheric water are considered as well as the effects of water in a liquid or

solid state on aggregation. Such a model was then implemented by Folch et al. (2010) in the FALL3D ash-dispersal model and validated with the data of the 18 May 1980 eruption of Mount St. Helens and the 17 September 1992 Crater Peak eruption of Mount Spurr (USA) with good agreement. The models proposed by Costa et al. (2010) and Folch et al. (2010) demonstrate that wet aggregation is limited to the cloud region characterized by temperatures above the freezing point of water, i.e. in the vertical column, as aggregation due to the presence of ice seems is less effective. Costa et al. (2010) also demonstrated how wet aggregation strongly depends on the ratio between the residence time of aggregating particles within the cloud region where liquid water exists and the time required for aggregates to form. As a result, aggregation processes are likely to be more effective in moderate explosions and small plumes than in vigorous Plinian and sub-Plinian eruptions characterized by fast ascending plumes.

6. Discussion, conclusions and suggestions for future work

Aggregation plays a fundamental role in the sedimentation of fine volcanic particles, both in proximal and distal areas, and influences atmospheric ash concentrations and tephra deposition. Fine ash particle aggregation results in greater fallout of fine ash close to source and has the effect of reducing distal (100s-1000s km) atmospheric ash concentrations. Even though ash-poor deposits can still be described without accounting for particle aggregation (Bonadonna and Phillips, 2003), models that do not account for aggregation have proven unable to reproduce tephra deposits rich in fine ash (Bonadonna et al., 2002a; Carey and Sigurdsson, 1982; Macedonio et al., 1988; Folch et al., 2010). Enhanced sedimentation of volcanic ash is important both in proximal areas and in medial-distal areas (e.g. Montserrat 1997 Vulcanian explosions and dome collapses; Bonadonna et al., 2002b; Mount St. Helens 1980; Folch et al., 2010). Very fine ash (<10 μm) falling as millimetric aggregates in populous regions are easily remobilized by human activity (e.g. by traffic, cleaning, horticulture) and pose a significant threat to human health (e.g. Baxter et al., 1999). In addition, integrating aggregation into operational forecast models of tephra dispersal is essential to avoid mismatches between predicted and observed ash concentrations in the atmosphere (especially at distances 100s-1000s km from the volcano) during explosive eruptions. Accurate parameterization volcanic ash sedimentation is crucial both to real-time forecasting

of ash dispersal (e.g. for aviation purposes; e.g. Bonadonna et al., 2011) and to hazard assessment of ground tephra accumulation (e.g. for long-term land use planning). In particular, key aspects of volcanic-ash aggregation (e.g. particle sticking properties and the role of hydrometeor formation) still need to be characterized in order to develop realistic numerical models that can be combined with models of tephra transport and sedimentation. This can only be accomplished through multidisciplinary efforts.

The presence of ash aggregates in co-PDC ash cloud fall deposits (e.g., Schumacher and Schmincke, 1991; Hobblit, 2000; Bonadonna et al., 2002b; Ritchie et al., 2002; Brown et al., 2010) indicates that conditions in ash-rich co-PDC plumes can approximate those conditions that may promote aggregation in parts of eruption columns. A large fraction of the total erupted material (35-50 vol. %) may be elutriated from PDCs by co-PDC deposit plumes (Sparks and Walker, 1977) and carried to the stratosphere (Sparks et al., 1997; Dartevelle et al., 2002), and the lofted ash is finer-grained than that dispersed by Plinian columns (Evans et al., 2009).

Ash aggregates occur with a range of morphology and internal structure that ranges from fragile particle clusters (i.e., PC1 and PC2) to accretionary pellets (i.e., AP1-3). Typically the structure of particle clusters and liquid pellets are not preserved in the stratigraphic record, whereas poorly structured and concentrically structured pellets (AP1 and AP2) are more resistant. The particle size of ash aggregates has a fairly consistent distribution for a given morphological-type that ranges from ~5 phi both for AP1 and AP2 aggregates to ~6 phi for PC1 (Bonadonna et al., 2002b; Durant et al., 2009; Durant et al., 2011). The availability of water within an eruption column or an eruption cloud and the residence time of ash particle with the eruption column and the eruption cloud control how and where aggregation occurs. Condensation occurs when the temperature falls below the dewpoint temperature and freezing occurs in a narrow range between T ≥ -13 °C to -23 °C (Durant et al., 2008). Aggregation in an evolved ash cloud (>24 hours, 100s - 1000s km from source) in the upper troposphere is driven by the growth of ice on ash particles which causes ash clouds to subside at rates that are orders of magnitude faster than terminal settling velocities of individual particles. Additionally, electrostatically-driven aggregation will become more important as the fraction of water in the cloud decreases.

Future work needs to focus on observational constraints of the process to inform and improve tephra dispersal and sedimentation models. As volcanic ash clouds contain a range

of silicate ash and aerosol particles that collectively span over seven orders of magnitude in size (Durant et al., 2010), no single technique can measure the full range of particle size that includes individual ash particles (<1 μm – 2 cm) and growing aggregates (>10 μm – >4 cm), so an integrated strategy is required (e.g., Bonadonna et al. 2011). Therefore, empirical studies should incorporate multiple observational techniques that include remote sensing (radar observations and thermal infrared imaging cameras) combined with direct analysis of fallout. Active remote sensing includes radar and lidar. Radar remote sensing is typically used to detect hydrometeor particles >1 mm diameter, although it is possible to detect silicate particles as small as 30 μm diameter (e.g., Bonadonna et al., 2011). Radar observations can constrain the spatial distribution of aggregate formation both in the vertical and horizontal, and potentially identify water phases due to vertical changes in reflectivity, and should involve a combination of high power short wavelength scanning systems with smaller vertically pointing radar rain-gauge disdrometers to directly observe fallout rates. Thermal infrared imaging systems (e.g., Prata and Bernardo, 2009) are sensitive to the fine ash fraction and could detect the onset of particle aggregation and observe changes in cloud mass loading over time. Dedicated laboratory investigation can also provide important insight on the evolution of particle aggregates both in wet and dry environments through direct control of environmental conditions (temperature and humidity). Observations remain critical in understanding ash aggregation and combining remote approaches with detailed physical analyses of aggregates, accounting for how aggregates may change with time during an eruption and with distance away from a volcano will provide much needed constraints for tephra dispersal models and for experiments aimed at investigating the physical and chemical conditions that control aggregation.

