

Sedimentation of long-lasting wind-affected volcanic plumes: the example of the 2011 rhyolitic Cordón Caulle eruption, Chile

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Abstract

Sedimentation processes and fragmentation mechanisms during explosive volcanic eruptions can be constrained based on detailed analysis of grain-size variations of tephra deposits with distance from vent and Total Grain-Size Distribution (TGSD). Grain-size studies strongly rely on deposit exposure and, in case of long-lasting eruptions, can be complicated by the intricate interplay between eruptive style, atmospheric conditions, particle accumulation and deposit erosion. The 2011 Cordón Caulle eruption, Chile, represents an ideal laboratory for the study of long-lasting eruptions thanks to the good deposit accessibility in medial to distal area. All layers analysed are mostly characterized by bimodal grain-size distributions, with both the modes and the fraction of the coarse sub-population decreasing rapidly with distance from vent and those of the fine sub-population being mostly stable. Due to gradually-changing wind direction, the two sub-

populations characterizing the deposit of the first two days of the eruption are asymmetrically distributed with respect to the dispersal axis. The TGSD of the climactic phase is also bimodal, with the coarse sub-population representing 90 wt.% of the whole distribution. Polymodality of individual samples is related to size-selective sedimentation processes, while polymodality of the TGSD is mostly related to the complex internal texture (e.g. size and shape of vesicles) of the most abundant juvenile clasts. The most representative TGSD could be derived based on a combination of the Voronoi tessellation with a detailed analysis of the thinning trend of individual size categories. Finally, preferential breakage of coarse pumices on ground impact was inferred from the study of particle terminal velocity.

Keywords: Tephra; Particle Aggregation; Grain-size Bimodality; Total Grain-size Distribution; Particle Terminal Velocity; Inversion strategies

1. Introduction

Critical insights into eruption dynamics can be inferred from the study of the size of pyroclastic fragments, i.e. tephra (Kaminski and Jaupart 1998; Kueppers et al. 2006; Perugini et al. 2011; Rust and Cashman 2011; Dingwell et al. 2012). In addition, grain-size distributions and their variation with distance from vent can highlight important sedimentation processes that occurred during the eruption, e.g. particle aggregation and convective instabilities (Durant and Rose 2009; Durant et al. 2009; Brown et al. 2012; Carazzo and Jellinek 2012; Carazzo and Jellinek 2013). Finally, Total Grain-size Distribution (TGSD) represents a critical source parameter in numerical models for the description of tephra transport and sedimentation, used to compile both long-term hazard assessments and real-time forecasts crucial to mitigation strategies of volcanic risk. An accurate description of the amount of fine particles has also significant implications for the assessment of specific tephra hazards (e.g. impact on human and animal health, long-range threat to aviation, long-term ash resuspension) (e.g. Rose and Durant 2009). Nonetheless, the determination of TGSD is not straightforward due to: i) poor exposure of many tephra deposits, with very proximal and very distal areas being often of difficult access; ii) variable rate of deposit erosion; and iii) the complexity of integrating individual grain-size samples to derive the TGSD of the whole deposit (Bonadonna and Houghton 2005). TGSD of long-lasting eruptions is even more complicated by the combination of multiple pulses that are often not easy to correlate in the field and which are possibly associated with different mechanisms of magma fragmentation and clast dispersal.

Only a few TGSDs are available in the literature, and those available often lack either the fine or the coarse fraction, or both, and often represent a combination of multiple pulses (e.g. Rotongaio Ash (Walker 1981); Askja 1875 (Sparks et al. 1981); Mt St Helens

1980 (Carey and Sigurdsson 1982; Durant et al. 2009); Fuego 1974 (Murrow et al. 1980; Rose et al. 2007); Etna 2001 (Scollo et al. 2007); Spurr 1992 (Durant and Rose 2009); Eyjafjallajökull 2010 (Bonadonna et al. 2011); Pululagua 2450BP (Volentik et al. 2010); 4-ka Rungwe Pumice (Fontijn et al. 2011)). Here we investigate the grain-size and sedimentological features of the 2011 Cordón Caulle long-lasting eruption and assess the uncertainties associated with the determination of the TGSD. A detailed characterization of the stratigraphy and deposit features is presented by Pistolesi et al. (submitted).

2. Stratigraphy and eruption evolution

After about 41 years of repose, the Puyehue-Cordón Caulle volcanic complex (Central Andes) erupted on June 4, 2011, from a system of vents located about 1500 m above sea level (a.s.l.) on a NW-SE fracture of the Cordón Caulle system (Castro et al. 2013; Pistolesi et al. submitted). This resulted in the evacuation of ca. 4,000 people in Chile and, due to the prevailing winds, widespread disruption to various economic sectors and human activities in Argentina (Collini et al. 2013; Wilson et al. 2013). The eruption developed as a long-lasting rhyolitic activity with plume heights above the vent between ca. 9-12 km above the vent during the first 3-4 days, 4-9 km during the following week and <6 km after 14 June (GVP, Global Volcanism Program, <http://www.volcano.si.edu>; Siebert et al. 2010). The whole Cordón Caulle eruption was strongly affected by wind, with only the first plume on June 4 being classified as subplinian (Mass Flow Rate of the order of $\sim 10^7$ kg/s), with the rest of the eruption being small-moderate with the cumulative volume of the first 3-4 days characterized by a Volcanic Explosivity Index (VEI) of 5 (Bonadonna et al. 2015, in press). Thirteen tephra layers were identified, correlated and organized into four Units based on lithologic characteristics, grain-size features and discontinuity surfaces;

satellite images were then used to correlate each Unit with specific eruptive phases (Pistolesi et al. submitted).

Unit I (layers A to F) is a white to yellow lapilli-bearing deposit that represents the coarsest Unit of the whole eruption (with B, D and F being the coarsest layers). It was deposited towards the east-southeast on June 4 to 5 with negligible up-wind sedimentation. The six layers are characterized by a distinct, multiple reverse grading, and separated by planar discontinuities only evident in proximal-to-medial areas (<50 km from vent) mainly marked by sharp grain-size changes. Farther from vent (25-50 km), separation between the six layers becomes difficult to observe, and the deposit grades into a single thin (<10 cm), massive, fine lapilli to coarse ash bed. The second eruptive phase (June 5-6; Unit II with layers G and H) was marked by multiple wind shifts depositing the obsidian-rich layer G towards the north and the distinctive light orange-brown, pumice-rich layer H towards the east. Layer H was used as a marker to separate Units I and II from Unit III. The third phase started on the morning of June 7 depositing grey ash- to light-gray fine lapilli-bearing layers K1 to K5 towards the east (Unit III). This phase was also associated with the emission of large ballistic bombs in proximal area (Castro et al. 2013; Pistolesi et al. submitted). The fourth phase (Unit IV) deposited a thin layer of white fine ash around the eruptive vent up to the southern slopes of Puyehue volcano (i.e. 15 km from Cordón Caulle). No accurate time constraint could be made for the fourth phase based on the deposit. Tephra is characterized by the abundance of highly-to-extremely vesicular white pumice clasts, with minor banded and dense juvenile fragments, which largely increase after the first phase (Unit I); lithic clasts are present within Unit I and virtually absent in the following units. After the first three phases, emission of low-intensity eruptive plumes, mainly dispersing ash, characterized the

eruption. However, more intense pulses of ash emissions were observed in July, October, November 2011 and January 2012, which still caused disruption to aviation (Collini et al. 2013).

