Dear Dr. Tesche:

Below you will find the colleague reviews to this paper, along with our response. Reviewer comments are in black. Responses in *blue italics*.

Appended below the comments and responses is a copy of the manuscript text with changes tracked. The revised manuscript with figures is provided as a separate file.

We hope you find these responses adequate to merit publication.

Sincerely,

Larry Mastin

# Folch Review

 The parameterization scheme (section 3.2, lines 206---201) seems somehow arbitrary and could be better justified. On the other hand, if the TPSD is discretized on 1Φ intervals all the fines aggregate except for Φ=3, for which 50% of particles aggregate. This seems rather simplistic. To what extend the results depend on this choice? What if discretization is performed at 0.5Φ intervals and/or the limits are extended beyond Φ=4 (e.g. Φ=5 or 6)? Could the best---fit values (i.e. the conclusions of the paper) depend substantially on this?

We have rewritten Section 3.2 to explain more clearly why we chose our parameterization scheme. It was based on experimental field observations of grain sizes that aggregate under different circumstances. Some explanation was already in an appendix, which has been deleted and its material moved into the main text. Figure A1 was also moved into the main paper and is now Figure 2.

In Section 3.2 (lines 250-255) we added a few sentences indicating the effect on results of different choices of the aggregation-size threshold. The main effect is to alter the mass that contributes to the secondary thickness maximum by several percent to tens of percent.

 The authors find "optimal" values of aggregate density of about paggr=600 kg/m3 consistent with (but denser than) previous studies and observations. It is important to mention that this is also a consequence of the settling velocity model chosen. Note that, for fine particles, the Wilson and Huang model gives smaller settling velocities than other fits (see e.g. Figure 1 in Folch 2012; Journal of Volcanology and Geothermal Research 235---236, 96–115). In other words, other velocity model using a smaller aggregate density would give exactly the same fit...

Actually, rho\_agg was chosen, rather than being obtained by optimization as we did with mu\_agg and sigma\_agg. We chose 600 kg/m3 because it was toward the middle of the very large range of densities observed for aggregates. We have added a paragraph to the end of Section 3.2 explaining this. We also now emphasize that this choice may lead to an over- or underestimate of aggregate sizes. Our objective in this study is not to constrain the size of real aggregates, but to find a combination of parameters that can successfully replicate observed deposits. We now make this point in the first paragraph of the Discussion section.

In Section 3.4 we also added a brief section point out that our results are dependent on the fall model chosen.

• Figure 2 is misleading because (at the beginning of the paper) gives the impression that only one aggregated bin is considered, contradicting the text. It would be much clearer if the distribution of aggregates is shown as an inset.

Yes, you're right. We have modified Figure 2 (now Figure 3) to show inset histograms of aggregate sizes.

• Line 241---242. Values of settling velocity for a given particle strongly vary with height. Are these values at sea level or averaged?

As stated in the text and in the caption to Figure 5 (now 6), the fall velocities are averages. Specifically, they were averages of calculations made at 1-km intervals in the atmosphere, from 0 to 15 km. We now say this in the figure caption.

• It is unclear to me how the modeled "dispersal axis" is obtained and why topography causes the oscillations observed in Figures 10---13.

The dispersal axes in Figures 10-13 (now 11-14) are determined by finding the ground cell in each row (for Figs. 10, 11) or column (Figs. 12-13), with the highest mass load. The algorithm that finds this cell reads from an ASCII output file that give mass load at each cell center, in kg/m2, to three decimal places. At distal locations, the maximum load along a row or column may not be much greater than the precision of the output, causing a jagged appearance when spurious cells are picked. We have added an explanation to the caption of the new Fig. 11 that now explains the calculation.

• Line 320. Typo (And)

# Corrected—thanks.

• Figure 1 and lines 322---327. The fact that diffusion can be ignore and still obtaining a reasonable fit is because the (Eulerian) model adds numerical diffusion. It is difficult to extract conclusions from here since this strongly depends on the numerical scheme, different from model to model.

Good point. At the end of Section 4.1 we have added a sentence noting that these results may be different in other models or model configurations.

• Line 428. "hundreds to thousands"? sure?

Changed to "many"

• Lines 424 to 444 in the discussion are rather speculative but interesting. I understand that the proposed "empirical" aggregation scheme would hold to model the finer aggregates (i.e. formed during transport), not for the larger aggregates (mm size) formed in the plume. That would explain why so different eruption conditions end up with similar mean and dispersal. Right?

If I understand your point, you seem to be suggesting that perhaps we're able to match these four deposits with similar aggregate sizes in part because we're excluding other, near-source processes, that could produce a more complicated and disparate outcome. If that's your point, I agree. It doesn't appear that you are suggesting that anything needs to be changed in this passage.

# di'Michieli Vitturi Review

The paper presents a study of the modelling of volcanic ash in the atmosphere, with a particular focus on the effect of ash aggregation on depositional pattern. Several eruptions are investigated in order to find the parameters controlling aggregation, which give best fits of the deposits. To this aim, the authors employed Ash3d, an Eulerian model that calculates tephra transport and deposition through a 3-D, timechanging wind field.

Despite the differences in the magnitude and styles of the eruptions studied, the parameters describing ash aggregates are found to be similar for all the events.

The phenomenon investigated is interesting and very relevant for the volcanic hazard associated with ash dispersal in the atmosphere and it presents important novelties for operational model forecast. For this reason, I think that the manuscript falls into the scope of Atmospheric Chemistry and Physics and it is scientifically sufficiently sound to be published, once some points detailed below are clarified, in particular concerning the way the grain size distribution has been discretized.

• Lines 174-179. While in most of the literature the Suzuki relation is described as the distribution of mass in the column, in the original paper it is defined as "probability density diffusion". This probability is related to the mass concentration of particles leaving the column at height z in the unit time, and it is different from the concentration of particles along the column.

Thank you for pointing this out. We have modified the text to indicate that we are using a modified version of the Suzuki equation, and that we are using this formula as a simple parameterization of mass distribution with height, with no attempt to relate it to physical process. The difference between a probability density function (which would not apply to our Eulerian model) and a function defining mass distribution in the column seems minor to me, unless I am misunderstanding something.

• Lines 189-193. In Wilson and Huang a, b and c are the principal axial lengths and not the semi-axes, and the values were measured for more than 155 particles. I am also not sure that the average value of the shape factor of 0.44 is reported in the Wilson and Huang paper.

*Thank you.* This was a typo, not an error in our calculations. We have corrected it in the text (3<sup>rd</sup> paragraph of Section 3.1).

You're right that the average shape factor of 0.44 was not reported in Wilson and Huang. We used their data to calculate an average shape factor. We have reworded the last sentence in the penultimate paragraph of section 3.1 to make this clearer.

• Section 3.2. It is not clear to me the choice of the bins for the discretization of the TPSD. Why bins of 0.5phi are used for the non-aggregated particles and bins of 0.1phi are used for the aggregated? If the settling velocity and the depositional process is sensitive to bins of 0.1phi for the aggregates, I think this should be true also for the non-aggregated particles.

Bins of 0.5 phi or coarser were used for the non-aggregated particles based on what was available in the published literature for these deposits. The finer, 0.1 phi bins were used for aggregates because, as shown in Figs. 6 and 9, where the aggregates land is highly sensitive to aggregate size, for the rather

narrow range of sizes and densities that would put fine ash at medial distances. For non-aggregated grains, this high sensitivity is only true for particles ~50-100 microns, as illustrated in Fig. 6. Most particles of this size have already aggregated. We have added a paragraph to Section 3.4 pointing out these constraints.

It is also reported that aggregates are described by a Gaussian size distribution, but the amount of fine ash assigned to different size bins, reported in Table 4, is not representative of a Gaussian distribution. The values should be computed using the error function:

# F(mu+x sigma)-F(mu-x sigma) = erf(x/sqrt(2))

You're totally right (gasp!); our distribution is not strictly Gaussian. And the values of sigma\_agg were inaccurate for the distributions given. We have modified the values in Table 4 and re-run all the simulations so that truly Gaussian distributions are represented. In the caption to Table 4 we also describe exactly how these values are derived (i.e. using a Gaussian formula). This change required us to re-derive Figures 10-13, 15, 16, all the supplementary figures. It also changed the results slightly, requiring slight rewording in the Results section.

• Section 3.3. I think that the first and third indexes, defined in Table 3, should not have the square root (exponent ½).

# Thank you. This has been changed, and the error indexes recalculated.

• Section 3.4. Aggregate size. Why is the range for sigma\_agg so small? Is it supported by observations or experiments? This doubt is also due to the results, showing a small sensitivity of the results with such a small range.

The small range that we use is a consequence of the high sensitivity between aggregate size and distance traveled (Fig. 6). For each simulation, we wanted to use a size distribution such that the range of distances traveled between the smallest and largest aggregates was a few hundred kilometers, as illustrated in Fig. 9. This limited the range of aggregate sizes to tenths of a phi unit. Broadening the size range would have caused a large fraction of aggregates to deposit outside the range of distances we were studying. This point was made in Section 3.4.

However to accommodate this concern, we have slightly broadened our range of sigma\_agg values. The sigma\_agg value is still small (0.3 phi), but larger than previously. When calculated properly using a Gaussian best-fit, our old maximum sigma\_agg value was 0.12.

With this new analysis, we can show that almost none of the optimal fits in Fig. 10 occur at the maximum value of sigma\_agg, suggesting that the range is now large enough to include the optimal value. Also, a perusal of the supplementary figures shows that, when sigma\_agg=0.3, the secondary thickness maximum is broader and less thick than observed, for example, at Mount St. Helens (Fig. S028) and Ruapehu (Fig. S128).

If we had compared the model result with more proximal sample locations, it is likely we would have obtained a wider optimal range of aggregate sizes. We chose not to include more proximal locations because the indexes we used, particularly delta^2, can be overwhelmed by proximal sample points, since their importance is directly proportional to the absolute value of the difference in mass load between the model and the measured deposit. Proximal deposition also involves processes such as hail-forming

aggregation or fallout from the vertical column, that are not accurately simulated in a widespread fallout model like Ash3d. Finally, if we had included these proximal sample locations, the optimal aggregate-size distribution would probably not have produced a secondary thickness maximum, because it would have been optimizing to fit the proximal deposit. The secondary thickening is a key feature of three of these deposits. Not reproducing it would have yielded an unacceptable result in our opinion.