Acknowledgements

References
Alvarado, G.E., Soto, G.J., 2002. Pyroclastic flow generated by crater-wall collapse and outpouring of the lava pool of Arenal volcano, Costa Rica. Bull. Volcanol. 63, 557-568.

Andrews, G.D.M., Branney, M.J., Bonnichsen, B., & McCurry, M. 2008. Rhyolitic PDC deposits in the Rogerson graben, southern Snake River plain volcanic province: Volcanic stratigraphy, eruption history and basin evolution. Bull. Volcanol. 70, 269-291.

Armienti, P., Macedonio, G., Pareschi, M.T., 1988. A numerical-model for simulation of tephra transport and deposition - applications to May 18, 1980, Mount-St-Helens eruption. J. Geophys. Res. 93, 6463-6476.

Baer, E.M., Fisher, R., Fuller, M., Valentine, G., 1997. Turbulent transport and deposition of the Ito pyroclastic flow: Determinations using anisotropy of magnetic susceptibility. J. Geophys. Res., 102, 22565–22586.

Baker, B.A., Lawson R.P., 2006. In Situ Observations of the Microphysical Properties of Wave, Cirrus, and Anvil Clouds. Part I: Wave Clouds. J. Atmos. Sci. 63, 3160–3185.

Barsotti, S., Neri, A. Scire, J. S., 2008. The VOL-CALPUFF model for atmospheric ash dispersal: 1. Approach and physical formulation, J. Geophy. Res., 113, B03208, doi:10.1029/2006JB004623.

Barsotti, S., Neri, A., 2008. The VOL-CALPUFF model for atmospheric ash dispersal: 2. Application to the weak Mount Etna plume of July 2001. J. Geophys. Res. 113. B03209, doi:10.1029/2006JB00462.

Baxter, P.J., Bonadonna, C., Dupree, R., Hards, V.L., Kohn, S.C., Murphy, M.D., Nichols, A., Nicholson, R.A., Norton, G., Searl A., Sparks, R.S.J. and Vickers, B.P., 1999. Cristobalite in volcanic ash of the Soufrière Hills Volcano, Montserrat: Hazards implications. Science 283, 1142-1145.

Bednarz, U., Schmincke, H-U., 1990. Evolution of the Quaternary melilite-nephelinite Herchenberg volcano East Eifel. Bull. Volcanol. 52, 426-444.

Bonadonna, C., Folch, A., Loughlin, S., Puempel, H., 2011. "Ash Dispersal Forecast and Civil Aviation Workshop - Consensual Document," https://vhub.org/resources/503.

Bonadonna, C., Genco, R., Gouhier, M., Pistolesi, M., Cioni, R., Alfano, F., Hoskuldsson, A., Ripepe, M. Submitted. Tephra sedimentation during the 2010 Eyjafjallajökull eruption (Iceland) from deposit, radar and satellite observations, J. Geophys. Res.

Bonadonna, C., Ernst, G.G.J., Sparks, R.S.J., 1998. Thickness variations and volume estimates of tephra fall deposits: the importance of particle Reynolds number. J. Volcanol. Geotherm. Res. 81, 173-187.

Bonadonna, C., Macedonio, G., Sparks, R.S.J., 2002a. Numerical modelling of tephra fallout associated with dome collapses and Vulcanian explosions: application to hazard assessment in Montserrat. In: Druitt, T.H., Kokelaar, B.P. (eds) The eruption of Soufrière Hills Volcano, Montserrat, from 1995 to 1999. Geological Society of London, Memoir, pp 517-537.

Bonnadonna, C., Mayberry, G.C., Calder, E.S., Sparks, R.S.J., Choux, C., Jackson, , Lejeune, A.M., Loughlin, S.C., Norton, G.E., Rose, W.I., Ryan, G., Young, S.R., 2002b. Tephra fallout in the eruption of Soufrière Hills Volcano, Montserrat: In: Druitt, T.H., Kokelaar, B., (Eds) The eruption of Soufrière Hills Volcano, Montserrat from 1995 to 1999. Geol. Soc. London Memoir, 483-516.

Bonadonna, C., Connor, C.B., Houghton, B.F., Connor, L., Byrne, M., Laing, A., Hincks, T., 2005a. Probabilistic modeling of tephra dispersion: hazard assessment of a multiphase eruption at Tarawera. NZ J. Geophys. Res. 110 (B3, B03203).

Bonadonna, C., Phillips, J.C., 2003. Sedimentation from strong volcanic plumes. J. Geophys Res. 108, 2340-2368.

Bonadonna, C., Phillips, J.C., Houghton, B.F., 2005b. Modelling tephra sedimentation from a Ruapehu weak plume eruption. J. Geophys. Res. 110 (B8, B08209).

Branney, M.J., 1991. Eruption and depositional facies of the Whorneyside Tuff Formation, Lake District: An exceptionally large-magnitude phreatoplinian eruption: Geol. Soc. Am. Bull. 103, 886-897.

Branney, M.J., Bonnichsen, B., Andrews, G.D.M., Ellis, B., Barry, T.L., McCurry, M., 2007. Snake River SR-type volcanism on the Yellowstone hotspot track: distinctive products of unusual high-temperature silicic super-eruptions. Bull. Volcanol. 79, 293-314.

Brazier, S., Davis, A.N., Sigurdsson, H., Sparks, R.S.J., 1982. Fallout and deposition of volcanic ash during the 1979 explosive eruption of the Soufrière of St-Vincent: J. Volcanol. Geotherm. Res., 14, 335-359.

