Hail formation triggers rapid ash aggregation in volcanic plumes

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During explosive eruptions, airborne particles collide and stick together, accelerating the fallout of volcanic ash and climate-forcing aerosols. This aggregation process remains a major source of uncertainty both in ash dispersal forecasting and interpretation of eruptions from the geological record. Here we illuminate the mechanisms and timescales of particle aggregation from a well-characterized ‘wet’ eruption. The 2009 eruption of Redoubt Volcano, Alaska, incorporated water from the surface (in this case, a glacier), which is a common occurrence during explosive volcanism worldwide. Observations from C-band weather radar, fall deposits and numerical modelling demonstrate that hail-forming processes in the eruption plume triggered aggregation of ~95% of the fine ash and stripped much of the erupted mass out of the atmosphere within 30 min. Based on these findings, we propose a mechanism of hail-like ash aggregation that contributes to the anomalously rapid fallout of fine ash and occurrence of concentrically layered aggregates in volcanic deposits.

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number of explosive volcanic eruptions have been likened to ‘dirty thunderstorms’ due to their powerful convective updrafts, elevated water contents and electrical activity. All volcanic plumes contain some water originating from the magma, which is typically in the range of 2–7 wt.%. However, wet eruptions also incorporate water from external sources, by interacting with, for example, glaciers or aquifers. This water involvement is important because cloud microphysical processes—such as condensation and ice formation—impact volcanic plume development by scrubbing fine ash and sulfur from the atmosphere. Despite recent observations that a major proportion of erupted fine ash never makes it into the downwind cloud, the actual mechanisms of near-source aggregation are poorly constrained, and therefore difficult to predict.

For decades, it has been proposed that spherical pellets of ash created during explosive eruptions (also referred to as accretionary lapilli) are akin to hailstones due to their similar size range, concentric structure and, sometimes, ice content. For example, ash-laden hailstones fell during the Icelandic eruptions of Grimsvötn in 2011 (ref. 12) and Eyjafjallajökull in 2010 (refs 13,14) and ‘ice-cold mudballs’ splattered when they landed up to 10 mm in diameter.19 Our analysis focuses on the downwind ash cloud were recorded in C-band Doppler radar and water and ice in triggering the fallout of fine ash close to source. All volcanic plumes contain some water originating from the magma, which is typically in the range of 2–7 wt.%. However, wet eruptions also incorporate water from external sources, by interacting with, for example, glaciers or aquifers. This water involvement is important because cloud microphysical processes—such as condensation and ice formation—impact volcanic plume development by scrubbing fine ash and sulfur from the atmosphere. Despite recent observations that a major proportion of erupted fine ash never makes it into the downwind cloud, the actual mechanisms of near-source aggregation are poorly constrained, and therefore difficult to predict.

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particles that were left behind (note: these components have different densities and fall velocities).

3D simulation of the eruption plume using ATHAM. The frozen ash aggregates and ‘regular’ hailstones originated near the eruptive source, indicated by their rapid size increase towards Redoubt Volcano (Fig. 1a). These observations strongly suggest that hail formation occurred directly within the volcanic plume. To test the physical plausibility of this process, we modelled event 5 using the Active Tracer High-resolution Atmospheric Model (ATHAM), which is a cloud-resolving large-eddy simulation for explosive eruptions\textsuperscript{23–25}. The simulation was initialized from well-constrained volcanic and atmospheric characteristics of the event, including the post-aggregation grain-size distribution (GSD) derived from our measurements (Fig. 1d, Supplementary Fig. 2 and Supplementary Table 4). The 3D volcanic plume simulation generates high concentrations of hail, which fall out within 20 km of Redoubt Volcano (Fig. 2; Supplementary Movie 1). Locations and timescales of the hail formation show excellent agreement with C-band Doppler radar measurements and field deposits (Figs 2 and 3). Ten minutes after eruption initiation, the modelled plume reaches 19.2 km above sea level, matching the radar echo top (Fig. 4a). Shortly thereafter, coarse particles > 500 μm, including hail and ash aggregates, begin to separate from the suspended mixture by gravitational fallout.

Figure 2 | Comparison of 3D large-eddy simulation and measured radar reflectivity. (a–d) Modelled plume at \(T = 5, 10, 15\) and 20 min after eruption start. Note: eruption ends at \(T = 6\) min. Total volcanic particles (sum of single-particle and aggregate size bins) shown as an isosurface coloured by height and hail as a transparent blue isosurface, both at 1 mg m\(^{-3}\) concentration. Dashed white lines show maximum height of hail isosurface. Fallout of hail in the model corresponds to the rapid descent of radar echo tops from ref. 20, which is shown in e–h as north–south cross-sections at roughly equivalent time steps. Distance between vertical lines is 5 km.