## We have substantially revised Section 3.1 and now emphasize these points in that section.

• Section 4.1. It is not clear why some points are excluded from the analysis in Figure 10b and 10c. In the caption it is written that for panel (b) "grey dots lay outside the range of downwind distances covered by trend lines in Fig. 6", and are excluded from the calculation of Delta^2. I don't understand why the trend lines are involved in the point-by-point index, and also why Figure 6 should be used.

Values of Delta<sup>2</sup> can be dominated by differences in proximal locations, where mass per unit area is greatest, and where processes such as fallout from the vertical column are not accurately simulated. Therefore we exclude these proximal points from the calculation. At the beginning of Section 3.4 we note that we ignore proximal fallout, but perhaps didn't do an adequate job explain why. We have modified the explanation of the point-by-point method in Section 3.3 to add this explanation.

Also for panel (c) the caption is not clear, referring to Delta<sup>2</sup>\_area, while the figure is reporting a value for Delta<sup>2</sup>\_downwind.

# Thanks for pointing out this typographical error. It's now corrected.

In any case, I think that the criteria to exclude points from the measures of the fit should be discussed more in the main text.

## I think the above-mentioned changes to Section 3.3 address this.

• Lines 322-325. It is stated that adding turbulent diffusion "visually improve the fit". For this reason, I think it would be useful to quantify how much the fit is improved, through the different statistical measures of fit presented in the paper.

We tried this, and found that delta^2 actually shows a worse fit for the MSH case when diffusion is turned on! Apparently, the improved fit on the margins of the deposit is more than offset but poorer fit along the dispersal axis. We will note that in the last paragraph of section 4.1.

It is also interesting to note that the numerical results seems to show a diffusion in the results, and this is probably due to a numerical diffusion associated with the Eulerian approach. Is it possible to quantify or discuss the effects such diffusion, in relation with the grid-size?

## I'm not sure. At the moment, I can't think of how this would be done.

• The choice to neglect diffusion in the model is justified by the decrease in run time from 30 to 10 minutes for operational conditions. It would be interesting to compare this time with the characteristic timing of the depositional process.

This might be beyond the scope of the paper, but an interesting problem.

# Neri Review

The manuscript aims to investigate the role and effect of particle aggregation in explosive eruptions. This is done by using a numerical model of ash dispersal and by adopting a simple parametrization of the aggregation process. Optimal parameters of such model are then derived by optimizing the comparison between model predictions and deposit evidence. The underlying hypothesis is that the effect of aggregation may be accounted for by a simple modification of the original grain---size distribution at source.

Based on the results and analysis presented the above hypothesis appears quite well justified. This is actually quite surprising given the wide range of eruptive conditions considered and the complexity of the aggregation process. On this basis, the study appears able to provide a first---order approximation of the effect of particle aggregation by simply modifying the grain---size distribution at source. This is quite relevant for improving the accuracy of operational ash dispersal models.

I found the study very interesting, well---presented and certainly worth of publication after minor revision. The organization of the manuscript, as well as the figures and tables, are clear and informative. I suggest to further investigate just a few points listed below in order to make the outcomes of the study and its presentation even more robust and effective. A few minor technical points are also listed.

# Main points:

Section 3.1, lines 189---193: the Authors assume a constant particle shape factor for all particles and eruptions considered (except for the aggregates). This is probably a quite important assumption that should be acknowledged and commented given the main sensitivity of the dispersal process to such a parameter (see e.g. Scollo et al., JGR 2008; Bagheri et al., Pow. Tech. 2015; Pardini et al., JGR 2016). This is also quite evident from Fig. 6 where the shape factor of the aggregates has been varied. A similar assumption has been made for the density of the aggregates which, as explained in the text, also varies largely (lines 244---246). A brief discussion of the implications of these assumptions could be appropriate.

Thank you for this observation. A similar point was raised in A. Folch's review, and it shows we need to emphasize that our objective is to see whether "standard" values of these parameters (even if locally unrealistic) can successfully match observations. During an eruption, these values cannot be scrutinized and there is a need to have a set of standard values that are known to work well in reproducing observations. We have added a short paragraph to the end of section 3.4 emphasizing this point, and also a couple of sentences to the first paragraph of the Discussion section. Also in the first paragraph of the Discussion section, we emphasize that our results depend on the specific inputs chosen.

 Section 3.4, lines 258---261: the justification of the range of particle aggregate size and distribution (standard deviation) does not appear sufficiently clear as reported in the text. Why the assumption that most deposits fall in the region of interest is able to constrain the size of the aggregate? We assume that most aggregates fall in the region of interest because studies suggest that most erupted mass, probably >90%, falls to form a recognizable deposit rather than transporting farther downwind as a distal cloud. This is now mentioned in Section 3.4, when describing constraints on aggregate size

Is this valid/assumed just for the MSH case (Fig. 8) or for all the four eruptions?

We use the observations from Mount St. Helens to derive these constraints, but assume it applies to all four eruptions. We now state this explicitly in Section 3.4, when describing constraints on aggregate size.

Also the extension of the mapped area is not clear.

It is the area shown in the new Fig. 9. This is now mentioned in Section 3.4, in the paragraph describing aggregate size.

This key point should be better explain to me for both the mean and the standard deviation values. In particular the range of the standard deviation appears very narrow (i.e. 0.1---0.3) given the uncertainties involved and the results obtained, which, in some cases, indicate optimal values close to, or larger than, 0.3 (see Fig. 9).

In fact, the standard deviation of aggregate sizes was even narrower than we stated. After calculating a proper Gaussian best-fit using the size distribution of our previous manuscript, the maximum value of sigma\_agg was only 0.12, not 0.3. We have revised the aggregate-size distributions as shown in Table 4, using 4 values now (sigma\_agg=0, 0.1, 0.2, 0.3). We don't think a wider distribution is justified, for two reasons

(1) using sigma\_agg=0.3, too much fine ash flows out of the model domain. For example, in the Mount St. Helens case, even for the optimal value of mu\_agg (2.4), only 75% of the erupted deposit lands within the mapped area. For mu\_agg=2.5, 2.7, and 2.9, the values drop to 70%, 60%, and 48%. These values are too low to be realistic, in our opinion.

(2) For the Mount St. Helens case, the value of sigma\_agg=0.3 produces a secondary thickening that is broader and more diffuse than observed, for example, in fig. 11c. For the other deposits, the secondary thickening is not sufficiently well defined to judge. We make this point in the first paragraph of Section 4.1.

• Section 4, lines 264---266 and Tab. 4. The way the aggregates are assigned to the various bins is not clear. In particular the distributions shown in Tab. 4 are not Gaussian as expected. This should be corrected.

This point was also made in Mattia di'Michieli Vitturi's review. We have changed the distribution of aggregates in Table 4 so that they are now Gaussian, and added an explanation of how they were calculated to the Table 4 caption. This change required us to run all the simulations again and derive new results.

It would be also interesting to see the effect of a different discretization of the Phi units of the aggreagates so to estimate the effects on the optimal parameters (units of 0.2 or 0.5 Phi instead of 0.1).

I think this point is addressed in our response to the previous bullet.

Section 4, lines 291---297: in the description of the consistency with other studies the Authors could also mention the studies of Biass et al. (NHESS 2014) and Barsotti et al. (BV 2015) on Icelandic volcanoes and Vesuvius, respectively, that show similar optimal parameters of the aggregation process.

Thanks for reminding me of these papers. I now cite them in the first paragraph of the Discussion section. It's interesting that they use a wider range of aggregate sizes, based partly on observations by Bonadonna et al. (2002) at Montserrat. We found a smaller size range to be optimal in this study. But the difference may lie partly in the fact that we optimized the fit to sample locations at distances of several tens to hundreds of kilometers. Including more proximal sample points may have resulted in a wider optimal aggregate-size range. I now point that out when citing them.

# Minor technical points:

- Line 374: D should be 3x10^2. *Corrected—thanks!*
- Line 850: Fig. 6 should be replaced by Fig. 3? *Yes, thanks. Should now be Fig. 4.*
- Line 853: Fig. 7a should be replaced by Fig. 3a? *Yes, corrected. (now Fig. 4a)*

Augusto Neri

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6	Adjusting particle-size distributions to account for
7	aggregation in tephra-deposit model forecasts
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## 21 Abstract

22 Volcanic ash transport and dispersion models (VATDs) are used to forecast tephra deposition 23 during volcanic eruptions. Model accuracy is limited by the fact that fine ash aggregates, altering patterns of deposition. In most models this is accounted for by ad hoc changes to model 24 25 input, representing fine ash as aggregates with density  $\rho_{\rm agg}$ , and a log-normal size distribution 26 with median  $\mu_{agg}$  and standard deviation  $\sigma_{agg}$ . Optimal values may vary between eruptions. 27 To test the variance, we used the Ash3d tephra model to simulate four deposits: 18 May 1980 28 Mount St. Helens; 16-17 September 1992 Crater Peak (Mount Spurr); 17 June 1996 Ruapehu; 29 and 23 March 2009 Mount Redoubt. In <u>158-192</u> simulations, we systematically varied  $\mu_{agg}$ 30 and  $\sigma_{\rm agg}$  , holding  $\rho_{\rm agg}$  constant at 600 kg m<sup>-3</sup>. We evaluated the fit using three indices that 31 compare modeled versus measured (1) mass load at sample locations; (2) mass load versus 32 distance along the dispersal axis; and (3) isomass area. For all deposits, under these inputs, the 33 best-fit value of  $\mu_{agg}$  ranged narrowly between ~2.13-2.57 (0.2320-0.18mm15mm), despite 34 large variations in erupted mass (0.25-50Tg), plume height (8.5-25 km), mass fraction of fine 35 (<0.063mm) ash (3-59%), atmospheric temperature, and water content between these eruptions. This close agreement suggests that aggregation may be treated as a discrete process that is 36 37 insensitive to eruptive style or magnitude. This result offers the potential for a simple, computationally-efficient parameterization scheme for use in operational model forecasts. 38 39 Further research may indicate whether this narrow range also reflects physical constraints on 40 processes in the evolving cloud.