Brown, R.J., Barry, T.L., Branney, M.J., Pringle, M.S., Bryan, S.E., 2003. The Quaternary pyroclastic succession of southern Tenerife, Canary Islands: explosive eruptions, related subsidence and sector collapse: Geol. Mag. 140, 265-288.

Brown, R.J., Branney, M.J., 2004. Event-stratigraphy of a caldera-forming PDC deposit eruption on Tenerife: the 273 ka Poris Formation: Bull. Volcanol. 66, 392-416.

Brown, R.J., Kokelaar, B.P., Branney, M.J., 2007. Widespread dispersal of pyroclastic density currents during the construction of a large tuff-ring: the Ordovician Glaramara Tuff, English Lake District. Sedimentology 54, 1163-1189.

Brown, R. J., Branney, M. J., Maher, C., Dávila-Harris, P. 2010. Origin of accretionary lapilli within ground-hugging density currents: Evidence from pyroclastic couplets on Tenerife. Bull. Geol. Soc. Am. 122, 305-320.

Bursik, M.I., Carey, S.N., Sparks, R.S.J., 1992a. A gravity current model for the May 18, 1980 Mount St. Helens plume. Geophys. Res. Lett. 19, 1663-1666.

Bursik, M.I., Sparks, R.S.J., Gilbert, J.S., Carey, S.N., 1992b. Sedimentation of tephra by

volanic plumes: I. Theory and its comparison with a study of the Fogo A plinian deposit, São Miguel (Azores). Bull. Volcanol. 54, 329-344.

Carey, S.N., Sigurdsson, H., 1982. Influence of particle aggregation on deposition of distal tephra from the May 18 1980 eruption of Mount St. Helens volcano. J. Geophy. Res. 87, 7061-7072.

Carey, S., Sigurdsson, H., Mandeville, C., Bronto, S., 1996. Pyroclastic flows and surges over water: An example from the 1883 Krakatau eruption. Bull.Volcanol. 57, 493-511.

Casadevall, T.J., 1994. The 1989-1990 eruption of Redoubt volcano, Alaska: Impacts on aircraft operations. J. Volcanol. Geotherm. Res., 62, 301-316.

Cole, D., Scarpati, C., 1993. A facies interpretation of the eruption and emplacement mechanisms of the upper part of the Neapolitan Yellow Tuff, Campi Flegrei, southern Italy. Bull. Volcanol. 55, 311-326.

Cole, P.D., Guest, J.E., Duncan, A.M., Pacheco, J. 2001. Capelinhos 1957-1958, Faial, Azores: Deposits formed by an emergent surtseyan eruption. Bull. Volcanol. 63, 204-220.

Collela, A., Hiscott, R.N., 1997. Pyroclastic surges of the Pleistocene Monte Guardia sequence Lipari Island, Italy: depositional processes. Sedimentology, 44, 47-66.

Connor, C.B., Hill, B.E., Winfrey, B., Franklin, N.M., La Femina, P.C., 2001. Estimation of volcanic hazards from tephra fallout. Nat. Haz. Rev. 2, 33-42.

Connor, L.G., Connor, C.B., 2006. Inversion is the key to dispersion: understanding eruption dynamics by inverting tephra fallout. In: Mader, H., Cole, S., Connor, C.B., Connor, L.G. (eds) Statistics in Volcanology. Geol. Soc. London. pp 231-242.

Cornell, W., Carey, S., Sigurdsson, H., 1983. Computer-simulation of transport and deposition of the Campanian Y-5 Ash. J. Volcanol. Geotherm. Res.17, 89-109.

Costa, A., Macedonio, G., Folch, A., 2006. A three-dimensional Eulerian model for transport and deposition of volcanic ashes. Earth Planet. Sci. Lett. 241, 634-647.

Costa, A., Folch, A., Macedonio, G., 2010. A model for wet aggregation of ash particles in volcanic plumes and clouds: 1. Theoretical formulation, J. Geophys. Res.115, B09201, doi:10.1029/2009JB007175.

D'Amours, R., Malo, A., Servranckx, R., Bensimon, D., Trudel, S., Gauthier, J.-, 2010. Application of the atmospheric Lagrangian particle dispersion model MLDP0 to the 2008 eruptions of Okmok and Kasatochi volcanoes, J. Geophys. Res. doi:10.1029/2009JD013602.

Dartevelle, S., Ernst, G.J., Stix, J., Bernard, A., 2002. Origin of the Mount Pinatubo climactic eruption cloud: Implications for volcanic hazards and atmospheric impacts. Geology, 30, 663-666.

Delmelle, P., Lambert, M., Dufrêne, Y., Gerin, P., & Óskarsson, N. 2007. Gas/aerosol-ash interaction in volcanic plumes: New insights from surface analyses of fine ash particles. Earth Planet. Sci. Lett. 259, 159-170.

Delmelle, P., Villiéras, F., & Pelletier, M., 2005. Surface area, porosity and water adsorption properties of fine volcanic ash particles. Bull. Volcanol. 67, 160-169.

De Rita, D., Giordano, G., Esposito, A., Fabbri, M., Rodani, S., 2002. Large volume phreatomagmatic PDC deposits from the Colli Albani Volcano Middle Pleistocene, Italy: J. Volcanol. Geotherm. Res., 118, 77-98.

Draxler, R.R., G.D. Hess, 1998. An overview of the HYSPLIT_4 modeling system of trajectories, dispersion, and deposition. Aust. Meteor. Mag., 47, 295-308.

Durant, A.J., Shaw, R.A., Rose, W.I., Mi, Y., Ernst, G.G.J., 2008. Ice nucleation and overseeding of ice in volcanic clouds. J. Geophy. Res., 113D9:doi: 10.1029/2007JD009064.

Durant, A.J., Rose, W.I. 2009. Sedimentological constraints on hydrometeor-enhanced particle deposition: 1992 Eruptions of Crater Peak, Alaska, J. Volcanol. Geotherm. Res., 186, 40-59.