3. Methodology

The tephra deposit associated with the 2011 Cordon Caulle eruption was studied based on ca. 70 outcrops located between 1 and 240 km from the active vent (see Pistolesi et al. (submitted) for more details). Isopach maps for layers A-B, A-F, H and K2 were converted into isomass maps based on mean values of deposit density, i.e. 560 kg m⁻³ for A-B and A-F and 600 kg m⁻³ for H and K2 (Fig. 1) (see Pistolesi et al. (submitted) for details on isopach maps). Selected samples of layers A-B, A-F, H and K2 were dried and weighed in the lab and their volume was measured with a graduated cylinder in order to calculate the deposit density. Samples collected for grain-size analysis were dried and mechanically dry-sieved at half- ϕ intervals down to 0.5 mm ($\phi = -\log_2 D$, where D is the particle diameter in millimeters); the fine fraction (<0.5 mm) was characterized using a CILAS 1180 laser particle size analyzer. The combination of sieving and laser-diffraction data was validated on selected samples by overlapping data resulting from the two techniques for the fractions between 0.5-0.063 mm. Md ϕ and sigma values were calculated according to Inman (1952). 110 grain-size analyses of selected layers of Unit I, II and III were carried out. However, given that the single layers forming Unit I (A-F) were only distinguishable within 50 km from the vent, grain-size analyses of proximal outcrops were obtained based on the thickness and density of each layer in order to obtain a cumulative A-F grain-size distribution and investigate variations with distance from vent. Maps of lapilli fraction, coarse-ash fraction and fine-ash fraction and of individual ϕ categories of the cumulative

Unit I (layers A-F) were then compiled (Fig. 2 and Appendix 1). Grain-size analyses were also treated with the SFT (Sequential Fragmentation Transport; (Wohletz et al. 1989)) software to investigate single sub-populations. For almost all the analyzed samples, two main sub-populations were identified, here indicated as sub-population 1 (SP1; coarse grained) and sub-population 2 (SP2; fine grained), respectively. Componentry data, juvenile density, morphological and textural analysis, vesicularity and chemistry are reported and discussed in detail by Pistolesi et al. (submitted).

4. Variations of grain-size with distance from vent

In agreement with the isomass maps of the whole deposit (Fig. 1), a strong elongation along the main wind direction is also shown by the isomass maps of individual size classes, accompanied by a substantial lack of up-wind sedimentation (Fig. 2 and Appendix 1). Values of mass/area of Unit I are plotted on a log-axis versus distance from vent, instead of square root of contour area, in order to investigate better sedimentation trends and allow comparison with another deposit produced by a bent-over plume, i.e. the 1996 Ruapehu eruption, New Zealand (Bonadonna et al. 2005) (Fig. 3); the detailed comparison is presented in the discussion section. Lapilli clasts (diameter between 2-64 mm) are dispersed within the first 90 km from the vent with a load of more than 100 kg m⁻² within the first 40 km (Figs. 2a and 3a). Maximum accumulation of coarse ash is less significant than for the lapilli clasts, being just over 30 kg m⁻² within the first 45 km and decreasing to 5 kg m⁻² around 110 km from vent (Figs 2b and 3a). Fine ash represents the predominant size fraction beyond 100 km from vent (see for example the maps of lapilli and coarse ash; Figs. 2 and 3a). Isomass maps of individual ϕ categories confirm that the low abundance of coarse ash is mainly related to the scarce sedimentation of particles in

the 3 ϕ class (250-125 μ m), while ash in the categories 4 ϕ and \geq 5 ϕ categories (between 63-125 μ m and <63 μ m) clearly extend beyond 250 km from vent (Appendix 1). Multiple accumulation maxima are clearly visible only in the fine-ash map, with the two most significant being located around 20 and 100 km from vent. However, these multiple maxima are not visible in the total deposit map (Figs 2c and 3a).

Detailed analyses of individual samples highlight a rapid decay of mean grain-size for all layers from proximal (M ϕ =-3.5, -1.6 and -0.8 for A-F, K2 and H respectively) to distal samples (M ϕ =5 for A-F and K2), followed by a less steep decrease between 50 and 250 km (Fig. 4a). Between 50 and 120 km from vent, M ϕ values range between 0.5-5.2 for A-F and K2 and between 4.8-5.2 for H, reaching 5.4 at our most distal sample of A-F and K2 (i.e., 240 km from the vent) (Fig. 4a). Sorting does not show any clear trend with distance from vent, varying between 0.3-2.9 for A-F (very well sorted to poorly sorted), 0.7-2.4 for H (very well sorted to poorly sorted) and 1.2- 3.3 for K2 (well sorted to poorly sorted) (Fig. 4b). Many of these values stay at the upper limit of the fallout field of the M ϕ - $\sigma\phi$ of Fisher and Schmincke (1984). A detailed analysis of the sorting data shows how the best sorted samples (mostly for A-F and H) are those associated with the deposit south of the dispersal axis (Fig. 4b). Due to a clear increase of particle density with the decrease of particle size (Pistolessi et al. submitted), the increase of the M ϕ value with distance from vent is the main cause for the increase of deposit density (Fig. 5). In fact, deposit density varies between 330 and 920 kg m⁻³ between about 24 and 240 km from vent. However, the most rapid change (from 330 to 790 kg m⁻³) is observed between 24 and 50 km from vent, where M ϕ has the largest variability (Figs. 4a and 5). The correlation of deposit density with M ϕ is remarkably linear (i.e. exponential with the representative particle diameter in mm) (Fig. 5b).

Most of the sampled deposits (A-F, H and K2), with the exception of our most distal sample at 240 km from vent, are characterized by a clear grain-size bimodality, which can be deconvoluted into two main Gaussian sub-populations (SP1 and SP2) using the SFT software (Fig. 6). The weight fraction associated with the coarse sub-population (SP1) is >75% for most samples south of the dispersal axis (Fig. 6a), which are also characterized by the best sorting (Fig. 4b). The mode of SP1 decreases with distance from the vent from -2.6 to 1.8 ϕ (between 15-100 km from vent) and within the first 100 km it mostly coincides with Md ϕ values of the whole samples (Figs. 4a and 6b). The decay trend of SP1 is similar for all layers, with A-F being typically the coarsest and H the finest through the whole deposit (Fig. 6b). The mass loading of SP1 of all layers decreases regularly with distance from vent, going from about 455, 38 and 84 kg m⁻² at 15-20 km from vent (for A-F, H and , K2 respectively) to <10 kg m⁻² beyond about 80 km from vent for all layers and even to <0.1 kg m⁻² for layer H (Fig. 7a). In contrast, the mode of the fine-grained sub-population (SP2) remains nearly constant between 4.5-5.9 ϕ (Fig. 6b), while the decay of the mass loading is less regular than for SP1, and always remains between 0.1-15 kg m⁻² (Fig. 7b). The samples with the highest mass loading for SP1 are those south of the dispersal axis (Fig. 7a). Layer H is characterized by the smallest SP2, which varies between 0.3-1.4 kg m⁻² throughout the whole deposit (i.e., 15-104 km from vent; Fig. 7b). Nonetheless, beyond 50 km from the vent, SP2 of H samples is always more abundant (0.3-0.6 kg m⁻²) than the associated SP1 (<0.1 kg m⁻²).

Grain-size variations are asymmetrically distributed across the dispersal axis, the relative abundance of SP1 in the southern samples being larger than that of the northern samples (Fig. 8). This is particularly evident in samples from Unit I (A-F layers) and can

explain the poor correlation of $\sigma\phi$ with distance from vent (Fig. 4b), the relative amount of SP1 and SP2 (Fig. 6a) and the corresponding mass loadings (Figs. 7).

5. Distribution of terminal velocity

In order to investigate particle sedimentation, terminal velocity was determined for the coarse clasts (-4.5 to 0ϕ) that fell at a locality 15 km downwind from the vent along the dispersal axis (Fig. 9 and Appendix 2). First, the clasts were grouped into two different density categories based on the detailed componentry and density analysis carried out by Pistolesi et al. (submitted): a high-constant density category, and a variable size-dependent density category (for -4.5 to 0ϕ : 435 to 978 kg m^{-3} , layers A to F, Fig. 9b; 440 to 863 kg m^{-3} , layer H, Fig. 9e; 460 to 1016 kg m^{-3} , layer K2, Fig. 9h). The high-density category combines clasts with high-constant density ($2,700\text{ kg m}^{-3}$), i.e. lithic and obsidian fragments, while the variable size-dependent density category consists of juvenile fragments with variable vesicularity, i.e. banded pumice clasts, dense juvenile clasts and white pumice clasts (componentry classes defined as in Pistolesi et al. (submitted)).