### 41 Keywords

volcanic ash, volcanic plume, ash clouds, aerosols, aggregation, volcanic eruptions, tephradeposition

### 44

## 45 1 Introduction

Airborne tephra is the most wide-reaching of volcanic hazards. It can extend hundreds to
thousands of kilometers from a volcano and impact air quality, transportation, crops, electrical
infrastructure, buildings, water supplies, and sewerage. During eruptions, communities want
to know whether they may receive tephra and how much might fall. Volcano observatories

50 typically forecast areas at risk by running volcanic ash transport and dispersion models

51 (VATD). As input, these models require information including eruption start time, plume

52 height, duration, the wind field, and the size distribution of the falling particles. Of these inputs,

53 the particle size distribution is perhaps the hardest to constrain.

54 Particle size (along with shape and density) determines settling velocity, which controls where

55 particles land in a given wind field. For different eruptions, the total particle-size distribution

56 (TPSD) can vary. Large eruptions produce more fine ash than small ones for example; and

57 silicic eruptions produce more than mafic (Rose and Durant, 2009). The TPSD is difficult to

estimate (e.g., Bonadonna and Houghton, 2005); hence estimates exist for only a handful of

deposits. And even in cases where the TPSD is known, that TPSD, entered into a dispersionmodel, will not accurately calculate the pattern of deposition (Carey, 1996).

61 This inaccuracy results from the fact that complex processes, not considered in models, cause 62 particles to fall out faster than theoretical settling velocities would predict. These processes include scavenging by hydrometeors (Rose et al., 1995a), gravitational instabilities that cause 63 dense clouds to collapse en masse (Carazzo and Jellinek, 2012; Schultz et al., 2006; Durant, 64 2015; Manzella et al., 2015), and aggregation, in which ash particles smaller than a few hundred 65 66 microns clump into clusters. The rate of aggregation, and the type and size of resulting 67 aggregates, depend on atmospheric processes such as ice accretion, electrostatic attraction, or 68 liquid-water binding whose importance varies from place to place.

69 Although one VATD model, Fall3d, calculates aggregation during transport for research studies 70 (Folch et al., 2010; Costa et al., 2010), no operational models consider it. Instead, aggregation 71 is accounted for by either setting a minimum settling velocity in the code (Carey and 72 Sigurdsson, 1982; Hurst and Turner, 1999; Armienti et al., 1988; Macedonio et al., 1988), or, 73 in the model input, adjusting particle size distribution by replacing some of the fine ash with aggregates of a specified density, shape, and size range (Bonadonna et al., 2002; Cornell et al., 74 75 1983; Mastin et al., 2013b). These strategies will probably prevail for at least the next few 76 years, until microphysical algorithms replace them.

These adjustments are mostly derived from *a posteriori* studies, where model inputs have been adjusted until results match a particular deposit. It is unclear how well the optimal adjustments might vary from case to case. For model forecasts during an eruption, we need some understanding of this variability. This paper addresses this question, using deposits from four well-documented eruptions. We derive a scheme for adjusting TPSD to account for

aggregation, optimize parameter values to match each deposit, and then see how much these

83 optimal values vary from one deposit to the next.

### 84 2 Background on the deposits

85 The IAVCEI Commission on Tephra Hazard Modeling has posted data from eight well-mapped eruption deposits, available for use by modeling groups to validate VATD simulations 86 87 (http://dbstr.ct.ingv.it/iavcei/). Of these, we focus on eruptions that lasted for hours (not days); where the TPSD included at least a few percent of ash finer than 0.063mm in diameter; and 88 89 where data were available from distal (>35 km) sample locations. Four eruptions met these 90 criteria: the 18 May 1980 eruption of Mount St. Helens, 16-17 June 1996 eruption of Ruapehu, 91 and the 16-17 September and 18 August 1992 eruptions of Crater Peak (Mount Spurr), Alaska. 92 The August Crater Peak eruption was already studied using Ash3d (Schwaiger et al., 2012) and 93 therefore not included here, reducing the total to three. To these we add event 5 from the 23 94 March 2009 eruption of Mount Redoubt, Alaska. Although an Ash3d study was made of this 95 event (Mastin et al., 2013b), aggregation has been unusually well characterized in recent years (Wallace et al., 2013; Van Eaton et al., in press). 96

Below are key observations of these events. Deposit maps are shown in Fig. 1, digitized frompublished sources.

99 1) The 18 May 1980 deposit from Mount St. Helens remains among the best documented of any in recent decades (Durant et al., 2009; Sarna-Wojcicki et al., 1981; Waitt and Dzurisin, 100 101 1981; Rice, 1981). This 9 hour eruption expelled magma that was dacitic in bulk composition 102 but contained about 40% crystals and 60% rhyolitic glass (Rutherford et al., 1985). The 103 eruption start time (1532 UTC) and duration are well documented (Foxworthy and Hill, 1982); 104 the time-changing plume height was tracked by Doppler radar (Harris et al., 1981) and satellite 105 (Holasek and Self, 1995) (Table 2). The deposit was mapped within days, before modification 106 by wind or rainfall, to a distance of ~800 km and to mass load values as low as a few hundredths 107 of a kilogram per square meter (Sarna-Wojcicki et al., 1981). Estimated volume of the fall 108 deposit in dense-rock equivalent (DRE) is 0.2 km<sup>3</sup> (Sarna-Wojcicki et al., 1981) based on what 109 fell in the mapped area. A TPSD was estimated by Carey and Sigurdsson (1982) and later by 110 Durant et al. (2009) to contain about 59% ash <63 um in diameter (Table S1), with a modal 111 peak in particle size that coincided with the median bubble size of tephra fragments (Genareau 112 et al., 2012). Some fine ash may have been milled in pyroclastic density currents on the 113 afternoon of 18 May and in the lateral blast that morning. A secondary maximum in deposit

114 thickness in Ritzville, Washington (~290 km downwind) was inferred by Carey and Sigurdsson 115 (1982) to have resulted from fine ash aggregating and falling en masse, perhaps as the cloud 116 descended and warmed to above-freezing temperatures (Durant et al., 2009). Wind directions 117 that were more southerly at low elevations combined with elutriation off pyroclastic flows in 118 the afternoon to feed low clouds, producing a deposit that was richer in fine ash along its 119 northern boundary than in the south (Waitt and Dzurisin, 1981; Eychenne et al., 2015). 120 Aggregates sampled by Sorem (1982) in eastern Washington consisted mainly of dry clusters 121 0.250 to 0.500 mm in diameter, containing particles <0.001mm to more than 0.040mm in 122 diameter, though no aggregates were visible in the fall deposit except at proximal locations (e.g. 123 Sisson (1995)). The eruption began under clear weather conditions. Clouds increased 124 throughout the day. Some precipitation in the form of mud rain was noted within tens of 125 kilometers of the vent (Rosenbaum and Waitt, 1981), probably due to entrainment and condensation of atmospheric moisture in the rising plume. But no precipitation was recorded 126 127 at more distal locations during the event.

128 2) The 16-17 September 1991 eruption from Crater Peak, Mount Spurr, Alaska, was the 129 third that summer from this vent. The eruption start time (0803 UTC September 17) and 130 duration (3.6 hours (Eichelberger et al., 1995)) were seismically constrained. The maximum 131 plume height, measured by U.S. National Weather Service radar (Rose et al., 1995b) increased 132 for the first 2.3 hours and then fluctuated between about 11 and 14 km above mean sea level 133 (MSL) until the plume height abruptly decreased at 1110 UTC. The andesitic tephra consisted 134 of two main types; tan and gray, which were both noteworthy for their low vesicularity (~20-135 45%) and high crystallinity (40-100%) (Gardner et al., 1998). The deposit was mapped rapidly after the eruption (Neal et al., 1995; McGimsey et al., 2001) to a distance of 380 km and mass 136 137 loads around 0.050 kg m<sup>-2</sup>. This deposit displays a weak secondary thickness maximum 260-138 330 km downwind. Durant and Rose (2009) derived a TPSD for this deposit, estimating about 139 40% smaller than 0.063 mm. Milling in proximal pyroclastic flows that accompanied this 140 eruption (Eichelberger et al., 1995) could have contributed fine ash. The eruption occurred at 141 night under clear skies (Neal et al., 1995).

3) The 17 June 1996 eruption of Ruapehu produced a classic weak plume that was modeled
by Bonadonna et a. (2005), Hurst and Turner (1999), Scollo et al. (2008), Liu et al. (2015), and
Klawonn et al. (2014), among others. The main phase involved two pulses, one beginning 16

 $145 \qquad \text{June at 1910 UTC and lasting 2.5 hours, and the second at 2300 UTC and lasting approximately}$ 

146 1.5 to 2 hours. Ash-laden plumes reached to about 8.5 km altitude above MSL based on satellite infrared images (Prata and Grant, 2001). The deposit was mapped out to the Bay of Plenty 147 148 (190 km), sampled at 118 locations to mass loads less than 0.01 kg m<sup>-2</sup>, and yielded a total mass 149 of about 0.001 km<sup>3</sup> DRE (Bonadonna and Houghton, 2005). Ejecta consisted mainly of scoria 150 containing 75% glass and 25% crystals, with glass containing about 54 wt% SiO<sub>2</sub> (Nakagawa 151 et al., 1999). A TPSD estimate based on the Voronoi tessellation method (Bonadonna and 152 Houghton, 2005) suggested that ash <0.063 mm composed only about 3% of the deposit. A 153 minor secondary thickness maximum was constrained by mapping at about 160 km downwind 154 (Bonadonna et al., 2005) (Fig. 1c). Although some witnesses at distal locations observed loose, millimeter-sized clusters falling, no aggregates or accretionary lapilli were present in the 155 156 deposit (Klawonn et al., 2014). The eruption was not accompanied by significant pyroclastic 157 density currents and occurred during clear weather.

158 4) Event 5 of the 23 March 2009 eruption of Redoubt Volcano, Alaska erupted through a 159 glacier and entrained a variable amount of water into a high-latitude early-spring atmosphere. 160 It began at 1230 UTC, lasted about 20 minutes on the seismic record (Buurman et al., 2013), 161 and sent a plume briefly to about 18 km as seen in both National Weather Service NEXRAD 162 Doppler radar from Anchorage, and a USGS mobile C-band radar system in Kenai, Alaska (Schneider and Hoblitt, 2013). Within a few days after the eruption, the deposit was mapped 163 164 by its contrast with underlying snow in satellite images (NASA MODIS), and sampled for mass 165 load and particle size distribution at 38 locations, at distances up to ~250 km and mass loads as 166 low as 0.01 kg m<sup>-2</sup> (Wallace et al., 2013). During Ash3d modeling of this eruption, Mastin et 167 al. (2013b) found that wind vectors varied rapidly with both altitude and time, making the 168 dispersal direction highly sensitive to both the plume height (which varied from ~12 to 18 km 169 during the 20-minute eruption) and the vertical distribution of mass in the plume. In the deposit, 170 Wallace et al. (2013) described abundant frozen aggregates with size decreasing with distance 171 from the vent, from about 10mm at 12 km distance. Schneider et al. (2013) attributed the high 172 (>50 dBZ) reflectivity of the proximal plume in radar images, and a rapid decrease in maximum 173 plume height over a period of minutes, to formation and fallout of ashy hail hydrometeors in 174 the rising column. Van Eaton et al. (2015) combined analysis of the aggregate microstructures 175 with a 3-D large-eddy simulation to show that the ash aggregates grew directly within the 176 volcanic plume from a combination of wet growth and freezing, in a process similar to hail

177 formation.