Durant, A.J., Rose, W.I., Sarna-Wojcicki, A.M., Carey, S., Volentik, A.C.M., 2009. Hydrometeor-enhanced tephra sedimentation: Constraints from the, 18 May, 1980 eruption of Mount St. Helens. J. Geophy. Res., 114:doi:10.1029/2008JB005756.

Durant, A.J., Bonadonna, C., Horwell, C.J., 2010. Atmospheric and Environmental Impacts of Volcanic Particulates. Elements 6, 235-240.

Durant, A., Villarosa, G., Rose, W.I., Delmelle, P., Prata, A.J., Viramonte, J.G., 2011. Long-range volcanic ash transport and fallout during the 2008 eruption of Chaitén volcano, Chile. Phys. Chem. Earth. this issue.

Edgar, C. J., Wolff, J. A., Olin, P. H., Nichols, H. J., Pittari, A., Cas, R. A. F., Reiners, P. W., Spell, T. L., Marti, J., 2007. The late quaternary Diego Hernandez Formation, Tenerife: Volcanology of a complex cycle of voluminous explosive phonolitic eruptions. J. Volcanol. Geotherm. Res. 160, 59-85.

Edmonds, M., Herd, R.A., 2005. Inland-directed base surge generated by the explosive interaction of pyroclastic flows and seawater at Soufrière Hills volcano, Montserrat. Geology, 33, 245-248.

Edmonds, M., Herd, R.A., Strutt, M.H., 2006. Tephra deposits associated with a large lava dome collapse, Soufrière Hills Volcano, Montserrat 12–15 July 2003. J. Volcanol. Geotherm. Res. 153, 313–330.

Ellis, B., Branney, M. J., 2010. Silicic phreatomagmatism in the snake river plain: The Deadeye Member. Bull. Volcanol. 72, 1241-1257.

Evans, J.R., Huntoon. J.E., Rose, W.I., Varley, N.R., Stevenson, J.A., 2009. Particle sizes of andesitic ash fallout from vertical columns and co-pyroclastic flow clouds, Volcan de Colima, Mexico. Geology 37, 935-938.

Field, P.R., et al., 2001. Ice nucleation in orographic wave clouds: Measurements made during INTACC. Quart. J. Roy. Meteor. Soc., 127, 1493-1512.

Fisher, R.V., Schmincke, H-U., 1984, Pyroclastic rocks. Springer-Verlag, 472 pp.

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. J. Geophys. Res. 115, B09202, doi:10.1029/2009JB007176.

Gardner, C.A., Neal, C.A., Waitt, R.B., Janda, R.J., 1994. Proximal pyroclastic deposits from the 1989-1990 eruption of Redoubt Volcano, Alaska Stratigraphy, distribution, and physical characteristics. J. Volcanol. Geotherm. Res., 62, 213-250.

Gilbert, J.S., Lane, S.J., 1994. The origin of accretionary lapilli. Bull. Volcanol. 56, 398-411.

Giordano, G., 1998. Facies characteristics and magma-water interaction of the white trachytic tuffs (Roccamonfina volcano, southern Italy). Bull. Volcanol. 60, 10-26.

Glaze, L.S., Self, S. 1991. Ashfall dispersal for the 16 September 1986, eruption of Lascar, Chile, calculated by a turbulent-diffusion model. Geophys. Res. Lett. 18, 1237-1240.

Graf, H.F., Herzog, M., Oberhuber, J.M., Textor, C., 1999. Effect of environmental conditions on volcanic plume rise. J. Geophys. Res. 104D20, 24309-24320.

Harris, D.M., Rose, W.I., Roe, R., Thompson, M.R., Lipman, P.W., Mullineaux, D.R., 1981. Radar observations of ash eruptions. In: The, 1980 Eruptions of Mount St. Helens, Washington. U. S. Geological Survey, Reston, VA, pp 323-333.

Hayakawa, Y., 1990. Mode of eruption and deposition of the Hachinohe phreatoplinian ash from the Towada Volcano, Japan. Geograph. Rep. Tokyo Met. Uni. 25, 167-182.

Herzog, M., Oberhuber, J.M., Graf, H.F., 2003. A prognostic turbulence scheme for the nonhydrostatic plume model ATHAM, J. Atmos. Sci. 6022, 2783-2796.

Hildreth, W., Drake, R.E., 1992. Volcan Quizapu, Chilean Andes. Bull. Volcanol. 54, 93-125.

Hoblitt, R.P., 2000. Was the 18 May 1980 lateral blast at Mt St Helens the product of two explosions? Phil. Trans. R. Soc. Lond. A 358, 1639-1661.

Hobbs, P.V., Lyons, J.H., Locatelli, J.D., Biswas, K.R., Radke, L.F., Weiss, R.R., Rangno, A.L., 1981. Radar Detection of Cloud-Seeding Effects. Science, 213, 1250-1252.

Horwell, C.J., Baxter, J., 2006. The respiratory health hazards of volcanic ash: a review for volcanic risk mitigation. Bull. Volcanol. 69, 1-24.

Houghton, B.F., Smith, R.T., 1993. Recycling of magmatic clasts during explosive eruption: estimating the true juvenile content of phreatomagmatic volcanic deposits. Bull. Volcanol. 55:414-420.

Houze, R.A., 1994. Cloud Dynamics. Academic Press.

Hurst, A.W., Turner, R., 1999. Performance of the program ASHFALL for forecasting ashfall during the 1995 and 1996 eruptions of Ruapehu volcano. NZ J. Geol. Geophys. 42, 615-622.

James, M.R., Gilbert, J.S., Lane, S.J., 2002. Experimental investigation of volcanic particle aggregation in the absence of a liquid phase. J. Geophy. Res., 107, 2191.

James, M.R., Lane, S.J., Gilbert, J.S., 2003. Density, construction, and drag coefficient of electrostatic volcanic ash aggregates. J. Geophy. Res., 108, 2435.