Clasts in the range -4.5 - 0ϕ represent about 90 wt.% of the deposit of all samples considered at this location, practically coinciding with SP1 (Fig. 9). Componentry plots also show that high-density clasts are most abundant within the finest size categories (i.e. 0 to -3.5ϕ for A-F and 0 to -1.5ϕ for H and K2), with the coarsest categories being mostly free of high-density clasts (i.e. -4.0 to -4.5ϕ for A-F and -2 to -4ϕ for H and K2) (Figs. 9a,d,g and Appendix 2). Polymodality of the grain-size distribution largely disappears when clast size is converted into terminal velocity, which peaks at a single value for all layers. The peak value mostly depends on plume height, i.e. $\sim 20\text{ m/s}$ for A-F, $\sim 15\text{ m/s}$ for H and ~ 10 - 15

m/s K2 (maximum plume height of 14 km, 12 km and 10 km for A to F, H and K2 respectively).

It is also interesting to note how the spread of all velocity distributions is significantly narrower than that of grain-size. When both high-density clasts and variable-density clasts are normalized to 100 and plot together, it is clear how, individually, they cannot always be described by a Gaussian distribution (Figs. 9c,f,i and Appendix 2). In addition, the two density categories overlap in the central velocity classes, whereas the highest and the lowest velocity classes are only represented by high-density and variable-density clasts, respectively, with mostly no overlap on the tails (bins of Fig. 9). In particular, the distribution of the variable-density clasts (i.e. juvenile vesicular clasts) seems to be shifted by about one bin category towards the low-velocity values (based on 5 m/s binning; Figs. 9c,f,i and Appendix 2). This suggests an excess of clasts smaller than the modal size, probably related to clast breaking upon impact with ground. High-density clasts, less prone to break, do not show this feature, and their terminal velocity frequency distribution is generally much less peaked (more platykurtic) than that of variable-density clasts (Appendix 2). Based on the shift of the peak value of A-F from 20 m/s (high-density clasts) to 15 m/s (variable-density clasts) (i.e. reduction of mean diameter value from ~28 to ~13 mm; Fig. 9), we can derive that the diameter of variable-density clasts was reduced by about 50% due to clast breakage. Breakage seems to be much more effective for large clasts, as evidenced by the general lack of variable-density clasts with high terminal velocity.

6. Total grain-size distribution

The TGSD was determined for the climactic phase of the Cordón Caulle eruption based on different strategies: i) Voronoi tessellation (Bonadonna and Houghton 2005); ii) weighted average; and iii) mass of individual ϕ categories (Fig. 10 and Table1). The mass of individual ϕ categories was derived in three different ways: a) integration of the exponential and b) Weibull fitting of the isomass maps of individual ϕ categories of Appendix 1 (Pyle 1989; Bonadonna and Costa 2012); and c) inversion of values of mass/area of individual ϕ categories using the model TEPHRA2 following Volentik et al. (2010). Particles $<4\phi$ were not considered in the last technique, as size-selective processes, such as particle aggregation and convective instabilities, cannot be currently described by inversion strategies (Fig. 10b and Appendix 4). The Voronoi tessellation was applied to two data sets: the complete data set of all our A-F samples (dataset 1) and the combination between dataset 1 and six additional distal points (around 610 km from vent) derived from two isopach maps compiled soon after the eruption that account for the distal deposit (dataset 2) (see Appendix 3 for details). We have assigned to these six distal points a grain-size distribution based on the relative proportion of lapilli, coarse ash and fine ash as derived from Fig. 3a. In fact, the best fit of the three size fractions considered, indicates that around 610 km from vent the deposit mostly consists of fine ash (see Appendix 3). Nonetheless, even if six distal samples are reconstructed and considered in the calculation, the resulting grain-size does not change significantly (Fig. 10a and Table 1). Relative increase in fine ash for dataset 2 with respect to dataset 1 is ~ 4 wt.% (i.e., 13 wt.% and 9 wt.%, respectively; Table 1). Not enough grain-size data were available to determine the TGSD of layers H and K2.

All strategies result in polymodal distributions with a clear lack of particles in the 3ϕ category, which is always <1 wt.%. However, while the TGSD resulting from the Voronoi

276 tessellation, weighted average and inversion strategies are mostly bimodal with similar
277 $Md\phi$ and sorting values (Table 1), the TGSD derived from the mass associated with
278 individual ϕ categories are more complex and polymodal, regardless of whether the
279 associated mass is determined based on the exponential or the Weibull fitting. The TGSD
280 derived with the Voronoi tessellation strategy is slightly coarser than the weighted
281 average strategy because the Voronoi tessellation allows for a better representation of
282 individual outcrops even when the distribution is not uniform, as is the case for the
283 proximal samples from the Cordón Caulle eruption (e.g. Fig. 1). The TGSD derived with the
284 inversion is also coarser, mostly because the fine-ash fraction could not be considered in
285 the calculation (i.e. particle $<4\phi$). In all cases, TGSD shows two main sub-populations. The
286 modal value of the coarse sub-population is consistent for all datasets and strategies,
287 with the exception of the exponential and Weibull integrations (i.e. -1.9 to -2.5ϕ ; Table 1).

288 The advantage of the inversion on grain-size with respect to the inversion on
289 mass/area and the other strategies to derive TGSD is the derivation of the plume height,
290 which, in the case of the climactic phase of the Córdon Caulle eruption resulted in an
291 average best-fit value of ~ 14 km a.s.l. with most particles being released between ~ 9 - 10
292 km a.s.l., in agreement with observations (Appendix 4) (targeted inversion on mass/area
293 resulted in average plume height of ~ 13 km a.s.l.; Bonadonna et al. 2015, in press).
294 However, the Root Square Mean Error (RSME) minima associated with the best-fit values
295 of plume height for the climactic phase of the Córdon Caulle eruption are not as well
296 constrained as when applied to plumes developing in almost no wind (e.g. Pululagua 2450
297 BP Plinian eruption, Ecuador (Volentik et al. 2010), ~ 4 -ka Rungwe Pumice Plinian
298 eruption, Tanzania (Fontijn et al. 2011)). In fact, similarly to mass/area inversion
299 (Bonadonna et al. 2015, in press), the erupted mass of individual size categories is better

constrained than plume height (Appendix 4). This is probably due to the stronger influence of wind entrainment on the plume rise, which increases the complex interaction amongst input parameters.

7. Discussion

The 2011 Córdon Caulle eruption represents an ideal case study to investigate grain size and sedimentological features of long-lasting eruptions and to assess the complexities and uncertainties associated with the determination of the associated TGSD.

Particle sedimentation

As shown by our study, grain-size features vary significantly based on distance from the vent, eruptive style, intensity and atmospheric conditions (e.g. Figs. 3 to 8). Further complexities in sedimentation can be added by the presence of multiple sub-populations in the TGSD, as is the case for the climactic phase of the Cordón Caulle eruption. This has obvious implications on the long-term assessment of eruption impact, including impact on human and animal health, impact on vegetation, generation of secondary lahars and ash resuspension. Grain-size variation and mass accumulation with distance from vent can also be significantly affected by specific sedimentation processes, such as particle aggregation and convective instabilities. In order to investigate the general trend of particle deposition, we compare the sedimentation of the Cordón Caulle Unit I (plume height of 11-14 km a.s.l. and average wind speed along plume rise of $\sim 20 \text{ m s}^{-1}$; Bonadonna et al. 2015, in press) with the sedimentation from another eruption strongly

affected by wind for which detailed grain-size data are also available: the Ruapehu 1996 eruption, New Zealand (Fig. 3b).