Figure 3 | Gravitational separation of particles in the event 5 volcanic plume from Redoubt Volcano. (a) Time series showing maximum heights of the radar-detected plume (green circles) and modelled particles (lines). Mean radar heights derived from the highest-angle scan containing the cloud in plan view; error bars give maxima and minima. Solid line shows fine particles (<250 μm); dashed shows coarse particles (≥500 μm), both at concentrations ≥10 mg m\(^{-3}\). (b) Modelled vertical distribution of mass at \(T = 15\) min, showing horizontally integrated mass of fine ash (solid) and hail (dot-dash).
of particles or rapid freezing of supercooled droplets lead to dry growth—for example, by electrostatic attraction. Liquid water, favouring wet growth of ash aggregates similar to raindrop formation or glazing in hailstones (stage i, Fig. 4). These warmer regions of the volcanic updraft contain abundant liquid water, condensing on ash particles, coalescing rapidly into unstructured pellets of wet ash. These warmer regions of the volcanic updraft contain abundant liquid water, favouring wet growth of ash aggregates similar to raindrop formation or glazing in hailstones (stage i, Fig. 4). Freezing subsequently occurs in the outer margins of the plume that entrain cold atmospheric air (stage ii, Fig. 4). Where temperatures fall below $-20^\circ C$, the sparse amounts of liquid water lead to dry growth—for example, by electrostatic attraction of particles or rapid freezing of supercooled droplets (riming). Many of the frozen aggregates may fall out of the plume at this point, recording only a single pass through the warm, moist updraft. Alternatively, further layers of ash may be accreted during recapture into rising currents (stage iii, Fig. 4), modulated by turbulent eddies, changes in the eruption velocity at source, and bending or tilting of the volcanic plume by the ambient wind field. The layers in Redoubt Volcano’s ash aggregates (Fig. 1b,c) are strong evidence for multiple passes through updrafts and downdrafts—this layering feature is commonly observed in aggregates from other deposits of wet eruptions in the geological record. As the eruption wanes (stage iii, Fig. 4), updraft velocities can no longer support the ash aggregates and they settle out of the atmosphere, leaving behind only a small proportion of the original fine ash content ($<5\%$ in the case of Redoubt Volcano).

We suggest that hail-like growth of ash aggregates is most likely to occur when volcanic plumes: (1) ascend to atmospheric levels colder than $-20^\circ C$, where ash particles become effective ice nuclei, (2) incorporate water from an external source (for example, a glacier), and (3) produce sustained updrafts, keeping particles aloft long enough to grow. These conditions were met during the eruptions of Grímsvötn in 2011, Eyjafjallajökull in 2010 and Mount St Helens in 1980. Indeed, volcanic hail was observed from each of these eruptions.

It is worth noting that hail formation may not be constrained to eruptions in the mid- or high-latitudes. The $-20^\circ C$ isotherm is typically near or below 10 km a.s.l., even in the tropics. This height is commonly reached by eruptions of moderate to high intensity (mass flux $\geq 10^{6}$–$10^{7}$ kg s$^{-1}$), suggesting that some amount of freezing and wet growth of aggregates may be a common occurrence during powerful, water-rich eruptions in general. However, there are certainly other scenarios in which hail formation is unlikely to occur, including eruptions that are too weak to rise higher than the atmospheric freezing level, or contain very little water from the magma, surface or atmosphere. In these cases, aggregation proceeds in the absence of ice—mainly by wet growth, electrostatic attraction or some combination of the two.

The hail-like growth process we have described played a key role in the overall extent of the fall deposits from the 2009 eruption of Redoubt Volcano. Using the volcanic ash transport and dispersal model Ash3d (refs 36,37), we compared the fall deposit resulting from event 5 with and without an aggregated GSD (see Methods). In agreement with the mapped deposits,
our results indicate that aggregation increased the near-source ashfall more than fivefold and reduced the long-distance ashfall by about half, defined by the areas receiving ≥1,000 g ash per m², respectively (Supplementary Fig. 3). Aggregation scrubbed the volcanic plume of fine particles and shifted much of the erupted mass into lower regions of the atmosphere, causing early fallout and leaving behind a dilute cloud of remaining ash particles that escaped aggregation. This phenomenon has important implications for remote sensing during eruption response and ash hazards in general. To illustrate, our plume simulation (Figs 2 and 3) shows a cloud of fine-grained particles, including cloud ice, persisting at heights >10 km a.s.l. long after the fallout of larger hail and ash aggregates. The model results are in agreement with thermal infrared satellite images of an upper-level ash cloud drifting southeast >4 h after the event20,21. This upper-level cloud was not detected by the C-band radar, due to its small particle sizes and relatively dilute concentrations (ATHAM modelling suggests <250 μm and ≤4 g m⁻³, respectively). Based on this observation, it can be seen that volcanic plumes undergoing rapid aggregation may be visible in radar only briefly after the end of the eruption, while continuing to impact aviation at higher altitudes for hours to days.