Jones, A.R., Thomson, D.J., Hort, M. Devenish, B., 2007. The U.K. Met Office's next-generation atmospheric dispersion model, NAME III], in Borrego, C. and Norman, A.-L. (Eds) Air Pollution Modeling and its Application XVII Proceedings of the, 27th NATO/CCMS International Technical Meeting on Air Pollution Modelling and its Application, Springer, p 580-589.

Junqueira-Brod, T.C., Gaspar, J.C., Brod, J.A., Kafino, C.V., 2005. Kamafugitic diatremes: their textures and field relationships with examples from the Goia's alkaline province, Brazil: J. South Am. Earth. Sci. 18, 337-353.

Koyaguchi, T., Ohno, M., 2001. Reconstruction of eruption column dynamics on the basis of grain size of tephra fall deposits. 1. Methods. J. Geophys. Res. 106, 6499-6512.

Kueppers, U., Auer, B., Cimarelli, C., Scolamacchia, T., Dingwell, D.B., 2011. Experimentally constraining the boundary conditions for volcanic ash aggregation. Geophys. Res. Abst., 13, EGU2011-11999.

Lane, S.J., Gilbert, J.S., Hilton, M., 1993. The aerodynamic behaviour of volcanic aggregates. Bull. Volcanol. 55, 481-488.

Lawson, R.P., Stewart, R.E., Angus, L.J., 1998. Observations and numerical simulations of the origin and development of very large snowflakes. J. Atmos. Sci., 3209-3229.

Lerbekmo, J.F. 2002. The Dorothy bentonite: An extraordinary case of secondary thickening in a Late Campanian volcanic ash fall in central Alberta. Can. J. Earth Sci. 39, 1745-1754.

Lorenz, V., 1974. Studies of the Surtsey tephra deposits. Surtsey. Res. Progr. Report. 7, 72-79.

Macedonio., G., Pareschi, M.T., Santacroce, R., 1988. A numerical simulation of the Plinian fall phase of 79 A.D. eruption of Vesuvius. J. Geophys. Res., 93, 14,817-14,827.

Macedonio, G., Costa, A., Longo, A., 2005. A computer model for volcanic ash fallout and assessment of subsequent hazard. Comp. Geosci. 31, 837-845.

Mather, T.A., Harrison, R.G., 2006. Electrification of volcanic plumes. Surv. Geophys. 27, 387-432.

Mayberry, G.C., Rose, W.I., Bluth, G.J.S. 2002. Dynamics of volcanic and meteorological clouds produced on 26 December (Boxing Day) 1997 at Soufriire Hills Volcano, Montserrat. In: Druitt, T.H., Kokelaar, B.P. (eds) The Eruption of Soufriere Hills Volcano, Montserrat. from 1995 to 1999. Geol. Soc. Lond. Mem. 21, 539-555.

McGimsey, R.G., Neal, C.A., Riley, C.M. 2001. Areal Distribution, Thickness, Mass, Volume, and Grain Size of Tephra-Fall Deposits from the 1992 Eruptions of Crater Peak Vent, Mt. Spurr Volcano, Alaska. U.S. Geological Survey Open-File Report 01–370.

McPhie, J., 1986. Primary and redeposited facies from a large-magnitude rhyolitic, phreatomagmatic eruption: Cana Creek tuff, Late Carboniferous, Australia. J. Volcanol. Geotherm. Res., 28, 319-350.

Moore, J.G., 1967. Base surge in recent volcanic eruptions. Bull. Volcanol. 30, 337-363.

Moore, J.G., Peck, D.L., 1962. Accretionary lapilli in volcanic rocks of the western continental United States. J. Geol. 70, 182-194.

Moore, J.G., Melson, W.G., 1969. Nuee ardentes of the 1968 eruption of Mayon volcano, Philippines: Bull. Volcanol. 33, 600-620.

Moore, J.G., Nakamura, K., & Alcarez, A., 1966. The 1965 eruption of Taal Volcano: Science. 151, 955-960.

Oberhuber, J.M., Herzog, M., Graf, H-F., Schwanke, K. 1998. Volcanic plume simulation on large scales. J. Volcanol. Geotherm. Res. 871-4, 29-53.

Oskarsson, N., 1980. The interaction between volcanic gases and tephra: Fluorine adhering to tephra of the 1970 Hekla eruption. J. Volcanol. Geotherm. Res.8, 251-266.

Oswalt, J.S., Nichols, W., O'Hara, J.F., 1996. Meteorological observations of the 1991 Mount Pinatubo Eruption. In: Newhall, C.G., Punongbayan. S. (eds), Fire and Mud: eruptions of Pinatubo, Philippines. Philippine Inst Volcanol Seismol, Quenzen City. University of Washington Press, Seattle: 545-570.

Palladino, D.M., Gaeta, M., Marra, F., 2001. A large K-foiditic hydromagmatic eruption from the early activity of the Alban Hills Volcanic District, Italy. Bull. Volcanol. 63, 345-359.

Prata, A.J., Bernardo, C., 2009. Retrieval of volcanic ash particle size, mass and optical depth from a ground-based thermal infrared camera. J. Volcanol. Geotherm. Res. 186, 91-107.

Reimer, T.O., 1983. Accretionary lapilli in volcanic ash falls: physical factors governing their formation. In: Peryt, T.M., (Ed) Coated grains. Springer, Berlin Heidelberg New York, p 56-68.

Ritchie, L.J., Cole, P.D., Sparks, R.S.J., 2002. Sedimentology of deposits from the pyroclastic density currents of 26 December 1997 at Soufrière Hills Volcano, Montserrat, in The eruption of Soufrière Hills Volcano, Montserrat, from 1995 to 1999, edited by T. H. Druitt and B. P. Kokelaar, Geol. Soc. London Mem. 21, 435-456.

Rogers, R.R., Yau, M.K., 1989. Short course in cloud physics. Third edition.

Rose, W.I., 1977. Scavenging of Volcanic Aerosol by Ash - Atmospheric and Volcanologic Implications, Geology, 5, 621-624.