The Ruapehu eruption was characterized by a smaller erupted mass, a slightly lower plume and a slightly higher wind speed than the climactic phase of the Cordón Caulle eruption (plume height of 8.5 km a.s.l. and average wind speed along plume rise of $\sim 24 \text{ m s}^{-1}$; (Prata and Grant 2001; Bonadonna et al. 2005)). However, the three main size fractions show a very similar thinning trend (i.e. lapilli, coarse ash and fine ash) (Fig. 3). In fact, even though the total A-F deposit can be fitted by at least two exponential segments on a semi-log plot of mass/area versus distance from vent, individual size fractions could be well fitted by single exponential segments with slopes that are similar to those of the Ruapehu deposits (slope values for Cordón Caulle/Ruapehu: $-0.108/-0.108$ for lapilli clasts, $-0.027/-0.023$ for coarse ash, $-0.004/-0.005$ for fine ash; Fig. 3). Due to the presence of an active lava flow emplaced during the eruption that could not be crossed during the field surveys, we could not collect data in the proximal downwind sector of Cordón Caulle ($<15 \text{ km}$ from the vent), precluding the possibility to recognize the most proximal exponential segments that can be identified at Ruapehu (Fig. 3b). The most distal break-in-slope for both cumulative thinning trends is associated with the intersection between the coarse-ash and the fine-ash trends (i.e., 110 km and 150 km from vent for Cordón Caulle Unit I and Ruapehu, respectively; Fig. 3). Due to a smaller erupted mass ($5 \times 10^9 \text{ kg}$), smaller accumulations are observed for Ruapehu at any distance from vent (Fig. 3b). In addition, probably due to the lower plume, sedimentation of lapilli clasts is mostly confined within the first 10 km for the Ruapehu deposit, which results in a different relative proportion of the size components at any distance from vent. As an example, the relative proportions of lapilli clasts, coarse ash and fine ash at about 40 km

from vent is 56%, 33% and 11% for Cordón Caulle A-F and 1%, 83% and 16% for Ruapehu (total accumulation being about 90 and 1 kg m⁻² for the two deposits, respectively). Fine ash becomes dominant for both deposits beyond 200 km from vent with a similar decay.

The similar deposit features shown by the Cordón Caulle Unit I and the Ruapehu eruptions, suggest that sedimentation from bent-over plumes is associated with similar thinning decay with distance from vent, even though the accumulation at any distance from vent is directly proportional to the total erupted mass and the relative fractions of lapilli clasts, coarse ash and fine ash, which might change depending on plume height (with higher plumes dispersing coarse particles further from vent). It is also important to bear in mind also that the Ruapehu eruption was mostly associated with a unimodal and coarser TGSD, and, therefore, the thinning trend of size fractions seems to be independent from source conditions. We stress how such a detailed analysis of the thinning trend of individual size categories can provide insights into the grain-size distribution of distal deposits (e.g. Fig. 3 and Appendix 3). As an example, such a critical analysis clearly shows how the deposit mostly consists of fine ash beyond 200 km, where the coarse ash is only about 0.5 kg m⁻² (15% of the total loading).

Grain-size bimodality

The layers analyzed in detail, i.e. A-F (4-5 June), H (6 June) and K2 (7 June) were all mostly characterized by bimodal grain-size distributions. Similarly, samples collected from the entire A-F sequence all have the same grain-size features, evidencing no significant changes through the sequence (Pistolesi et al. submitted). An interesting feature is the decrease in grain-size of the SP1 mode with distance from vent for all layers and the stable mode of SP2 around 4-6 ϕ (Fig. 5b), which has already been observed in other

tephra deposits (e.g. Montserrat, West Indies; St Vincent, West Indies; El Chichon, Chile; Mt St Helens, USA; Mt Mazama, USA (Brazier et al. 1983; Varekamp et al. 1984; Bonadonna et al. 2002; Durant et al. 2009; Brown et al. 2012; Durant et al. 2012). However, a more interesting aspect is the poorly variable mass loading ($0.1\text{--}15\text{ kg m}^{-2}$) of SP2 that does not show any obvious trend with distance from vent and with the mass loading of SP1 (Fig. 6b), although samples collected south of the dispersal axis generally have a lower mass loading with respect to samples from similar distances collected north of the axis. While the regular decrease of the SP1 mode with distance from vent suggests normal settling of individual clasts dominated by plume height and wind advection, the ubiquitous presence of a fine sub-population with a constant grain-size distribution is interpreted as an evidence of settling dominated by size-selective sedimentation processes (e.g. particle aggregation and/or convective instabilities) that may have affected particle fallout even 15 km from the vent.

Particle aggregates were only observed within the K1 deposit at a few sites close to Villa La Angostura (48 km from the vent) (i.e. millimetric structured accretionary pellets; Pistolesi et al. submitted), suggesting that fine ash mostly fell as very fragile aggregates whose structure was not preserved during impact with ground (Brown et al. 2012). Unfortunately, we could not sample the deposit in more proximal areas to verify if aggregation occurred at distances <15 km and, if so, in which form. Premature fallout of fine particles can also be associated with convective instabilities often observed during sedimentation of small-moderate eruption plumes, which could also enhance particle aggregation due to the increased particle concentrations within the resulting fingers (Bonadonna et al. 2002; 2005; 2011; Carazzo and Jellinek 2013; Manzella et al. 2015, in press). Convective instabilities are expected to have played an important role also during

the Cordón Caulle eruption, even though no clear pictures of downwind sedimentation during the climactic phase of the first few days are available to confirm this hypothesis. Considering that A-F, H and K2 layers were generated by plumes with different intensities, we conclude that aggregation and/or convective instabilities were persistent processes throughout the whole eruption, not significantly affected by small differences in plume height.

A persistent sub-population of fine particles generating bimodal grain-size distribution has also sometimes been reported as being generated by co-PDC plumes (Carey and Sigurdsson 1982; Bonadonna et al. 2002; Eychenne et al. 2012). However, Cordón Caulle PDCs had runout distances too short (i.e. ~ 10 km) and possibly volumes too small (ca. 10% of A-F total deposit; Pistolesi et al. submitted) to disperse a significant amount of fine ash up to 100 km from vent where bimodality is still observed (Pistolesi et al. submitted). In addition, PDCs were only observed on 4, 5 (with a runout of ~ 10 km; (SERNAGEOMIN/OVDAS 2011)), 8 and 14 June, whereas layers H and K2 also show bimodal distributions and they were produced on 6-7 June 2011. However, we do not exclude that part of the ash component in the proximal A-F deposit (generated on 4-5 June) could be a combination of co-PDC and main plume fallout.

Finally, grain-size bimodality also affects deposit sorting. In fact, sorting is typically improved by wind advection, with tephra deposits generated during low-wind conditions being usually poorly sorted. However, this is not the case for the Cordón Caulle eruption. As an example, the Pululagua 2450 BP Plinian eruption, Ecuador (Volentik et al. 2010) and the ~ 4 -ka Rungwe Pumice Plinian eruption, Tanzania (Fontijn et al. 2011) are characterized by sorting between 1.1-3.0 and 1.3-2.0, respectively, which are in the same range shown by layers A-F, H and K2 (i.e. 0.3-2.9, 0.6-2.4 and 1.2-3.3, respectively; Fig.

4b). For the two no-wind eruptions, sorting seems to improve with distance from vent (particularly for the Pululagua eruption) and the associated TGSD is mostly unimodal. Regardless of the strong wind advection, sorting at Cordón Caulle is not significantly better than for the Pululagua and Rungwe Pumice eruptions, most probably because of the bimodality of grain-size distributions at all outcrops (e.g. Fig. 6).

The sorting of the deposits of the climactic phase of the Cordón Caulle eruption was shown to be worst north of the dispersal axis due to a strong asymmetry of the distribution of the two sub-populations SP1 and SP2 (Fig. 8). Such an asymmetry was also observed for the small-moderate Mt St Helens eruptions (USA) on May 25 through August 7, 1980, which was explained as mostly due to distribution-rotation of wind direction with altitude, and variability in the duration, height, and qualitative density of eruption columns (Waitt et al. 1981). Similarly, the preferential accumulation of SP1 and SP2 south and north of the dispersal axis, respectively, can be related to the progressive wind shift towards the north that occurred at the end of June 5 associated with a lowering of the plume height (e.g. Bonadonna et al. 2015, in press; Pistolesi et al. submitted).