178 These eruptions vary from weak (Ruapehu) to strong (Redoubt) plumes, from mid-latitude (St.

Helens, Ruapehu) to high-latitude (Spurr, Redoubt), from dry (Ruapehu) to relatively wet
(Redoubt), from basaltic andesite (Ruapehu) to dacite (St. Helens), and from ~3% to 59% ash

181 <0.063 mm in diameter. Inferred aggregation processes range from dry (Ruapehu) to wet within

182 the downwind cloud (St. Helens), to liquid+ice in the rising column (Redoubt).

## 183 3 Methods

### 184 3.1 The Ash3d model

We model these eruptions using Ash3d (Schwaiger et al., 2012; Mastin et al., 2013a), an Eulerian model that calculates tephra transport and deposition through a 3-D, time-changing wind field. Ash3d calculates transport by setting up a three dimensional grid of cells, adding tephra into the column of source cells above the volcano, and distributing the mass in the column following the probability density function of Suzuki (Suzuki, 1983), modified by Armienti et al. {Armienti, 1988 #4546}

191 
$$\frac{dQ_m}{dz} = Q_m \frac{k^2 (1 - z / H_v) \exp(k(z / H_v - 1))}{H_v [1 - (1 + k) \exp(-k)]},$$
 (1)

where  $Q_m$  is the mass eruption rate,  $H_v$  is plume height above the vent, z is elevation (above the vent) within the plume, and k is a constant that adjusts the mass distribution. Suzuki {Suzuki, 1983 #2071} defines this function as a "probability density of diffusion" of mass from the column as particles fall out. Here we regard it as a simplified parameterization of mass distribution with no implication for physical process.

197 At each time step, tephra transport is calculated through advection by wind, through turbulent 198 diffusion, and through particle settling. For wind advection, simulations of Mount St. Helens, 199 Crater Peak, and Redoubt use a wind field obtained from the National Oceanic and Atmospheric 200 Administration's (NOAA's) NCEP/NCAR Reanalysis 1 model ("RE1") (Kalnay et al., 1996). 201 For the Ruapehu simulations we used a local 1-D wind sounding, which gave more accurate 202 results. The RE1 model provides wind vectors on a global 3-D grid spaced at 2.5° latitude and 203 longitude, and 17 pressure levels in the atmosphere (1000-10 hPa), updated at 6-hour intervals. 204 Ash3d calculates turbulent diffusion using a specified diffusivity D (Schwaiger et al., 2012, Eq. 205 4). *D* is set to zero for simplicity, though later we show the effect of different values of *D*.

206 Settling rates are calculated using relations of Wilson and Huang (1979) for ellipsoidal particles. 207 Wilson and Huang define a particle shape factor F=(b+c)/2a, where a, b, and c are the 208 maximum, intermediate, and minimum diameters of the ellipsoid respectively. Wilson and 209 Huang measured a, b, and c for 155 natural pyroclasts. The From data published in Wilson and 210 <u>Huang, we calculate an</u> average F of their measurements was 0.44, which we use in our model. 211 For aggregates we use F=1.0 (round aggregates).

212 Other model inputs include the extent and nodal spacing of the model domain; vent location 213 and elevation; the eruption start time, duration, plume height, erupted volume, diffusion 214 coefficient D, and a series of particle size classes and associated densities. The size classes 215 may represent either individual particles or aggregates. These input values are given in Tables 216 1 and 2.

#### 3.2 Adjusting particle size distributions to account for aggregation 217

218	In deriving and particle size adjustment scheme we found it necessary to prioritize the type(s)	Formatted: Indent: First line: 0
219	of processes and products we wish to replicate. The rate and extent type of ash aggregation are	
220	sensitive to changes inknown to vary with both eruptive conditions and background	
221	meteorology. Despite the complexity of the process, field studies and laboratory experiments	
222	have highlighted key spatial and temporal controls. For example, lLarge aggregates, including	
223	frozen accretionary lapilli, tend to-form near the volcanic-source and and are particularly	
224	abundant in phreatomagmatic eruption-deposits (Van Eaton et al., 2015; Brown et al., 2012;	Field Code Changed
225	Houghton et al., 2015). These. They are associated with with precipitation forming processes	
226	occurring as particles collidinge in moist, turbulent updrafts within a rising plume rising above	
227	the volcanic vent or ground-hugging density currents (Fig. A1Fig. 2) or an elutriating ash cloud.	
228	Field measurements indicate that nThese near-source aggregates commonly exceed 1 cm	
229	diameter (Wallace et al., 2013; Swanson et al., 2014; Van Eaton and Wilson, 2013). In contrast,	Field Code Changed
230	the low-density aggregates that produced the Ritzville Bulge, 230 km downwind from Mount	
231	St. Helens, are thought to have been triggered by mammatus cloud instabilities (Durant et al.,	
232	(2009)). A as the cloud descended-to warmer atmospheric levels, warmed, the increasing	Field Code Changed
233	proportion of liquid water increased the rate of aggregation and falloutand ice melted into liquid	
234	water (red line, Fig. A1Fig. 2). These types of distal aggregates tend to be smaller than a	
235	millimeter, and forming in the downwind cloud up to hundreds of kilometers downwind from	
236	source (Sorem, 1982; Dartayat, 1932). At Mount St. Helens and perhaps other places,	
237	investigators found evidence for both large, wet, proximal accretionary lapilli (Sisson, 1995)	
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238	and distal, dry aggregates (Sorem, 1982). The latter type deposited over a larger area, involved	
239	a greater fraction of the total erupted mass, and affected a greater population. Thus it is the	
240	process whose deposits we wish to reproduce.	
241	Aggregation is also a highly size-selective process. The threshold size below which most	
242	particles aggregate and above which they don't varies with moisture and electrical charge,	
243	ranging from several tens of microns under dry conditions, to hundreds of microns when liquid	
244	water is present (Gilbert and Lane, 1994; Schumacher and Schmincke, 1995; Van Eaton et al.,	
245	$\underline{2012}$ ). Our aggregation scheme is too crude to distinguish the threshold size as a function of	
246	atmospheric conditions, hence we use a broad range such that:	
247	$\underbrace{For \ \phi \ge = 4, all \ ash \ aggregates}_{==}$	
248	For $\phi \leq =2$ , no ash aggregates.	
249	For 4> $\phi$ >2, the mass fraction that aggregates varies linearly with $\phi$ from 1 (when $\phi = 4$ ) to 0	
250	(when $\phi=2$ ).	
251	The TPSD used to model these four eruptions are listed in Table S1 and illustrated as gray bars	
252	in Fig. 3. Particle sizes that do not aggregate according to this scheme are illustrated as black	
253	bars. We assume that the aggregates collect into clusters having a Gaussian size distribution of	
254	mean $\mu_{agg}$ , and standard deviation $\sigma_{agg}$ (insets, Fig. 3). For deposit modeling, we ignore the	
255	small fraction of the erupted mass that goes into the distal cloud, typically a few percent (Dacre	C
256	et al., 2011; Devenish et al., 2012).	
257	In our study, the aggregated ash mostly deposits as a secondary thickness maximum. Different	
258	choices of a threshold size for particle aggregation would influence the mass building the	
259	secondary maximum. For Mount St. Helens, about 10% of the erupted mass lies between $\phi=2$	
260	and $\phi$ =4. For Spurr, Ruapehu, and Redoubt, the percentages are 28%, 6% and 11%. These	
261	values reflect the variability in mass of the secondary maximum that could result from different	
262	choices of the aggregation-size threshold.	
263	Aggregate Density: Liquid water also Different processes influences aggregate morphology,	
264	density <del>, and rate of formation. Laboratory experiments have shown that wWet ash (&gt;10-15</del>	
265	wt.% liquid water) rapidly produces dense, sub-spherical pellets with density >1,000 kg m <sup>-3</sup>	

266 (Schumacher and Schmincke, 1991; Van Eaton et al., 2012)<u>:- whereas-drier conditions lead to</u>

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267	low-density, electrostatically-bound clusters (Schumacher and Schmincke, 1995; Van Eaton et	
268	al., 2012) with density in the hundreds of kilograms per cubic meter (James et al., 2002;	
269	Taddeucci et al., 2011) <u>(Schumacher and Schmincke, 1995; James et al., 2002; Van Eaton et</u>	
270	al., 2012). (2002) Taddeucci et al. (2011) estimated densities of dominantly several	_
271	hundred ranging from $<100$ to $>1,000$ kg m <sup>-3</sup> in dry aggregates photographed falling 7 km from	
272	the Eyjafjallajökull vent. James et al., (2003) however estimate dry aggregate densities less	
273	than 200 kg m <sup>-3</sup> . For simplicity, we hold $\rho_{agg}$ constant at 600 kg m <sup>-3</sup> , toward the middle of the	_
274	observed range but higher than that of some dry aggregates. Our results, in terms of oOptimal	
275	aggregate sizes that we derive later in this paper, are determined by this assumed density, and	
276	may be larger or smaller than actual aggregate sizes depending on the density used here.	
277	The TPSD used to model these four eruptions are listed in Table S1 and illustrated in Fig. 2.	
278	We aim to adjust the TPSD in our model to better match the mapped deposits. In doing so, we	
279	assume that some fraction $(m_{agg})$ of ash smaller than some size $\phi_p^{max}$ collects into clusters having	
280	a density $ ho_{agg}$ and Gaussian size distribution of mean $\mu_{agg}$ , and standard deviation $\sigma_{agg}$ . For	
281	deposit modeling, we ignore the small fraction of the erupted mass that goes into the distal	
282	cloud, typically a few percent (Dacre et al., 2011; Devenish et al., 2012). In the Appendix we	
283	briefly review aggregation processes. We offer the following parameterization scheme:	
284	For $\phi >=4$ , all ash aggregates	
285	For $\phi \ll 2$ , no ash aggregates.	
286	For $4 \ge \phi \ge 2$ , the mass fraction that aggregates varies linearly with $\phi$ from 1 (when $\phi = 4$ ) to 0	
287	(when $\phi=2$ ). Based on this scheme, particle sizes that aggregate are depicted as gray bars in Fig.	
288	<del>2.</del>	
•		

## 289 **3.3 Statistical measures of fit**

For each eruption, we have done a series of model simulations, first using the TPSD without considering aggregation, and then systematically varying  $\sigma_{agg}$  and  $\mu_{agg}$  to include the effects of aggregation. We compare the resulting deposit with the mapped deposit using three methods presented in Table 3. Each has advantages and disadvantages.