Rose, W. I., Chuan, R.L., Woods, D.C., 1982. Small Particles in Plumes of Mount St-Helens, J. Geophys. Res. 87, 4956-4962.

Rose, W.I., Heiken, G., Wohletz, K., Eppler, D., Barr, S., Miller, T., Chuan, R., Symonds, R. 1988. Direct Rate Measurements of Eruption Plumes at Augustine Volcano: A Problem of Scaling and Uncontrolled Variables, J. Geophys. Res., 93, 4485-4499.

Rose, W.I., Delene, D.J., Schneider, D.J., Bluth, G.J.S., Krueger, A.J., Sprod, I., McKee, C., Davies, H.L., Ernst, G.G.J., 1995. Ice in the, 1994 Rabaul eruption cloud: implications for volcano hazard and atmosphere effects. Nature, 375, 477-479.

Rose, W.I., Bluth, G.J.S., Schneider, D.J., Ernst, G.G.J., Riley, C.M., Henderson, L.J., McGimsey, R.G., 2001. Observations of volcanic clouds in their first few days of Atmospheric residence: the, 1992 eruptions of Crater Peak, Mount Spurr volcano, Alaska. J. Geol.109, 677-694.

Rose, W.I., Bluth, G.J.S., Watson, I.M. 2004. Ice in volcanic clouds: When and Where? Proceedings of the 2nd International Conference on volcanic ash and aviation safety: OFCM Washington D.C., Session 3, 61.

Rose, W.I. Durant. A.J., 2009. El Chichon volcano, April 4, 1982: volcanic cloud history and fine ash fallout. Natural hazards, 51, 364-373.

Rosenbaum, J.G., Waitt, R.B., 1981. Summary of eyewitness accounts of the May 18 eruption. In: Eruptions of Mount St. Helens, Washington, edited by Lipman W.P. and Mullineaux, D.R., pp. 601-616, U.S. Geol. Surv. Prof. Paper Washington, D.C.

Rosi, M., 1992. A model for the formation of vesiculated tuff by the coalescence of accretionary lapilli. Bull. Volcanol. 54, 429-434.

Sarna-Wojcicki, A.M., Meyer, C.E., Woodward, M.J., Lamothe, P.J., Lipman, P.W., Mullineaux, D.R., 1981. Composition of air-fall ash erupted on May 18, May 25, June 12, July 22 and August 7. In: Eruptions of Mount St. Helens, Washington, edited by Lipman W.P. and Mullineaux, D.R., U.S. Geol. Surv. Prof. Paper Washington, D.C., 601-616,

Scasso, R.A., Corbella, H., and Tiberi, P., 1994. Sedimentological analysis of the tephra from the 12–15 August 1991 eruption of Hudson volcano: Bull. Volcanol. 56, 121–132.

Schumacher, R., 1994. A reappraisal of Mount St. Helens' ash clusters—depositional model from experimental observation. J. Volcanol. Geotherm. Res., 59, 253–260.

Schumacher, R., Schmincke, H-U., 1991. Internal structure and occurrence of accretionary lapilli - a case study at Laacher See Volcano. Bull. Volcanol. 53, 612-634.

Schumacher, R., Schmincke, H-U., 1995. Models for the origin of accretionary lapilli. Bull. Volcanol. 56, 626-639.

Schumann, U., Weinzierl, B., Reitebuch, O., Schlager, H., Minikin, A., Forster, C., Baumann, R., Sailer, T., Graf, K., Mannstein, H., Voigt, C., Rahm, S., Simmet, R., Scheibe, M., Lichtenstern, M., Stock, P., Rüba, H., Schäuble, D., Tafferner, A., Rautenhaus, M., Gerz, T., Ziereis, H., Krautstrunk, M., Mallaun, C., Gayet, J.F., Lieke, K., Kandler, K., Ebert, M., Weinbruch, S., Stohl, A., Gasteiger, J., Groß, S., Freudenthaler, V., Wiegner, M., Ansmann, A., Tesche, M., Olafsson, H., Sturm, K., 2011. Airborne observations of the Eyjafjalla volcano ash cloud over Europe during air space closure in April and May 2010. Atmos. Chem. Phys. 11, 2245-2279.

Schultz, D.M., Kanak, K.M., Straka, J.M., Trapp, R.J., Gordon, B.A., Zrnic, D.S., Bryan, G.H., Durant, A.J., Garrett, T.J., Klein, P.M., Lilly, D.K., 2006. The mysteries of mammatus clouds: observations and formation mechanisms. J. Atmos. Sci. 63, 2409-2435.

Scolamacchia, T., Macías, J.L., Sheridan, M.F., Hughes, S.R., 2005. Morphology of ash aggregates from wet pyroclastic surges of the 1982 eruption of El Chichón Volcano, Mexico. Bull. Volcanol. 68, 171-200.

Scott, W.E., McGimsey, R.G. 1994. Character, mass, distribution, and origin of tephra-fall deposits of the 1989-1990 eruption of Redoubt volcano, south-central Alaska. J. Volcanol. Geotherm. Res., 62, 251-272.

Searcy, C. Dean, K., Stringer, W., 1998. PUFF: A high-resolution volcanic ash tracking model. J. Volcanol. Geotherm. Res. 80, 1-16.

Self, S., 1983,. Large-scale phreatomagmatic silicic volcanism - a case-study from New-Zealand: J. Volcanol. Geotherm. Res. 17, 433-469.

Sheridan, M.F., Wohletz, K.H., 1983. Origin of accretionary lapilli from the Pompeii and Avellino deposits of Vesuvius: In, Gooley, R., Ed Microbeam Analysis. San Francisco Press, 35-38.

Sisson, T.W., 1995. Blast ashfall deposit of May, 18, 1980 at Mount St. Helens. Washington. J. Volcanol. Geotherm. Res. 66, 203-216.

Smoluchowski, M., 1916. Drei Vorträge über Diffusion, Brownsche Molekularbewegung und Koagulation von Kolloidteilchen. Phys Z, 17, 557-571 and 585-599.