Secondary maxima of accumulation

Multiple maxima of accumulation are shown by the fine-ash fraction of the A-F deposit of Cordón Caulle, but not by the total deposit (Figs. 2, 3 and Appendix 1). Secondary maxima of accumulation of the total deposit have often been associated with particle aggregation (e.g. Mt St Helens 1980 (Carey and Sigurdsson 1982; Durant et al. 2009); Hudson 1991 (Scasso et al. 1994); Quizapu 1932 (Hildreth and Drake 1992)). Multiple maxima of accumulation were also observed in the fine-ash fraction of the deposit associated with the Ruapehu 1996 plume, New Zealand, for which particle aggregation was not so evident

in the deposit but that was also strongly distorted by wind advection (Bonadonna et al. 2005). However, in the Ruapehu case, these multiple maxima coincide with multiple maxima in the total deposit (Fig. 3b). Even though, both for the Cordón Caulle eruption and the Ruapehu eruption, aggregation processes were not evident in the associated deposit, secondary maxima of accumulation are clearly related to size-selective sedimentation processes that affect only particles $\leq 63 \mu\text{m}$. These processes are most likely related to particle aggregation and convective instabilities even when field evidence is missing. As an example, particle aggregation has been shown to generate fragile clusters that easily break on impact with the ground (e.g. Brown et al. 2012). Finally, considering that tephra deposits are not always investigated based on isomass maps of individual grain-size categories, we can conclude that the fine-ash fraction tends to produce secondary maxima of accumulation even when these are not shown by the total tephra deposit.

Total Grain-Size Distribution

Our results show how the Voronoi tessellation, weighted average and inversion strategy give more consistent results for TGSD than the strategies based on the mass calculated from empirical fits (i.e. exponential and Weibull fits). This is due to the poor distribution of mass/area points for individual ϕ categories that result in isomass maps with mostly ≤ 3 contours (Appendix 1). Four and five contours could be well identified only for -1 and 0ϕ . The mass associated with 3ϕ had to be calculated with the method of Legros (2000) as only one contour could be drawn. The cumulative mass derived with the exponential and Weibull method is about 60% and 75%, respectively, of the total mass derived from the isopach map of the cumulative deposit of Pistolesi et al. (submitted) (i.e., ~ 2.8 and

3.7x10¹¹ kg versus ~5x10¹¹ kg). In addition, the Voronoi tessellation for the derivation of TGSD is preferred with respect to the weighted average because it can better characterize non-uniform point distributions (as shown by the higher sensitivity to the addition of 6 distal points; Fig. 10 and Table 1) and to the inversion of individual size categories because this cannot be applied to the fine-ash fraction that is likely to be affected by size-selective sedimentation processes (e.g. particle aggregation). In addition, the application of inversion strategies to eruptions strongly affected by wind entrainment seems to be more complex than the application to strong plumes (see also Bonadonna et al. 2015, in press). The TGSD of the A-F deposit as derived with the Voronoi tessellation applied to the dataset 1 is clearly bimodal, with the two main sub-populations peaked at -2.5 and 6.2 ϕ , a large fraction of the coarse population (90 wt.%) and a significant low cumulative wt.% (0.8) of particles between 0.250-0.125 mm (i.e. 2-3 ϕ) (Table 1).

Polymodality of TGSD has already been observed for other tephra deposits (e.g. Montserrat Vulcanian and dome collapse explosions (Bonadonna et al. 2002); Mt St Helens 1980 (Carey and Sigurdsson 1982; Durant et al. 2009); Mt Spurr 1992 (Durant and Rose 2009); Tunguragua 2006 (Eychenne et al. 2012); El Chichon 1982 (Varekamp et al. 1984); Pinatubo 1991 (Wiesner et al. 2004); 1979 Soufriere St Vincent, Mazama 7000 a. (Brazier et al. 1983)), for which it could be explained as mainly due to the contribution of sedimentation from co-PDC plumes. However, as already mentioned above, we consider the contribution of co-PDC sedimentation to be very limited for the Cordón Caulle eruption. In addition, such a striking polymodality is in contrast, for example, with the TGSD of another rhyolitic Plinian eruption (β layer, Chaitén 2008 eruption, Chile; (Alfano et al. submitted)), which is characterized by one single population peaked around 6 ϕ , i.e. mostly coinciding with the fine population of the A-F deposit. Considering that both

eruptions have been studied in detail and the TGSD has been integrated over a similar deposit extension (up to about 600-800 km from source) and with the same methodologies, we believe that the striking differences are related to the actual fragmentation of the erupted material. In particular, it is important to bear in mind that about 80 wt.% of the β layer of the 2008 Chaitén eruption consists of non-vesicular material and that the associated vesicular juveniles are characterized by a unimodal vesicle size distribution (Alfano et al. 2012). Fragmentation of such homogeneous pyroclastic material is more likely to result in one single population than the fragmentation of polycomponent material characterized by juveniles with complex vesicle textures, such as that of Cordón Caulle Unit I (Pistolesi et al., submitted; Schipper et al., 2013). Given the abundance of white-pumice clasts in the A-F cumulative layer of Cordón Caulle, we conclude that the grain-size polymodality is mostly due to the associated complex texture. In fact, white-pumice clasts are characterized by the coexistence of spherical and nearly isotropic vesicles together with vesicles with highly deformed and convoluted shapes (Pistolesi et al. submitted), which, when fragmented, are likely to result in multiple distinct peaks of particle sizes.

Terminal velocity

Our detailed analysis of terminal velocity at the reference outcrop 15 km from vent has shown how particles deposited at a given location are peaked around a characteristic velocity that mostly depends on plume height and particle density (Fig. 9 and Appendix 2; to note that maximum plume heights were considered for all calculations and therefore particle terminal velocities are to be considered as maximum values). However, high-density and variable-density particles show different modal values for terminal velocity,

generally overlapping only in a few central velocity classes. We have interpreted such a shift of the moderately to highly vesicular, variable-density clasts towards the low velocity categories as due to an important effect of clast breakage on impact with the ground. Based on the shift of the peak velocity values, we estimated such a breakage to be about 50% of the original diameter of the variable-density clasts.

This has important implications both for the determination of the largest clasts and of the TGSD. In fact, our results clearly support the recommendation of the collection of largest lithics as supposed to largest pumices for the compilation of isopleth maps (Sparks et al. 1981; Bonadonna et al. 2013). In addition, given the absence of variable-density clast sizes corresponding to the largest velocities, and to the evident increment of clasts in the modal terminal velocity (evidenced by the frequency distribution of terminal velocities for these clasts, highly peaked on a modal class with terminal velocity values equal or slightly lower than those of lithic fragments), we suggest that the coarsest variable-density clasts are the most affected by breakage at impact.

Considering that tephra deposits of subplinian-Plinian eruptions are typically mostly composed of variable-density (juvenile) clasts (e.g. variable-density clasts of A-F are ~73% of the total deposit; Fig. 9 and Appendix 2), we can also conclude that TGSD derived from tephra deposits are very likely to be shifted towards lower size values, with a possible general underestimation of the coarse clasts contribution. Dufek et al. (2012) have already shown the relation between amount of fine ash and fragmentation depth due to collisional processes in the conduit. Here we show how additional breakage of lapilli clasts can occur on ground impact, inducing a shift in the calculated TGSD. This has important implication for the associated discussion of magma fragmentation and simulations of particle dispersal.

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539 *Deposit erosion and particle resuspension*

540 Long-lasting eruptions are characterized by a complex interplay between tephra
541 accumulation and deposit erosion depending also on local soil and atmospheric
542 conditions (e.g. rain, snow, wind, vegetation cover, soil moisture). As an example, due to
543 persistent strong winds, significant resuspension of the distal part the Cordon Caulle
544 deposit affected air quality and resulted in the closure of important airports even months
545 after the beginning of the eruption (e.g. 14-18 October 2011 (Wilson et al. 2013; Folch et
546 al. 2014)). These resuspension events have also been recognized in other ash-rich
547 deposits (e.g. Katmai, USA (Hadley et al. 2004); Mount St Helens, USA (Hobbs et al. 1983);
548 Hudson, Chile (Wilson et al. 2011); Montserrat, West Indies (Hincks et al. 2006);
549 Eyjafjallajökull, Iceland (Leadbetter et al. 2012; Thorsteinsson et al. 2012)) and can be
550 comparable to the dispersal of primary ash fallout in terms of dispersal area, with the
551 potential to cause widespread impact on agriculture, aviation, livestock and human health
552 long after a volcanic eruption and far away from the original eruptive vent.