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294 1) The point-by-point index  $\Delta^2$  compares model results with sample data collected at specific 295 locations (dots, Fig. 1). It offers the advantage that the comparison is made directly with 296 measured values, not with interpreted or extrapolated contours of data. But  $\Delta^2$  values are 297 dominated by differences in proximal locations where mass per unit area is greatest; and values 298  $\frac{\partial f A^2}{\partial t}$  can be influenced by errors in the wind field, which cannot be adjusted in the model. More 299 importantly,  $\Delta^2$  can be dominated by differences in proximal locations where mass per unit area 800 is greatest, and where near-vent processes, such as fallout from the vertical column, are not 801 accurately simulated. For these reasons, we exclude proximal data, within a few column heights 302 distance from the vent, from the calculation of  $\Delta^2$ .

2) **The downwind thinning index**  $\Delta^2_{downwind}$ , compares modeled mass per unit area along the downwind dispersal axis with values expected at that distance based on a trend line drawn from field measurements (Fig. 3Fig. 4). The comparison is not made directly with measured values (a disadvantage). However the method does not suffer the limitation of over-weighting proximal data. And, more importantly, it still provides a useful comparison when wind errors cause the modeled dispersal axis to diverge from the mapped one.

309 3) The isomass area index  $\Delta_{area}^2$  compares the area within modeled and mapped isomass 310 lines. It is based on traditional plots of the log of isopach thickness versus square root of area 311 (Pyle, 1989; Fierstein and Nathenson, 1992; Bonadonna and Costa, 2012), which are assumed 312 to accurately depict the areal distribution of tephra while minimizing the effects of 3-D wind 313 on the distribution (Pyle, 1989). Fig. 4Fig. 5 shows plots for our four eruptions, using the log 314 of isomass rather than isopach thickness to avoid problems introduced by varying deposit 315 density.

The index  $\Delta_{area}^2$  is assumed to be insensitive to effects of wind (an advantage). However, model results are compared with isopach lines that are interpretive and may not be well constrained, depending on the distribution and number density of sample locations.

### 319 3.4 Sensitivity to various input values

We ignore complex, proximal fallout and concentrate on medial to distal areas, about 100 to  $\sim$ 500 km downwind for example at Mount St. Helens. There, under the average wind speed (15.1 m s<sup>-1</sup>) that existed below about 15 km, tephra falling from 15km at average settling velocities of 0.4-1.5 m s<sup>-1</sup> would deposit within this range (Fig. 5Fig. 6a). Tephra falling at

- $0.66-0.78\ m\ s^{-1}$  would land 290-340 km downwind, the distance of the secondary maximum at
- 325 Ritzville. A wide range of aggregate diameters *d* could fall at this rate depending on density

326  $\rho_{agg}$  (Fig. 5<u>Fig. 6</u>b).

327 Other factors listed below can also affect the results.

- 328 *Aggregate shape*. Aggregate shape can strongly affect the settling velocity and thus where 329 deposits fall, as illustrated in Fig. 6Fig. 7. For simplicity, we use round aggregates (F=1.0).
- 30 Suzuki k. Simulations of Mount St. Helens (Fig. 7 Fig. 8) show that increasing the Suzuki factor
- 331 from 4 to 8 increases the prominence of a secondary thickness maximum. But at k > 8, the
- 332 proximal deposit becomes unrealistically thin. Our simulations use k=8 to replicate the known
- 333 prominent secondary thickening while minimizing unrealistic thinning of proximal deposits.
- Aggregate size. The transport distance is highly sensitive to aggregate size. Reducing aggregate diameter *d* from 0.250 to 0.217 to 0.189 mm increases transport distance at Mount St. Helens from 300 to 366 to 448 km respectively (Fig. 5Fig. 6a). In simulations that use a single, dominant aggregate size, these variations produce conspicuous changes in the location of a secondary maximum (Fig. 8Fig. 9). Decreasing size also decreases the percent of erupted mass that lands in the mapped-area shown in Fig. 9: from 7063% to 5335% to 3915% for d=0.165, 0.143, and 0.125mm respectively ( $\phi$ =2.6, 2.8.3.0). At d=0.1mm ( $\phi$ =3.3), only 4% of
- 341 <u>the erupted mass lands in the mapped area.</u>
- 842 This constrains the range of aggregate sizes we may use in our simulations. Sparse observations 843 suggest that >90% of erupted mass falls as an observable deposit while less than several percent 844 is transported downwind as a distal cloud {Wen, 1994 #2861}{Devenish, 2012 #3029}. To 845 ensure a similar relationship in our simulations, nearly all of the aggregate-size distribution 846 must be coarser than about 0.1 mm. At the proximal end, for Mount St. Helens, Durant et al. 847 (2009) found that most fine ash fell at distances >150 km. This implies aggregate sizes coarser 848 than about 0.32mm ( $\phi$ =1.6) (Figs 6, 9). To ensure that the tails of our aggregate-size 849 distribution land in the area of interest, we must vary -Our simulations limit-µagg to-values of to 850 <u>a narrow range of about 1.89-3.14 (0.287-0.117mm12mm)</u>, and  $\sigma_{agg}$  to 0.1-0.34 to a small 851 fraction of this range, to ensure that most deposits fall in the region of interest. We assume that 852 similar constraints apply to all deposits in this study.
- *Fall-velocity model.* Different fall-velocity models are used for-in different tephra dispersion
- models. Ash3d allows users to choose from three; Pfeiffer et al. (2005), Ganser (1993), and

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855	Wilson and Huang (1979). Different fall-velocityThese models give slightly different values	 Formatted: Font color: Red
356	of fall velocityresults, and it should be noted that our results are specific to our choice of the	 Formatted: Font color: Red
357	Wilson and Huang fall model.	
358	Finally, we note that key parameters such as particle density, shape, Suzuki k etc. are held	
359	constant for all four eruptions even though they may vary from one eruption to another. Such	
360	parameters cannot easily be scrutinized when setting up simulations during an eruption. An	
361	objective is to see how well "standard" values, even if locally unrealistic, can reproduce	
362	observations.	Formatted: Font: Not Italic,

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### 363

#### 364 4 Results

865 We ran simulations at  $\mu_{agg} = 1.89, 1.92.0, 2.01, \dots 3.1\phi$ , and  $\sigma_{agg} 0.0, 0.1, 0.2$ , and  $0.3\phi$ . The latter used 1, 35, 7, and 5-11 aggregate size classes respectively, in each simulation, with the 366 percentage of fine ash assigned to each bin given in Table 4. Our calculations of  $\Delta^2$  and  $\Delta^2_{downwind}$ 367 368 only included sample points whose downwind distance lay within the range indicated by the 369 trend lines in Fig. 3Fig. 4.

Figure 9<u>Figure 10</u> shows contours of  $\Delta^2$ ,  $\Delta^2_{downwind}$ , and  $\Delta^2_{area}$  as a function of  $\sigma_{agg}$  and  $\mu_{agg}$ 870 371 for each of these four deposits. Values are given in Tables S3-S6. Although the three indices 372 compare different features of the deposit, they provide roughly similar optimal values of  $\sigma_{agg}$ 373 and  $\mu_{agg}$ . For Mount St. Helens, for example, the best-fit value of  $\mu_{agg}$  is about 2.34  $\phi$  using  $\Delta^2$  (Fig. 9Fig. 10a), 2.5 $\phi$  using  $\Delta^2_{downwind}$  (Fig. 9Fig. 10b), and 2.67 $\phi$  using  $\Delta^2_{area}$  (Fig. 9Fig. 874 875 <u>10</u>c). The fit does not depend very Optimal values of strongly on  $\sigma_{agg}$  but appears slightly 876 better at higher values are 0.1, 0.1, and 0.2 respectively. For Crater Peak, optimal  $\mu_{agg}$  values 877 are 2.360, 2.250, and 1.92.00 respectively. F, while for Ruapehu they are about 2.13-2.40 878 (poorly constrained),  $2.25\phi$ , and  $2.35\phi$ . For both Crater Peak and Ruapehu, the fit is also 879 insensitive to optimal values of  $\sigma_{agg}$ , though slightly better at higher values for Ruapehu using  $\Delta^2_{area}$  (Fig. 9i) range from 0.0 to 0.2. For Redoubt, optimal values are disparate:  $\mu_{agg} = 2.1$ -880 381  $\frac{2.25}{6}$ , 2.35, and  $\frac{42}{6}$  respectively. The Redoubt deposit is least constrained by field data and 382 the most difficult to match due to the complex wind conditions.

Figures  $\frac{1011-13-14}{1}$  show results for each of these eruptions using  $\mu_{agg} = 2.4\phi (0.29 \text{mm} 19 \text{mm})$ and  $\sigma_{agg} = 0.31\phi$ . The sizes of particles and aggregates used to generate these figures is given in Table S2. For all deposits these values are close to optimal, depending on which criterion is used. Similar figures for other values of  $\mu_{agg}$  and  $\sigma_{agg}$  are provided as Figs. S005-S172S212.

Figures S001-S004 show simulations using the original particle-size distribution, with no aggregation. Tephra fall beyond a few tens of kilometers is strongly underestimated in all these runs, especially for the three eruptions that contain more than a few percent fine ash. Values of  $\Delta^2$ ,  $\Delta^2_{downwind}$ , and  $\Delta^2_{area}$  are also higher than most simulations that use aggregates (Table S3-S6). For Mount St. Helens, Crater Peak, Ruapehu, and Redoubt, the percentages of the erupted mass landing in the mapped area are very low: 29%, 42%, 88%, and 59% respectively.