Sohn, Y.K., 1996. Hydrovolcanic processes forming basaltic tuff rings and cones on Cheju Island, Korea. Geol. Soc. Am. Bull. 108, 1199-1211.

Sorem, R.K., 1982. Volcanic ash clusters: tephra rafts and scavengers. J. Volcanol. Geotherm. Res., 13, 63-71.

Sparks, R.S.J., Self, S., Walker, G.P.L., 1973. Products of PDC deposit eruptions. Geology 1, 115-118.

Sparks, R.S.J., Walker, G.L., 1977. Significance of vitric-enriched air-fall ashes associated with crystal-enriched PDC deposits. J. Volcanol. Geotherm. Res., 2, 329-341.

Sparks, R.S.J., Bursik, M.I., Carey, S.N., Gilbert, J.S., Glaze, L.S., Sigurdsson, H., Woods, A.W., 1997. Volcanic Plumes. John Wiley & Sons, Chichester, p 574.

Spence, R.J.S., Kelman, I., Baxter, P.J., Zuccaro, G., Petrazzuoli, S. 2005. Residential building and occupant vulnerability to tephra fall. Nat. Haz. Earth Syst. Sci. 5, 477-494.

Stewart, C., Johnston, D.M., Leonard, G.S., Horwell, C.J., Thordarson, T., Cronin, S.J., 2006. Contamination of water supplies by volcanic ashfall: A literature review and simple impact modelling. J. Volcanol. Geotherm. Res. 158, 296-306.

Stohl, A., Forster, C., Frank, A., Seibert, P., Wotawa, G. 2005. Technical Note: The Lagrangian particle dispersion model FLEXPART version 6.2. Atmos. Chem. Phys. 5, 2461-2474.

Suzuki, T. 1983. A theoretical model for dispersion of tephra. In: Shimozuru D, Yokoyama I (eds) Arc Volcanism, Physics and Tectonics. Terra Scientific Publishing Company (TERRAPUB), Tokyo, pp 95-113.

Taddeucci, J., Scarlato, P., Montanaro, C., Cimarelli, C., Del Bello, E., Freda C., Andronico, D., Gudmundsson, M.T., 2011. Aggregation-dominated ash settling from the Eyjafjallojökull volcanic cloud illuminated by field and laboratory high-speed imaging. Geology, 39, 891-894.

Textor, C., Ernst, G.G.J., 2004. Comment on "particle aggregation in volcanic eruption columns" by Veitch, G., Woods, A.W. J. Geophys. Res. 109, B05202 1-6.

Textor, C., Graf, H.F., Herzog, M., Oberhuber, J.M., Rose, W.I., Ernst, G.G.J., 2006a. Volcanic particle aggregation in explosive eruption columns. Part I: Parameterization of the microphysics of hydrometeors and ash. J. Volcanol. Geotherm. Res., 150, 359-377.

Textor, C., Graf, H.F., Herzog, M., Oberhuber, J.M., Rose, W.I., Ernst, G.G.J., 2006b. Volcanic particle aggregation in explosive eruption columns. Part II: numerical experiments. J. Volcanol. Geotherm. Res., 150, 378-394.

Thordarson, T., 2004. Accretionary-lapilli-bearing pyroclastic rocks at ODP Leg, 192 Site, 1184: a record of subaerial phreatomagmatic eruptions on the Ontong Java Plateau: In Fitton, J.G., Mahoney, J.J., Wallace, J., Saunders, A.D., Eds, Origin and evolution of the Ontong Java Plateau. Geol. Soc. London. Spec. Pub. 229, 275-306.

Tomita, K., Kanai, T., Kobayashi, T., Oba, N., 1985. Accretionary lapilli formed by the eruption of Sakurajima volcano: J. Jap. Ass. Min. Pet. Econom. Geol. 80, 49-54.

Trusdell, F.A., Moore, R.B., Sako, M., White, R.A., Koyanagi, S.K., Chong, R., Camacho, J.T., 2005. The, 2003 eruption of Anatahan volcano, Commonwealth of the Northern Mariana Islands: chronology, volcanology, and deformation. J. Volcanol. Geotherm. Res., 146, 184-207.

Ui, T., Kobayashi, T., Suzuki-Kamata, K., 1992. Calderas and pyroclastic flows in southern Kyushu. In: Kato, H., Noro, H., 29th IGC field trip guidebook 4: volcanoes and geothermal fields of Japan 245–276.

Varekamp, J.C., Luhr, J.F., Prestegaard, K.L., 1984. The 1982 eruption of El Chichón volcano Chiapas, Mexico: character of the eruptions, ash fall deposits and gas phase. J. Volcanol. Geotherm. Res. 23, 39–68.

Veitch, G.W., Woods, A.W., 2001. Particle aggregation in volcanic eruption plumes: J. Geophy. Res. 106, 26425-26441.

Waitt, J.R., Dzurisin, D., 1981, Proximal air fall deposits from the May 18 eruption - stratigraphy and field sedimentology. In: Eruptions of Mount St. Helens, Washington, edited by Lipman W.P. and Mullineaux, D.R., U.S. Geol. Surv. Prof. Paper Washington, D.C. 601-616.

Wallace, P.J., 2005. Volatiles in subduction zone magmas: concentrations and fluxes based on melt inclusion and volcanic gas data. J. Volcanol. Geotherm. Res. 140, 217-240.
Wardman, J.B., Wilson, T.M., Bodger, P.S., Cole, J.W., Johnston, D.M., 2011. Investigating the electrical conductivity of volcanic ash and its effect on HV power systems. Phys. Chem. Earth, this issue.

Watanabe, K., Ono, K., Sakaguchi, K., Takada, A., Hoshizumi, H., 1999. Co-pyroclastic density current ash-fall deposits of the 1991 eruptions of Fugen-dake, Unzen Volcano, Japan. J. Volcanol. Geotherm. Res. 89, 95-112.