553 The source term necessary to model and forecast resuspension events can only be
554 characterized by a detailed study of the tephra deposit that can be potentially
555 remobilized by strong winds. Local grain-size distribution is likely to be very different from
556 the TGSD of the eruption and can only be constructed based on a selection of critical
557 outcrops. As a result, detailed stratigraphic studies of tephra deposits and the physical
558 characterization of eruptive parameters becomes the first step to develop effective
559 mitigation strategies and land-use planning. Resuspension mostly affected layers K1-K5
560 (Unit III) of the 2011 Cordon Caulle deposit, which are the only layers that were found
561 remobilized during our last field survey (i.e. February 2013). In fact, comparison between

observations made during the first surveys (i.e. July 2011 and October-November 2011) with observations made in May 2012 and February 2013 show that erosion did not affect layers A to H. In addition, Layers K1-K5 are mostly characterized by particles with diameter $<250\text{ }\mu\text{m}$, which was considered by Folch et al. (2014) as a threshold for resuspension for the Cordon Caulle deposit. The total thickness of remobilized material was then $<0.5\text{ cm}$. In general, a thinning plot showing the trend of the fine-ash fraction (e.g. Fig. 3a) can be used to derive the approximate amount of material that can be remobilized at any given distance.

8. Conclusions

In this paper we focus on particle size and sedimentation processes during the first eruptive phases of the 2011 Cordón Caulle eruption (i.e., June 4-7, 2011). Based on our detailed field campaigns and analytical studies we can conclude that:

1) All layers analysed (i.e., cumulative layer A-F, H and K2) are mostly characterized by bimodal grain-size distributions, with the mode of the coarse sub-population (SP1) decreasing rapidly with distance from vent (from -2.6 to 1.8ϕ between 15-100 km from vent) and the associated mass fraction varying from about 500 kg m^{-2} (15 km from vent) to $<10\text{ kg m}^{-2}$ (beyond 100 km from vent). In contrast, the mode of the fine sub-population (SP2) remains stable between 4.5 - 5.9ϕ with a mass loading always between 0.1 - 15 kg m^{-2} .

2) The fine sub-population has been interpreted as being mostly associated with size-selective sedimentation processes (e.g. particle aggregation, convective instabilities) confirmed by the presence of multiple accumulation maxima associated with the fine-ash fraction (i.e. particles $<63\text{ }\mu\text{m}$). Considering the similarity of grain-size features and

trends for the three layers analysed, patterns of fine-ash sedimentation during the Cordón Caulle eruption can be interpreted as being independent of plume height and eruptive style.

3) The presence of multiple maxima of accumulation for the fine-ash fraction but not shown by the total deposit suggests that size-selective sedimentary processes play an important role in most eruptions even when multiple maxima are not shown by the total deposit.

4) The coarse and fine sub-populations of the A-F layers are asymmetrically distributed with respect to the dispersal axis, with SP1 being more abundant in the southern samples. This is probably related to the progressive shift of the wind direction towards the north, which occurred at the end of the deposition of Unit I associated with a lowering of plume height (June 5).

5) The rapid decrease of $Md\phi$ within the first 50 km is associated with a rapid increase of deposit density from from 330 to 790 kg m⁻³. The highest value of deposit density observed is 920 kg m⁻³ at 240 km from vent (A-F layers).

6) The combination of Voronoi tessellation with a detailed analysis of the thinning trend of individual size categories (e.g. lapilli clasts, coarse ash and fine ash) was considered to provide the most representative TGSD of the climactic phase of the Cordón Caulle eruption, which is characterized by two main sub-populations peaked at -2.5 and 6.2 ϕ , with the coarse sub-population representing about 90 wt.% of the whole distribution.

7) Detailed investigations have shown that the grain-size bimodality is not related to the lack of distal samples but to a significant lack of particles in the 3 ϕ category. The addition of distal samples (600 km from vent) only slightly increases (<5 wt.%) the amount of fine ash. We suggest that the shape of the TGSD record a primary

fragmentation process possibly related to the complex textural features of the highly vesicular magma.

8) The derivation of TGSD based on weighted average and on the mass of individual size categories as derived from empirical fits and analytical inversions were of more difficult application and did not provide exhaustive results. The analytical inversion could provide important insights into plume height, but should be used with caution when applied to eruptions strongly affected by wind entrainment.

9) Bent-over plumes of both Cordón Caulle (June 4-5) 2011 and Ruapehu 1996 eruptions are characterized by similar sedimentation patterns, with thinning of lapilli, coarse ash and fine ash being described by exponential segments with similar slope. However, the relative fraction of individual components and the position of the break-in-slope might vary mostly depending on erupted mass and plume height.

10) Particle sedimentation at different distances from the vent can be described by a Gaussian distribution of terminal velocity with a mode that mostly depends on plume height and particle density. Nonetheless, we have observed a shift between high-density clasts and variable size-dependent density clasts at a same outcrop, mainly due to preferential breakage of coarse vesicular pumices at impact with the ground. We conclude that TGSDs derived from tephra deposits rich in highly vesicular juvenile material can be biased towards fine fractions generated during deposition in relation to the physical and textural features of the clasts (vesicularity, thermal history, density).

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	Voronoi (DS1)	Voronoi (DS2)	Weighted average (DS1)	Weighted average (DS2)	Inversion on grain-size	Exponential fitting	Weibull fitting
Md ϕ	-1.9	-2.1	-1.9	-1.9	-1.8	-0.5	-0.7
$\sigma\phi$	2.4	2.7	2.0	2.0	2.1	3.5	3.8
wt% fine ash	8.7	12.9	6.9	7.1	ND	19.5	16.0
Mode of SP1	-2.2	-2.5	-2.1	-2.1	-1.9	-1.4	-1.5
Fraction of SP1	0.9	0.9	0.9	0.9	1.0	0.8	0.8
Mode of SP2	5.1	6.2	5.4	5.4	ND	ND	ND
Fraction of SP2	0.1	0.1	0.1	0.1	ND	ND	ND