Optimal aggregates obtained from our study are similar in size but denser than those found optimal by Cornell et al. (1983) for the Campanian Y-5 ( $\mu_{agg}$ =2.3 $\phi$ ,  $\rho_{agg}$ =200 kg m<sup>-3</sup>). The unknown wind field during the prehistoric Campanian Y-5 eruption makes it difficult to compare Cornell et al.'s optimal value to the results here. Folch et al. (2010) matched the Mount St. Helens deposit using a similar aggregation scheme, but with aggregates of density 400 kg m<sup>-3</sup> (compared with our 600 kg m<sup>-3</sup>) and diameter of 0.2-0.3mm (compared with our ~0.2mm). Their results are broadly consistent with ours.

### 400 4.1 Mount St. Helens

401 For the Mount St. Helens case, the modeled deposit follows a dispersal axis (solid black line, 402 Fig. 10Fig. 11a) that matches almost exactly with the mapped one (dashed line). The agreement 403 reflects both the faithfulness of the numerical wind field to the true one and the appropriateness 404 of other inputs, such k, that influence dispersal direction. The measured mass loads in Fig. 405 10Fig. 11a, indicated by the color of markers, agree reasonably well with modeled mass loads 406 indicated by colors of the contour lines, except along the most distal transect, where modeled 407 loads are essentially zero while measured loads are about 10<sup>-1</sup> kg m<sup>-2</sup>. Figure 10Figure 11b 408 shows that modeled and measured mass loads generally agree within a factor of three or so, 409 except for those same distal, low-mass-load measurements, to the lower left of the legend label 410 (those where modeled values are truly zero do not show up on this plot). Figure 10Figure 11c 411 shows that the modeled mass load (black line with dots) contains a secondary thickening at

about the same location mapped (dashed line). It also has roughly the same downwind shape,

in contrast to results using  $\sigma_{agg}=0.2$  and 0.3 (Figs. S027-S028), in which the secondary

thickening is broader and thinner than observed. However, the modeled mass load is
consistently less than measured, especially at the most distal sites. In Fig. 10Fig. 11d, the log
of modeled mass load versus square root of area shows reasonable agreement with mapped

417 values until mass loads are less than about 1 kg m<sup>-2</sup>, where they diverge.

418 Notably, modeled mass loads somewhat underestimate the measured values along the dispersal 419 axis in Fig. 10Fig. 11c. The underestimate reflects the fact that the input erupted volume of 0.2420 km<sup>3</sup> DRE (Table 1) was based on estimates by Sarna-Wojcicki et al. (1981) of what lay within 421 the mapped area in Fig. 10Fig. 11a; yet only about 7978% of the modeled mass landed within 422 this area. Reducing the mean aggregate size to  $2.76\phi$  (0.153mm164mm, Figs. S032S036-S034) 423 improves the fit somewhat along distal transect near the dispersal axis but not along the entire 424 transect lengthbut degrades it near Ritzville. And the finer size moves the secondary maximum 425 too far east and reduces the percentage deposited to  $\frac{50-60-65}{60}$ %.

426 In Fig. 10Fig. 11 a, the modeled deposit is also slightly narrower than the mapped one. Adding 427 turbulent diffusion, with a diffusivity D of about  $3 \times 10^2 \text{ m}^2 \text{ s}^{-1}$  (Fig. 14Fig. 15) visually improves 428 the fit, and was likely important during this eruption due to high crosswind speeds that increased 429 entrainment (Degruyter and Bonadonna, 2012; Mastin, 2014). But adding diffusion slightly 430 increases  $\Delta^2$ , improving fit on deposit margins at the expense of the axis. Ignoring turbulent 431 diffusion also decreases run time by  $\sim 3x$ , from  $\sim 30$  to 10 minutes, yielding faster results for 432 operational runs, and is a reasonable compromise under operational conditions. Results with 433 other models may vary depending on model setup and configuration.

### 434 4.2 Crater Peak (Mount Spurr)

At Crater Peak (Mount Spurr), results in Fig. 11Fig. 12 a also show good agreement between the modeled dispersal axis and the mapped one (which is constrained by fewer sample locations than the Mount St. Helens case). The isomass lines in this plot are jagged and irregular due to effects of topography in this mountainous region. The modeled location of secondary thickening in Fig. 11Fig. 12 c agrees with the mapped location, about 250-300km downwind. Although Fig. 11Fig. 12 c shows a tendency to underestimate the mass load along the dispersal axis, there is less tendency to underestimate mass load in the most distal locations as occurred

442 at Mount St. Helens. In <u>Fig. 11Fig. 12</u>d, the areas covered by modeled isomass lines are 443 comparable to the mapped values, down to mass loads approaching 0.1 kg m<sup>-2</sup>.

### 444 4.3 Ruapehu

445 For Ruapehu (Fig. 12Fig. 13), simulations using the NCEP Reanalysis 1 numerical winds 446 produced an odd double dispersal axis whose average did not correspond well with the mapped 447 direction of dispersal (Fig. 1c). To improve the fit we used the 1-D wind sounding provided 448 for this eruption at the IAVCEI Tephra Hazard Modeling Commission web page 449 (http://dbstr.ct.ingv.it/iavcei/). Use of a 1-D wind sounding seems justified in this case because 450 this deposit covers a smaller area than the others, making a 3-D wind field less important in 451 calculating transport. The resulting dispersal axis (Fig. 12Fig. 13a) agrees with the mapped one 452 out to about 140 km distance, beyond which it strays eastward, reaching the coast, 180 km 453 downwind, about 10 km east of the mapped axis. This slight difference is enough to cause 454 misfits in point-to-point comparisons at measured mass loads of ~10<sup>-1</sup> kg m<sup>-2</sup> (Fig. 12Fig. 13b).

The modeled mass load along the dispersal axis (Fig. 12Fig. 13c) agrees with measurements to about 60-90 km distance. At 100-200 km, modeled values level off and show a hint of secondary thickening at ~180 km, in agreement with the mapped deposit (Fig. 1c and He13c), although the mapped secondary thickening is more prominent.

459 A large discrepancy is also apparent at distances of less than 60 km, where mass load along the 460 dispersal axis (Fig. 12Fig. 13c) and the area covered by thick isomass lines (Fig. 12Fig. 13d) is 461 greater than the mapped deposit. The implication is that too much mass is dropping out 462 proximally in the model. Underestimates of isomass area at  $<=10^{-1}$  kg m<sup>-2</sup> (Fig. 12Fig. 13d) also show that too little is falling distally. Simulations (not shown) that raise the plume height 463 464 or increase k to concentrate more mass high in the plume do not improve the fit. The discrepancy may reflect the coarse TPSD-50% of which is coarser than 1mm (compared with 465 466 2%, 12%, and 8% for the other three deposits in Table S1). An additional simulation used the 467 TPSD derived from technique B of Bonadonna and Houghton (2005) (Table S1), which divides 468 the deposit into arbitrary sectors, and calculates a weighted sum of the size distributions in each 469 sector following Carey and Sigurdsson (1982). Technique B yields a finer average particle size 470 than technique C, which uses Voronoi tessellation to sectorize the deposit. But the finer particle 471 size of the technique B TPSD does not improve the fit-(Fig. S173). Further exploration of this 472 discrepancy is beyond the scope of this paper; but other possible causes could include release

of different particle sizes at different elevations, or complex transport in the bending of theweak plume that can't be accommodated in this model.

475 A second, smaller discrepancy is that the modeled deposit is narrower than the mapped one 476 (Fig. 1c). As at Mount St. Helens, deposit widening due to cross flow entrainment is likely. 477 Increases in entrainment resulting from crossflow is widely known to both increase plume width 478 and decrease its height for a given eruption rate (Briggs, 1984; Hoult and Weil, 1972; Hewett 479 et al., 1971; Woodhouse et al., 2013). Adding turbulent diffusion, we get a visually improved 480 fit when  $D = -3 \times \frac{10^3}{10^2} \text{ m}^2 \text{ s}^{-1}$  (Fig. 15Fig. 16), consistent with findings by Bonadonna et al. 481 (2005) based on the rate of downwind widening of isomass lines. This diffusivity is also similar 482 to the visual best-fit value for Mount St. Helens (Fig. 14Fig. 15).

483 Despite the uncertainty in TPSD, simulations that systematically vary  $\mu_{agg}$  and  $\sigma_{agg}$  fit best in 484 Figs. 9g10g, h, and i when  $\mu_{agg}$  is about 2.2-3 to 2.45. Results similar to those presented in 485 Fig. 12Fig. 13c use other values of  $\mu_{agg}$  (Figs. S089S109-S130S160) and show a secondary 486 maximum migrating downwind as  $\mu_{agg}$  increases, coming into agreement with the mapped 487 distance at  $\mu_{agg} = 2.2-3$  to  $2.45\phi$  (0.1920-0.22mm18mm), where errors in Fig. 9Fig. 10g, h, and 488 i are lowest.

### 489 4.4 Redoubt

490 This deposit is the second smallest in our group, the least well-constrained by sampling, and 491 the only one in our group not known to include a secondary thickness maximum. Mastin et al. 492 (2013b) modeled this deposit using numerical winds from the North American Regional 493 Reanalysis model (Mesinger et al., 2006). During that eruption, the winds at 0-4 km, 6-10, and 494 >10 km elevation were directed toward the northwest; north, and northeast respectively, with 495 the highest speeds at 6-10 km. Mastin et al. found that the modeled cloud developed a north-496 oriented, northward migrating wishbone shape with the west prong at low elevation and the east 497 prong at high elevation. Mastin et al. also found that the modeled dispersal axis and the mass 498 load distribution roughly agreed with mapped values for a plume height of 15km, k=8, and a 499 particle size adjustment that involved taking 95% of the fine ash (<0.063mm) and distributing it evenly among the coarser bins. In this study we use the same plume height and k value, a 500 501 different wind field (RE1), and explore a different parameterization for particle aggregation.