Based on the present findings, we suggest that rapid, hail-like ash aggregation occurs directly within the plume arising from explosive, wet eruptions. Therefore, models of atmospheric dispersal for these scenarios may reasonably assume that ~95% of the fine ash (<250 μm) is instantaneously converted into aggregates (Fig. 4). We have emphasized that water-rich eruptions are most strongly affected by this process, although further work is required to identify the range of eruption styles and plume water contents capable of triggering significant hail formation. Refining predictive models of long-distance ash transport thus requires a sink term in the proximal area to account for ash aggregation and related instabilities in the volcanic plume23,38–40, in addition to the formation of weaker ash aggregates that may grow in the distal cloud41. We conclude that rapid, hail-like growth of ash aggregates offers a compelling explanation for a diverse range of observations from volcanic plumes and their deposits, and provides a conceptual model to guide future development of ash dispersal forecasting.

Methods

Analysis of volcanic deposits. Detailed textural analysis was undertaken on volcanic ash aggregates from a well-preserved proximal location ~12 km from Redoubt Volcano (Supplementary Fig. 1). The aggregates landed frozen and were archived in a freezer at the US Geological Survey (USGS) Alaska Volcano Observatory. The maximum aggregate diameter of ~10 mm was recorded by Wallace et al.19. To examine these deposits in detail, we freeze dried them to remove ice without melting the aggregate structures14. Water content and frozen bulk density of individual aggregates (sample sizes n = 8 and n = 46, respectively) were also obtained by measuring the three principal axes, and weighing before and after oven drying. A representative size distribution of the intact, freeze-dried aggregate sample (obtained from equatorial cross-sectional circular diameters of usually fitted ellipsoids using ImageJ) analysis of 2D images. Aggregates <0.5 mm were not measured using this technique. A selection of representative aggregates was gently disaggregated for size analysis of the single, constituent particles using a Beckman–Coulter LS 13 320 Laser Diffraction Particle Size Analyzer. Data are provided in Supplementary Tables 1–3. Analyses assumed a refractive index of 1.56 and an absorption coefficient of 0.1 for andesitic ash.

Calculating total GSD after aggregation. This study required an estimate of the total GSD in the volcanic plume after aggregation, which we refer to as GSD4. This represents the effective sizes of airborne particles during transport through the atmosphere. The calculation requires three key pieces of information, specifically the GSDs of: ‘single’ particles that were incorporated into aggregates (GSD1); whole aggregates that formed during transport (GSD2); and single particles originally produced by the eruption before aggregation (GSD3). For GSD1, we use the results of laser diffraction size analysis of ash aggregates collected 12 km from source (Supplementary Fig. 1), which were gently disaggregated before analysis. GSD2 required a mass-averaged size distribution of all aggregates in the deposit. To do this, we applied a method similar that of Murrow et al.42. We inferred the spatial extent of three aggregate isopleths (lines of equal aggregate diameter) using the mapped distribution of deposit mass from ref. 19 from six widely spaced sites along the dispersal axis (12 to 229 km from source) where aggregate sizes were noted in the field (Supplementary Table 1). Ideally, this extent should be based on observations at many more sites at many more distances from source, but with which frozen aggregates melt or are otherwise destroyed in harsh, high-altitude terrain precluded a larger data set. Nonetheless, our data set of ash aggregates is among the most detailed of any modern eruption yet documented. The GSD of aggregates contained within each of the three isopleths are assumed to follow a log-normal (Gaussian) distribution defined by a mean and s.d. from the measured values (that is, the maximum observed diameter was taken to represent the ~99th percentile). The assumption of a log-normal distribution is based on the detailed analysis of the aggregates within 12 km of Redoubt Volcano, which gives an excellent match to the Gaussian fit (linear R² = 0.91; see Supplementary Table 2).