Watt, S.F.L., Pyle, D.M., Mather, T.A., Martin, R.S., Matthews, N.E., 2009. Fallout and distribution of volcanic ash over Argentina following the may 2008 explosive eruption of chaitén, chile. J. Geophys. Res. 114 B04207, doi:10.1029/2008JB006219.

Wiesner, M.G., Wetzel, A., Catane, S.G., Listanco, E.L., Mirabueno, H.T., 2004. Grain size, areal thickness distribution and controls on sedimentation of the, 1991 Mount Pinatubo tephra layer in the South China Sea. Bull. Volcanol. 66, 226-242.

Williams, E.R., McNutt, S.R., 2004. Total Water Contents in Volcanic Eruption Clouds and Implications for Electrification and Lightning. In: Proceedings of the, 2nd International Conference on volcanic ash and aviation safety. OFCM Washington D.C., pp 67-71.

Wilson, C.J.N., 2001. The, 26.5 ka Oruanui eruption, New Zealand: an introduction and overview. J. Volcanol. Geotherm. Res.112, 133-174.

Wohletz, K.H., Sheridan, M.F., Brown, W.K., 1989. Particle-Size Distributions and the Sequential Fragmentation Transport-Theory Applied to Volcanic Ash. J. Geophys. Res. 94, 15703-15721.

Woods, A.W. 1993. Moist convection and the injection of volcanic ash into the atmosphere. J. Geophys. Res., 98, 17,627-17,636.

Figure Captions
Figure 1. Ash aggregate types. PC1 ash cluster (A) PC2 coated particle (B) and AP1 aggregates (C) from the 2010 eruption of Eyjafjallajokull volcano, Iceland (Bonadonna et al., submitted). D) AP1 from Soufriere Hills volcano, Montserrat (Bonadonna et al., 2002b) E) AP2 from Upper Scoria deposits, Santorini. F) Close-up image of rims of an AP2 concentrically structured accretionary pellet from Poris PDC deposit, Tenerife (Brown and Branney, 2004). G) Evaporated mud rain droplet (AP3) from 2010 eruption of Eyjafjallajokull volcano, Iceland (Bonadonna et al., submitted).

Figure 2. Isopach map for the May 18 1980 eruption of Mount St Helens. Distal accumulation maximum centred over Ritzville (from Durant et al., 2009).

Figure 3. Common aggregate bearing lithofacies found within PDC deposit sheets. A) matrix-supported AP2 aggregates within PDC deposit matrix. Note abundance of aggregate fragments. B) clast-supported AP1 aggregates with open framework texture in co-PDC deposit fall deposit. Examples from Poris PDC deposit and La Caleta PDC deposit, Tenerife (Brown et al., 2003).

Figure 4. Volcanic cloud from the, 18 May, 1980 Mount St Helens eruption delineated by satellite and radar observations. Radar reflectivity is represented by shaded regions with darker colour corresponding to higher reflectivity. There were two persistent regions of high reflectivity throughout the eruption: one at the volcano and the other centred over the region of distal ash mass accumulation. Dark grey shading corresponds to Level 2 reflectivity

of Harris et al. (1981) which is equivalent to stratiform rainfall rates of 2.5-12.5 mm/hr. The light grey shading corresponds to a weaker reflectivity. Modified from Folch et al. (2010).

Figure 5. Compared to raindrops, snowflakes have a complex morphology that results in high drag and lower fall velocities in the atmosphere. As a consequence, the volumetric mass loading of snowflakes in the atmosphere is greater than water drops, which have higher fall velocities and larger separation distances between hydrometeor particles. Additionally, because of greatly different dielectric properties, liquid hydrometeors have a higher radar reflectivity than frozen hydrometeors. A "bright band" in vertically-pointing radar reflectivities observations results from an increase in K (reflectivity) as snow melts to form liquid water during descent. As a snowflake passes through the 0 °C isotherm, the outside of the particle becomes liquid, while particle density and fall velocity remain almost unaffected. Consequently, particle mass loading remains similar but reflectivity rapidly increases, which is responsible for the increased radar reflectivity. As the snowflake fully melts, the structure collapses and forms a drop, and consequently fall velocity and particle separation increases which causes a decrease in reflectivity.

Figure 6. Model for mammatus-driven distal ash sedimentation, formation of aggregates, and distal mass deposition maxima in tephra deposits (Durant et al., 2009).

Figure 7. Particle size distribution of the deposit from the 18 May 1980 eruption of Mount St. Helens. The distributions were averaged along sampling transects at increasing distance from the volcano (DAVIES, 8 samples, average distance 328 km; WOOD, 4 samples, average distance 435 km; BATE, 7 samples, average distance 630 km).

Figure 8. Bulk aggregation coefficient of particles with different sizes as derived by James et al. (2003) and Gilbert and Lane (1994).

CANADA

Washington

cloud boundary
from GOES

radar reflectivity

Montana

○ Pullman

Idaho

Mount
St. Helens

100 km

Oregon

WY

snowflakes have high
drag and low fall velocity

separation
distance is small

0 C

BRIGHT BAND
HIGH
REFLECTIVITY

$|K|^2_{liquid} > |K|^2_{ice}$

outside of
hydrometeor becomes
liquid, while density
and fall velocity do
not change much

snowflake structure collapses
during melting and forms a drop

fall velocity and separation
distance increases

(1) Ash particles nucleate ice, become encased, and settle faster

(2) Mammatus generation as ice crystal number density and size increase

(3) Rapid cloud subsidence from ash-hydrometeor loading and sublimation at cloud base occurs at a rate far greater than particle terminal fall velocities

zone of aggregation

(4) Turbulence-induced ash-hydrometeor aggregation (T.>-36°C), analogous to snowflake production

(5) Melting, mixed phase hydrometeors and rapid wet aggregation

radar bright band high reflectivity

Ash-hydrometeor key
● ice
● liquid water
● ash

(6) En masse sedimentation of ash-hydrometeors from cloud base

(7) Sublimation / evaporation of water from ash-hydrometeors

Increasing downwind distance from volcano (or time) ⟶

(8) Deposition of ash clusters and formation of distal deposition maximum