502 In Fig. 13Fig. 14a, the modeled dispersal axis diverges about 20° westward from the mapped 503 axis. We do not correct this divergence by adjusting mass height distribution, since the optimal values of  $\mu_{agg}$  and  $\sigma_{agg}$  can still be obtained from  $\Delta^2_{downwind}$ , and  $\Delta^2_{area}$ . As with the Crater 504 505 peak (Spurr) simulations, the isomass lines are jagged and patchy; an artifact of high relief. 506 (The most distal sample location lies at 4.3 km elevation on the west shoulder of Mount 507 McKinleyDenali). Although the value of  $\mu_{agg}$  (2.4 $\phi$ , 0.20mm19mm) portrayed in Fig. 13Fig. 508 14 is close to optimal in Fig. 9Fig. 10, many sample points do not plot in Fig. 13Fig. 14b 509 because modeled mass load is zero. And most values of  $\Delta^2$  are high—0.99, largely because of 510 the disparity in axis dispersal directions and the consequent fact that sample points lie outside 511 the modeled deposit. The reason that  $\Delta^2$  shows a clear minimum, around  $\mu_{agg}=2.4\phi$ 512 (0.20mm19mm) in Fig. 9Fig. 10j, is apparent from Figs. S131S161-S172-S212 which show 513 that, as  $\mu_{agg}$  decreases in size, the modeled deposit extends farther north and takes a clear turn 514 to the northeast, overlapping more with the mapped deposit. These figures also illuminate why 515  $\Delta^2_{downwind}$  is optimal at  $\mu_{agg}$  =2.3; because modeled and mapped loads come into best agreement 516 along the dispersal axis for aggregates of this size.  $\Delta_{area}^2$  is optimized at  $\mu_{agg} < \frac{1-2}{2}$  because the 517 area of the 1 kg m<sup>-2</sup> isomass diverges below the mapped value, and the area of the 0.01 kg m<sup>-2</sup> 518 isomass diverges above observed, as aggregate size increases. The isomass lines are drawn 519 based on sparse data and are the least reliable of the datasets used in this comparison.

### 520 5 Discussion and Conclusions

521 The overall derived values of  $\mu_{agg}$  have a narrow range between  $\sim 2.43 - 2.57 \phi$  (0.4815-522 0.23mm20mm), despite large variations in erupted mass (0.25-50×Tg), plume height (8.5-25 523 km), mass fraction of fine (<0.063mm) ash (3-59%), atmospheric temperature, and water 524 content between these eruptions. The value of this narrow range depends strongly on other 525 inputs, such as particle density, shape factor, and Suzuki factor. Values assigned here may not 526 always be representative. Aggregate density for example is frequently less than 600 kg m<sup>-3</sup>. 527 And different assumptions on particle or aggregate shape could significantly change our results. 528 Moreover, our result is partly an artifact of our choice to optimize fit to deposits at medial distances of several tens to hundreds of kilometers. Including more proximal sample points 529 530 may have given optimal aggregate sizes that spanned a wider range, as used for example in 531 aggregation schemes for Vesuvius {Barsotti, 2015 #6790} or Iceland {Biass, 2014 #6789}.

532 But, holding those factors constant<u>Despite these considerations</u>, the similarity in this 533 rangeoptimal values of  $\mu_{agg}$  between these four eruptions is noteworthy.

The overall agreement in modeled mean aggregate size ( $\mu_{agg}$ ) suggests that accelerated fineash deposition may be treated as a discrete process, insensitive to eruptive style or magnitude. It seems unlikely that these varied eruptions would produce aggregates of the same size, density, and morphology. A combination of processes removed ash. Our approach captures these processes implicitly, ignoring the microphysics.

539 What sort of processes could evolve in the cloud? Some possibilities are illustrated in Fig. 540 A1Fig. 2. The evolution starts with ejection of particles from that the vent-whose, with size 541 ranges ranging from microns to meters. For an eruption having the TPSD of Mount St. Helens, 542 the rising plume would have contained  $10^{6}$ - $10^{8}$  particles per cubic meter with diameter between 543 10-30 µm that collided with larger particles hundreds of thousands of many times per second. 544 High collision rates and the availability of liquid water in the plume would have led to rapid 545 aggregation. Freezing of liquid water and riming would have shifted the maximum possible 546 size of aggregates towards mm to cm sizes. Mud rain, observed falling at Mount St. Helens 547 (Waitt, 1981) and ice aggregates collected near the vent at Redoubt (Van Eaton et al., in press), 548 are evidence of these processes.

549 In the downwind cloud particle concentrations were lower, turbulence was less intense, a 550 smaller range of particle sizes existed, and, for all four eruptions, atmospheric temperatures 551 near the plume top were well below freezing (Table 5), leading to presumably slow aggregation 552 rates. However, at least two other processes may help settle ash from downwind clouds. One 553 is gravitational overturn. Experiments (Carazzo and Jellinek, 2012) have observed that fine ash 554 settles toward the bottom of ash clouds as they expand and move downwind, accumulating 555 gravitationally unstable particle boundary layers that eventually overturn and cause the entire 556 air mass to settle rapidly. At Eyjafjallajökull in 2010, gravitational convective instabilities 557 formed within 10km of the vent, presumably as a result of accumulation of coarse ash over a 558 period of minutes (Manzella et al., 2015). The development of fine-ash particle boundary layers 559 presumably takes longer, perhaps hours, although the underlying processes remain a subject of 560 active research.

561 A second process is hydrometeor growth. In some cases, magmatic and (or) externally-derived 562 water in the eruption cloud may condense on ash particles and initiate hydrometeor growth.

563 Both hydrometeor growth and gravitational overturn have been suggested to produce the 564 mammatus clouds that developed in mid-day over central Washington on 18 May 1980 and 565 signaled mass settling (Durant, 2015; Durant et al., 2009; Carazzo and Jellinek, 2012). 566 Mammatus descent rates are typically meters per second (Schultz et al., 2006), much faster than 567 the settling rate of individual ash particles (<0.1 m s<sup>-1</sup>) or even of ash aggregates (<~1 m s<sup>-1</sup>, 568 Fig. 5Fig. 6).

The extent to which these processes operated at Crater Peak, Ruapehu, and Redoubt is unknown. Cloud structures were not observed during the nighttime eruptions of Redoubt and Crater Peak (Spurr). And although virga-like structures can be seen in some near-vent photos of Ruapehu (Bonadonna et al., 2005, Fig. 9a), we have seen no documentation of such instabilities farther downwind.

For operational forecasting, these mechanisms cannot be considered in any case, because no operational model has the capability to resolve these processes. The fact that these eruptions can all be reasonably modeled using similar inputs for aggregate size is convenient, even if the model does not calculate the processes involved. The agreement suggests that model forecasts can still be useful during the coming years. Future work will focus on the development of more sophisticated algorithms that account for cloud microphysics.

### 580 <mark>6 Appendix</mark>

581	The rate and extent of ash aggregation are sensitive to changes in both eruptive
582	conditions and background meteorology. Despite the complexity of the process, field studies
583	and laboratory experiments have highlighted key spatial and temporal controls. For example,
584	large aggregates, including frozen accretionary lapilli, tend to form near the volcanic source
585	and are particularly abundant in phreatomagmatic eruption deposits (Van Eaton et al., 2015;
586	Brown et al., 2012; Houghton et al., 2015). These are associated with precipitation forming
587	processes occurring as particles collide in moist, turbulent updrafts rising above the volcanic
588	vent or ground hugging density currents (Fig. A1). Field measurements indicate that near-
589	source aggregates commonly exceed 1 cm diameter (Wallace et al., 2013; Swanson et al., 2014;
590	Van Eaton and Wilson, 2013). In contrast, the low density aggregates that produced the
591	Ritzville Bulge, 230 km downwind from Mount St. Helens, are thought to have been triggered
592	by mammatus cloud instabilities (Durant et al., (2009). As the cloud descended to warmer
593	atmospheric levels, the increasing proportion of liquid water increased the rate of aggregation
594	and fallout (red line, Fig. A1). These types of distal aggregates tend to be smaller than a

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595	millimeter, forming in the downwind cloud up to hundreds of kilometers from source (Sorem,
596	<del>1982; Dartayat, 1932).</del>
597	Liquid water also influences aggregate morphology, density, and rate of formation. Laboratory
598	experiments have shown that wet ash (>10-15-wt.% liquid water) rapidly produces dense, sub-
599	spherical pellets, whereas drier conditions lead to low density, electrostatically bound elusters
600	(Schumacher and Schmincke, 1995; James et al., 2002; Van Eaton et al., 2012). Furthermore,
601	aggregation is a highly size selective process smaller particles (<0.25mm) have a much
602	greater likelihood of sticking (Gilbert and Lane, 1994; Schumacher and Schmincke, 1995; Van
603	Eaton et al., 2012). In this study, we do not attempt to address the detailed mechanisms of
604	aggregation, but consider the bulk impact on downwind deposits for practical applications in
605	ash dispersal forecasting.
606	Author contributions

607 L. Mastin conceived the study, did the model simulations and wrote most of the paper. A. Van

Eaton provided advice on aggregation processes-<del>and wrote the appendix</del>. A. Durant provided

the data for Mount St. Helens and Crater Peak, and advice on aggregation processes that

610 occurred during those two eruptions.

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## 862 Tables

# 863 Table 1: Input parameters for simulations. Vent elevation is given in kilometers above mean

sea level.

PARAMETER(S)	MOUNT ST. HELENS	SPURR	RUAPEHU	REDOUBT
MODEL DOMAIN	42-49°N 124-110°W 0-35 km asl	59-64°N 155.6- 141.4°W 0-17 km asl	39.5-37.5°S 175-177°E 0-12km asl	60-64°N 155-145°W 0-20km asl
VENT LOCATION	122.18°W 46.2°N	152.25°W 61.23°N	175.56°E 39.28°S	152.75°W 60.48°N
VENT ELEVATION (KM)	2.00	2.30	2.80	2.30
NODAL SPACING	0.1° horizontal 1.0 km vertical	0.1° horizontal 1.0 km vertical	0.025° horizontal 0.5 km vertical	0.07° horizontal 1.0 km vertical
ERUPTION START DATE (UTC)	1980.05.18	1992.09.17	1996.06.16	2009.03.23
(YYYY.MM.DD)			1996.06.17	
START TIME (UTC)	1530 <u>UTC</u>	0803 UTC	2030 UTC 0200 UTC	1230 UTC
PLUME HEIGHT, KM ASL	See Table 2	13	8.5	15
DURATION, HRS	See Table 2	3.6 4.5 2.0		0.33
ERUPTED VOLUME	0.2 (total)	0.014	0.000643	0.0017
KM <sup>3</sup> DRE			0.000357	
DIFFUSION COEFFICIENT D	0	0	0	0
SUZUKI CONSTANT K	8	8	8	8
PARTICLE SHAPE FACTOR F	0.44	0.44	0.44	0.44
AGGREGATE SHAPE FACTOR F	1.0	1.0	1.0	1.0

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867	Table 2: Time series of plume height and total erupted volume used in model simulations of
868	the Mount St. Helens ash cloud. H=plume height in km above sea level (a.s.l.), V=erupted
869	volume in million cubic meters dense-rock equivalent (DRE). The time series of plume height
870	approximates that measured by radar (Harris et al., 1981). We calculated a preliminary eruptive
871	volume for each eruptive pulse using the duration and the empirical relationship between plume
872	height and eruption rate (Mastin et al., 2009). This method underestimated the eruptive volume,
873	as noted in previous studies (Carey et al., 1990). Hence we adjusted the volume of each pulse
874	proportionately so that their total equals the 0.2 km <sup>3</sup> DRE estimated by Sarna-Wojcicki et al.
875	(1981). For the last two eruptive pulses, start times in UTC, marked with asterisks, are on 19
876	May in UTC time. All other start times are on 18 May.