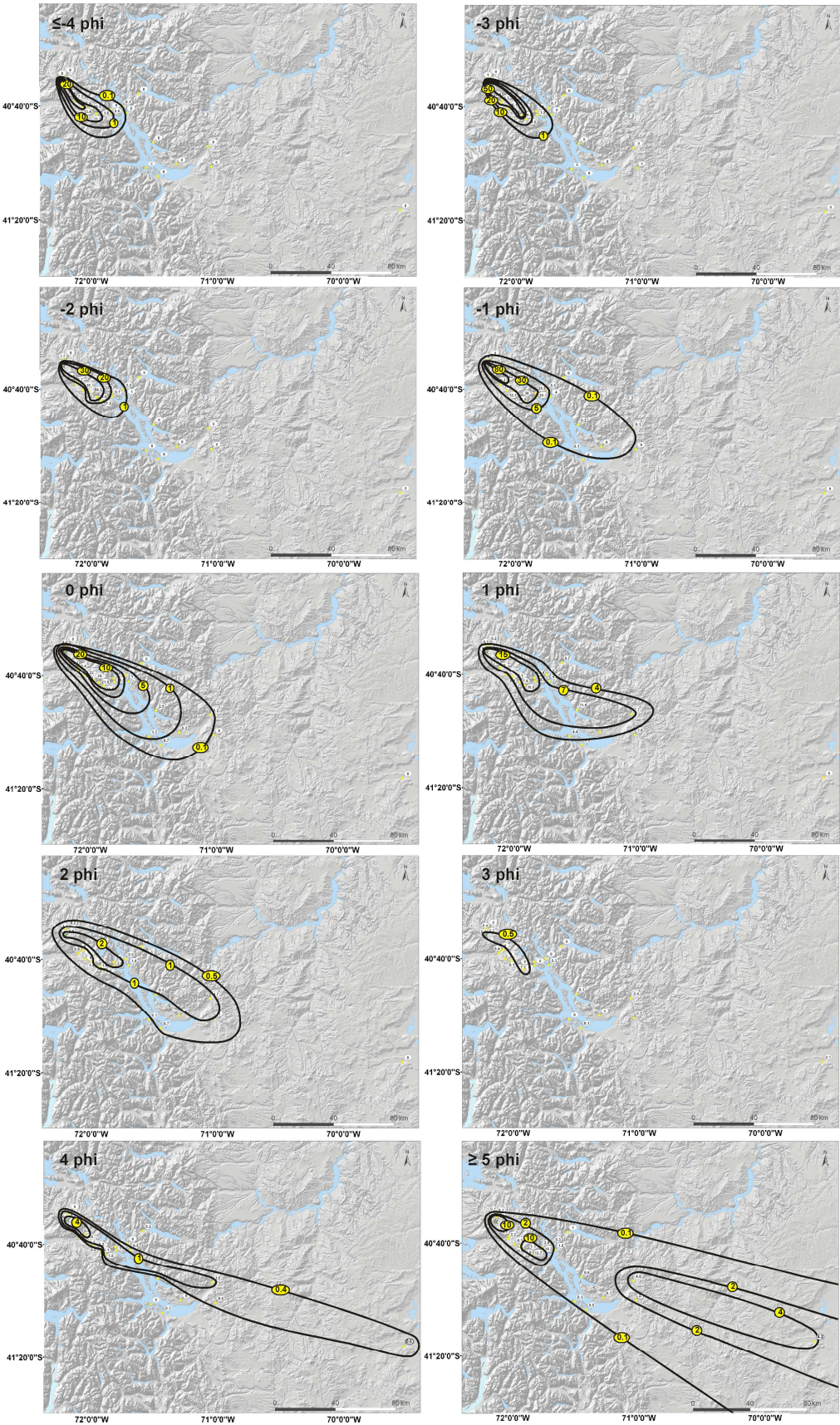
Table 1. TGSD grain-size parameters as derived with different strategies and for different datasets (see text for more details). Md ϕ and $\sigma\phi$ are from Inmann (1952). ND: not derivable. DS1 and DS2 are dataset 1 and 2 in Appendix 3. SP1 and SP2 are coarse- and fine-grained sub-populations, respectively.

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Appendix 1

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Isomass maps of individual ϕ categories for Unit I (cumulative A to F layers) in kg m^{-2}



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Appendix 2

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Calculation of particle terminal velocity

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Particle terminal velocity was calculated with the equation of Ganzer (1993) for the mean diameter within each half- ϕ size bin (assuming a Gaussian size distribution within each size bin) and averaged over sedimentation height (1 to 14 km a.s.l. for A-F, 1 to 12 km a.s.l. for H and 1 to 10 km a.s.l. for K2; maximum plume heights were considered for all phases). Sphericity was kept at 0.9 for both clast categories (e.g. Folch 2012). Particle density was measured for individual clast sizes by Pistolesi et al. (submitted) (see Figs 9b,e,h). In Fig. A2-1 we show results for individual layers A to F.

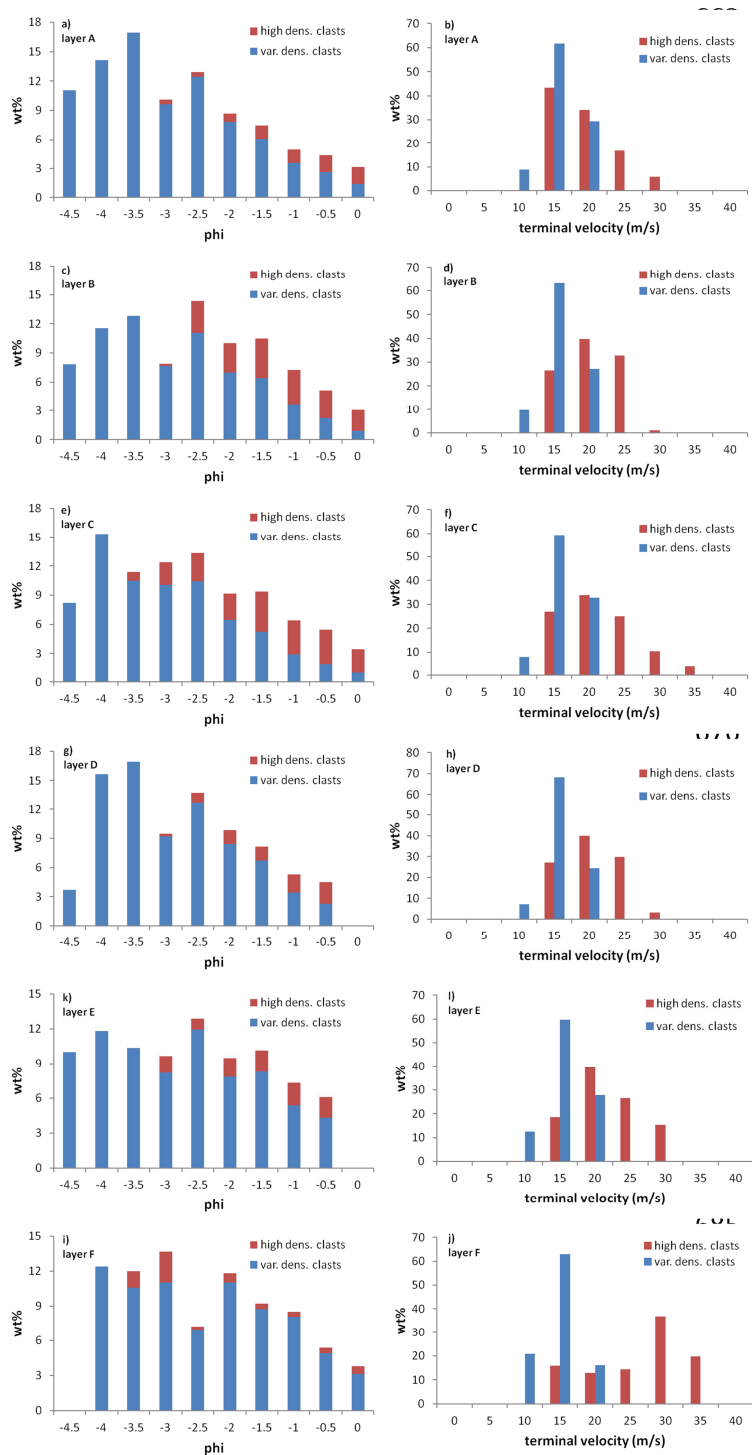


Fig. A2-1. Componentry (left column) and terminal velocity (right column) of tephra clasts ranging from 0 to -4.5 ϕ (i.e. 1-22.6 mm) for layers A to F at a locality about 15 km from vent on the dispersal axis for Unit I. See also Fig. 9 for results of the other layers.

Appendix 3

Calculation of the total grain-size distribution of Unit I

In order to account for the missing distal data in the derivation of the total grain-size distribution of the A-F cumulative layer (i.e. Unit I), we have derived the mass/area of 6 points about 600 km from vent from the isopach maps compiled soon after the eruption by Gáitan et al. (2011) and the INTA (2011). Based on these two maps that also describe the distal deposit, we assigned to the 6 distal data a thickness of 0.1 cm (i.e. 0.8 kg m^{-2} based on our most distal value of deposit density, i.e., 834 kg m^{-3} ; Fig. 5) and the grain-size shown in Fig. A3-1. The two maps (based on dataset 1 and 2) are compared in Fig. A3-2.

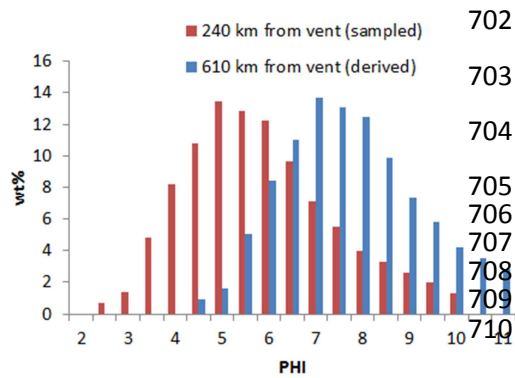


Fig. A3-1. Grain-size distribution derived for the distal points (610 km from vent; blue histograms) based on the distribution of our most distal sample at 240 km from vent (red histograms) and on the proportion of lapilli, coarse ash and fine ash as derived from the best-fit equations in Fig. 3a, main text (i.e. Gaussian distribution with mode of 7ϕ and composed of 99% fine ash).