For GSD3, we use the distribution calculated by Mastin et al.37 (their Table 2), which employed a volume-weighted average of 32 grain-size analyses using the VoroVolcanic module of the ATHAM’s bulk microphysical scheme describes the exchange of mass and energy between volcanic plumes and gas of initially uniform temperature25 (in this case, 575 K). Volcanic source parameters were derived from ATMOSPHERIC OBSERVATIONS detailed field observations (described in the following section). Atmospheric sounding data over Redoubt Volcano were interpolated from the 2.5-degree NCEP/NCAR Reanalysis 1 model45 (Supplementary Fig. 2). We used a stretched grid covering a domain of 100 × 100 × 30 km with 194 × 194 × 139 grid points. The maximum horizontal and vertical resolution of 50 m was centred at the volcano, stretching to 1.0 km (vertically) and 1.9 km (horizontally) at the model boundaries. Topography was interpolated onto the model grid from a 30-m resolution digital elevation model (Supplementary Table 1). The AT:

ATHAM’s bulk microphysical scheme describes the exchange of mass and energy between water vapour and four hydrometeors: cloud water, cloud ice, rain
and hail (see ref. 23 for full details). The hail tracer has a density of 700 kg m\(^{-3}\), which lies between that of true hail and lower-density graupel. There are 19 microphysical processes incorporated into the model, including condensation, evaporation, freezing, melting, sublimation, deposition, autoconversion and accretion. Hail growth is modelled by collection and freezing of rain and cloud water, and by deposition of cloud water on hail freezing. Velocities of hail and rain are taken from their volume mean radius, derived from artificial-parallel distributions, which depend on the mass concentration of each tracer. In contrast, the radii of cloud water and cloud ice are prescribed and assumed to be monodisperse. A simplification of the bulk microphysical scheme is that it does not include interactions between hydrometeors and volcanic particles, meaning they can coexist, but not combine. Therefore, processes of ice nucleation and ash removal by precipitation are not explicitly resolved in the model. To minimize these limitations, we do the following: (1) enable temperature-dependent statistical freezing of supercooled water beginning at 0°C (ref. 23), which is reasonable given that contact freezing of liquid droplets is initiated by ice particles at all temperature <0°C; and (2) use a coarser size distribution of volcanic particles representing the effective sizes after aggregation, based on measurements undertaken for this study (GSD4, Supplementary Table 3). Therefore, our simplifying assumptions are adequate for examining microphysical features in the volcanic plume, and identifying the regions in which hail-forming processes are thermodynamically favourable.

Constraints on volcanic source parameters. Eruptive event 5 from Redoubt Volcano was exceptionally well-monitored by the USGS Alaska Volcano Observatory and provided constraints on many of the properties required for modelling. The most sensitive inputs in our large-eddy simulation were related to eruption rate, including total erupted mass, eruption duration and initial velocity, followed by plume composition and plume temperature. Constraints on these key parameters are detailed below and summarized in Supplementary Table 4.

Erupted mass was calculated from the USGS data set of total eruptive index (ref. 23) as

\[ \text{mass} = \frac{1}{2} \times \text{mass rate} \times \text{eruption duration} \]

Some of the erupted mass was injected into the atmosphere during these initial 6 min, which is consistent with ash dispersal and therefore does not include any of the water or ice involved in the eruption. The initial temperature of the volcanic plume was calculated assuming a constant eruption rate of 1.53 \( \text{kg m}^{-2} \text{s}^{-1} \), and an additional 16.5 wt.% water incorporated from the glacier. We also included 0.5 wt.% \( \text{SO}_2 \) gas as a non-reactive, bulk tracer to account for all other magmatic volatiles. The initial temperature of the volcanic plume was calculated assuming that magma-water mixing cooled the erupted mixture during thermal equilibration at constant pressure, using a specific heat of magma of 1.000 kg K\(^{-1} \) K\(^{-1} \). We assumed that the erupting mixture (solid particles plus magmatic gas) was initially at 910°C (ref. 47) before mixing with 16.5 wt.% liquid water at 0°C, resulting in an equilibrated plume temperature of ~300°C.

Simulation of ash cloud dispersal using Ash3d. We used the Eulerian volcanic ash transport and dispersion model Ash3d (ref. 36), employing a model domain of 700 \( \times 700 \times 20 \text{ km} \) and constant grid resolution of 5 km. Rather than resolving the near-source dynamics of the volcanic plume, Ash3d specializes in long-range transport of ash particles in a time-changing, 3D wind field. Ash3d was not coupled to ATHAM for this study. To examine how ash dispersal from event 5 of Redoubt Volcano would have been different if aggregation had not occurred, we initialized an Ash3d simulation using the total GSD before and after aggregation (GSD3 and GSD4, Supplementary Table 3) and other input parameters from ref. 37 (including maximum plume height of 15 km and a vertical distribution of mass defined by a Suzuki distribution with a k-constant of 4). The wind field was derived from the 32 km North American Regional Reanalysis data set.

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