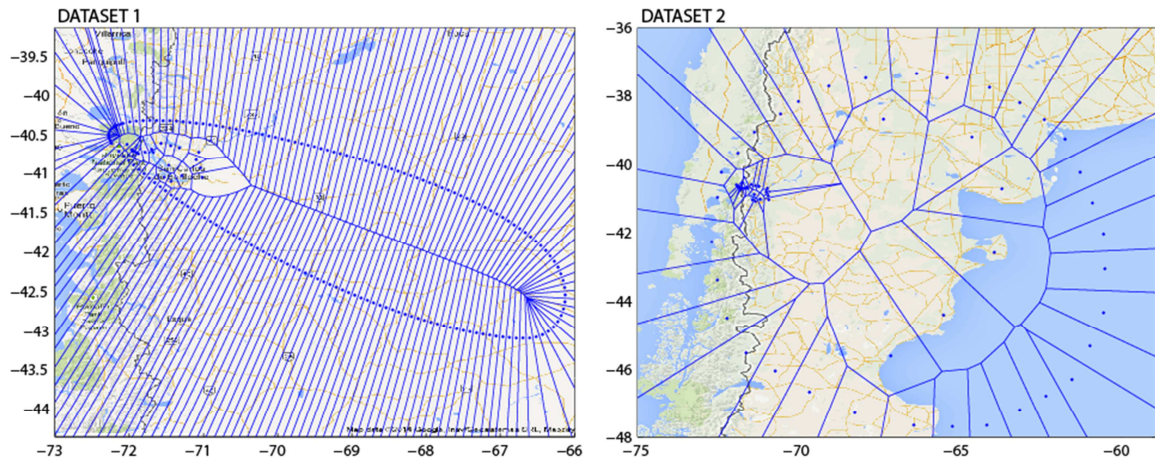
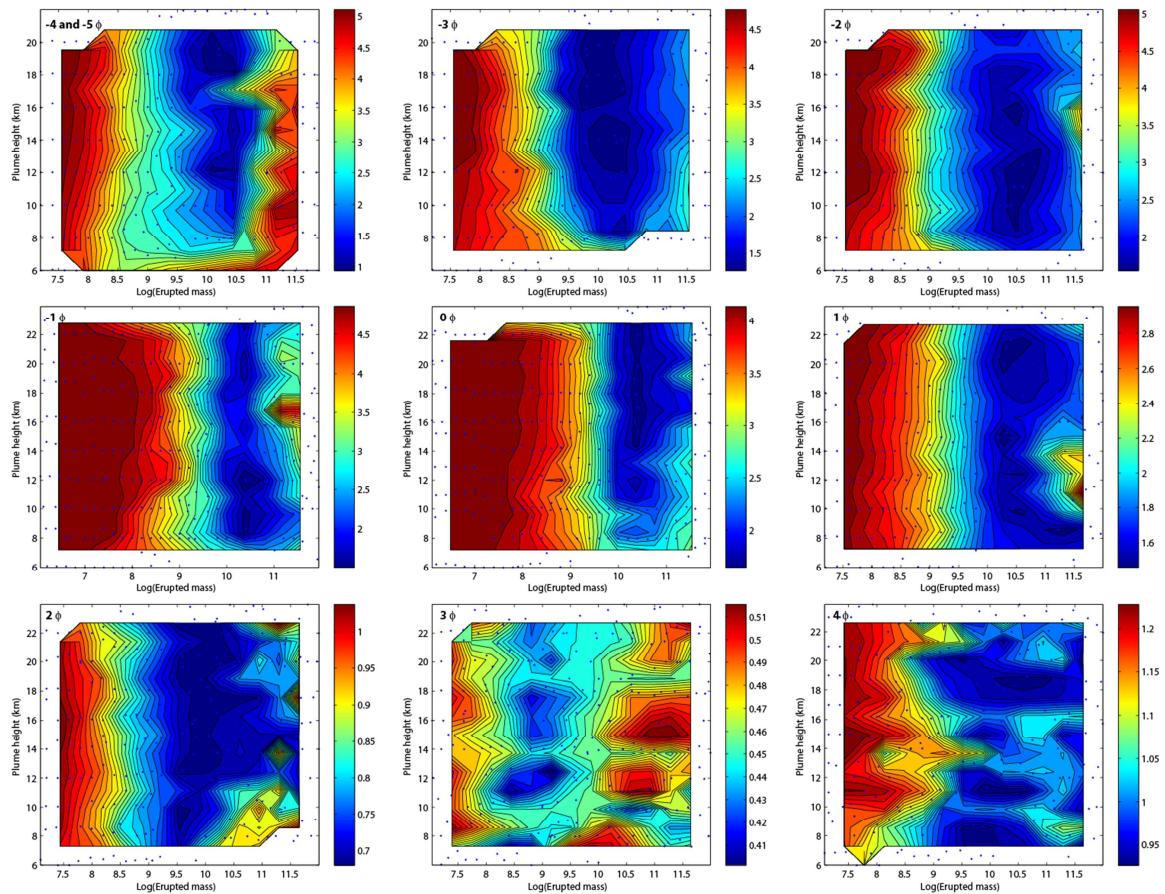


Fig. A3-2. Maps showing the Voronoi triangulation and the zero line used for the determination of the total grain-size distribution based on the Voronoi tessellation method of Bonadonna and Houghton (2005) and using the application of Biass and Bonadonna (2014) for: a) data-set 1 (i.e. samples collected in our field surveys) and b) data-set 2 (data-set 1 combined with six distal points derived from the isopach maps compiled soon after the eruption by Gáitan et al. (2011) and the INTA (2011)).

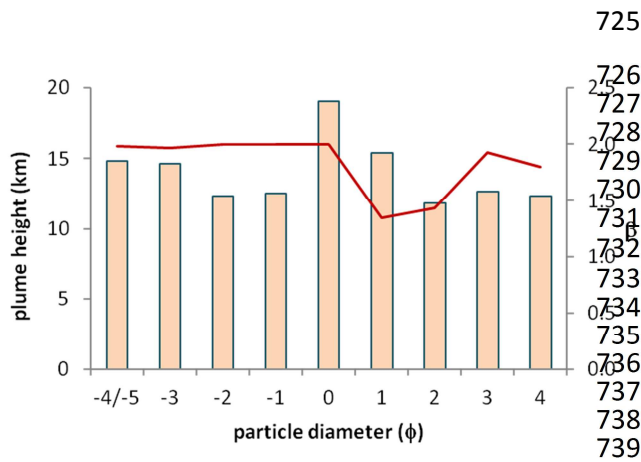
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Appendix 4
Inversion on grain-size using the model TEPHRA2



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Fig. A4.1 Plot of Log(erupted mass) vs. plume height (km) (above sea level) showing the minimum values (dark blue) of the goodness-of-fit measure (Root Mean Square Error, RMSE) associated with individual ϕ size categories (indicated in each plot) of the climactic phase of Córdon Caulle eruption (i.e. Unit I).



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Fig. A4-2. Best-fit plume height above sea level as derived from inversion on individual grain-size categories (average value=13.9 km). The mass along the eruptive plume in the model TEPHRA2 is described by a beta function characterized by two parameters, α and β (Connor et al. 2015, in press). For simplicity, in these inversion calculations α is kept constant (i.e. 3) and β is left vary between 0.5 and 2 (i.e. particles are mostly released at the top of the cloud) and 2 (i.e. particles are released at a height which is 60-70% the total height). Average best-fit value for the climactic phase of Córdon Caulle eruption is 1.8 (i.e. particles are mostly released at a height which is 70% the total height).

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