Plume height (H), duration (D) and volume (V)

st	start		Н	V
PDT	UTC	min	km asl	$ imes 10^6  m^3  DRE$
8:30	1530	30	25	3.247
9:00	1600	36	15.3	0.077
9:36	1636	54	13.7	0.356
10:30	1730	45	15.3	0.502
11:15	1815	30	16.1	0.426
11:45	1845	42	17.4	0.615
12:27	1927	48	17.4	0.615
13:15	2015	60	14.6	0.183
14:15	2115	45	14.7	0.535
15:30	2230	60	15.8	0.691
16:30	2330	60	19.2	0.700
17:30	0030*	60	7.7	1.945
18:30	0130*	60	6.2	0.020

878 Table 3. Statistical measures of fit used in the	his paper
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Name	Formula	Explanation
Point-by- point method	$\Delta^{2} = \left[\frac{\sum_{i=1}^{N} (m_{m,i} - m_{o,i})^{2}}{\sum_{i=1}^{N} m_{o,i}^{2}}\right]$	The mass load $m_{o,i}$ observed at each sample location <i>i</i> is compared with modeled mass load $m_{m,i}$ at the same location. Squared differences are summed to the total number of sample points <i>N</i> , and normalized to the sum of squares of the observed mass loads.
Downwind thinning method	$\Delta_{domination}^{2} = \frac{1}{M} \sum_{j=1}^{M} \left( \log \left( m_{m,j} / m_{o,j} \right) \right)^{2}$	The log of modeled mass load $m_{m,j}$ at a point <i>j</i> on the dispersal axis, is compared with the observation-based value $m_{o,j}$ expected at that location based on a trend line drawn between field measurements along the axis (Fig. <b>7</b> Fig. 4). Differences between $m_{m,j}$ and $m_{o,j}$ are calculated on a log scale, squared, and summed.
lsomass area method	$\Delta_{area}^{2} = \left[\frac{\sum_{i=1}^{L} (A_{m,i} - A_{o,i})^{2}}{\sum_{i=1}^{L} A_{o,i}^{2}}\right]$	This method calculates the area $A_{m,i}$ of the modeled deposit that exceeds a given mass load <i>i</i> by summing the area of all model nodes that meet this criterion. It then takes the difference between $A_{m,i}$ and the area $A_{o,i}$ within same isomass line mapped from field observations. The sum of the squares of these differences, normalized to the sum of the squared mapped isopach areas, gives the index $\Delta_{area}^2$ .

Table 4: percentage of fine ash assigned to different size bins for different values of  $\sigma_{agg}$ . The mass fraction  $m_{\phi}$  in each bin ( $\phi$ ) was calculated using the equation for a Poisson distribution,  $m_{\phi} = (1/\sqrt{2\pi}) \exp\left\{\left[-(\phi - \mu_{agg})\right]^2/(2\sigma_{agg})^2\right\}$ . Values of  $m_{\phi}$  were then adjusted proportionally so that their sum added to 1.  $\sigma_{agg}$  -0.4 $\phi$  -0.3 $\phi$  -0.2 $\phi$  -0.1 $\phi$   $\mu_{agg}$  +0.1 $\phi$  +0.2 $\phi$  +0.3 $\phi$  +0.4 $\phi$ 

$\sigma_{agg}$	- <del>0.4</del> ¢	-0	<del>.3</del> 0	<del>-0.2</del> ¢	<del>-0.1ø</del>	$\mu_{agg}$	<del>+0.1ø</del>	<del>+0.2<i>\$</i></del>	<del>+0.3<i>0</i></del>	<del>+0.4<i>¢</i></del>
<del>0.0</del>						<del>100%</del>				
<del>0.1</del>				<del>6%</del>	<del>24%</del>	<del>40%</del>	<del>24%</del>	<del>6%</del>		
<del>0.15</del>		<del>3.</del>	<del>5%</del>	<del>11%</del>	<del>22%</del>	<del>27%</del>	<del>22%</del>	<del>11%</del>	<del>3.5%</del>	
<del>0.2</del>	<del>2.8%</del>	<del>6.</del>	<del>7%</del>	<del>12%</del>	<del>18%</del>	<del>20%</del>	<del>18%</del>	<del>12%</del>	<del>6.7%</del>	<del>2.8%</del>
Bin	<u>σ<sub>agg</sub>=0</u>	<u>0.1</u>	<u>0.2</u>	<u>0.3</u>						
<u>µ<sub>аgg</sub> -0.6ф</u>				<u>1.9</u>						
<u>µ<sub>аgg</sub> -0.5ф</u>			<u>0.9</u>	<u>3.4</u>						
μ <sub>agg</sub> -0.4φ			<u>2.7</u>	<u>5.6</u>						
μ <sub>agg</sub> -0.3φ			<u>6.5</u>	<u>8.3</u>						
<u>μ<sub>agg</sub> -0.2φ</u>		<u>6</u>	<u>12</u>	<u>11.0</u>						
<u>μ<sub>agg</sub> -0.1φ</u>		<u>24</u>	<u>18</u>	<u>13.0</u>						
<u>µ<sub>agg</sub></u>	<u>100</u>	<u>40</u>	<u>20</u>	<u>13.7</u>						
μ <sub>agg</sub> +0.1¢	2	24	<u>18</u>	<u>13.0</u>						
<u>μ<sub>agg</sub> +0.2</u>		<u>6</u>	<u>12</u>	<u>11.0</u>						
μ <sub>agg</sub> +0.3¢	2		<u>6.5</u>	<u>8.3</u>						
<u>μ<sub>agg</sub> +0.4</u>	2		<u>2.7</u>	<u>5.6</u>						
μ <sub>agg</sub> +0.5¢	2		<u>0.9</u>	<u>3.4</u>						
μ <sub>agg</sub> +0.6¢				<u>1.9</u>						

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Table 5: Atmospheric temperature profiles during the eruptions at Mount St. Helens, Crater Peak (Spurr), Ruapehu, and Redoubt volcanoes. Profile for Mount St. Helens is for 18 May

890 1980, 1800 UTC, interpolated to the location of Ritzville, Washington (47.12°N, 118.38°W).

For Crater Peak (Spurr) the profile is for 17 September 1992, 1200 UTC, interpolated to the

892 location of Palmer, Alaska (61.6°N, 149.11°W). For Ruapehu the temperature profile is for

893 17 June 1996, 0000 UTC, interpolated to the location of Ruapehu. For Redoubt the sounding

was for 23 March 2009, 1200 UTC, at 62°N, 153°W. All soundings were taken from using
 RE1 reanalysis data at <a href="http://ready.arl.noaa.gov/READYamet.php">http://ready.arl.noaa.gov/READYamet.php</a>. For Mount St. Helens,

the freezing elevation was also checked using data from the North American Regional

897 Reanalysis (NARR) model (Mesinger et al., 2006), available at the same NOAA site, and

898 899 found to be 3.3 km, similar to that given below by the RE1 model.

	Mount St. Helens		Crater Peak (Spurr)		Ruape	ehu	Redoubt		
p (hPa)	z (m)	T (C)	z (m)	T (C)	z (m)	T (C)	z (m)	T (C)	
10	31,381	-39.9	31,137	-41.8	30,632	-54.9	30,179	-61.9	
20	26,713	-47.5	26,535	-51.0	26,239	-57.9	25,891	-62.1	
30	24,067	-52.1	23,920	-54.4	23,673	-56.6	23,385	-61.3	
50	20,786	-55.7	20,660	-55.5	20,441	-57.1	20,185	-57.6	
70	18,646	-55.8	18,515	-55.6	18,307	-56.4	18,049	-55.1	
100	16,377	-55.4	16,241	-55.3	16,041	-56	15,759	-53.1	
150	13,782	-55.1	13,646	-56.0	13,439	-54.2	13,133	-51	
200	11,962	-58.3	11,833	-58.9	11,613	-58.6	11,255	-50.4	
250	10,552	-53.4	10,412	-51.3	10,214	-58.3	9,814	-54.7	
300	9,355	-44	9,200	-41.0	9,057	-53.4	8,652	-55.5	
400	7,355	-28.5	7,174	-25.0	7,151	-38.9	6,764	-41.9	
500	5,716	-16.4	5,519	-15.5	5,576	-26.7	5,225	-33.9	
600	4,318	-6.9	4,126	-10.2	4,231	-15.5	3,929	-27.4	
700	3,100	0.1	2,929	-6.7	3,049	-8.6	2,802	-19.5	
850	1,515	10.3	1,397	-2.0	1,524	-1.4	1,330	-9.7	
925			722	-0.2	844	3.8	675	-8.9	

## 902 Figure captions

903 Figure 1: Maps of the deposits investigated in this work: (a) Mount St. Helens, 18 May 1980; 904 (b) Crater Peak, 16-17 September, 1992; (c) Ruapehu, 17 June, 1996; and (d) Redoubt, 23 905 March, 2009. Isomass lines for Mount St. Helens were digitized from Fig. 3Fig. 438 in Sarna-906 Wojcicki et al. (1981); for Crater Peak from Fig. 15 Fig. 16 in McGimsey et al. (2001); for 907 Ruapehu from Fig. 1 of Bonadonna and Houghton (2005); and for Redoubt from Wallace et 908 al. (2013). Isomass values are all in kg m<sup>-2</sup>. Colored markers represent locations where isomass 909 was sampled, with colors corresponding to the mass load shown in the color table. Black dashed 910 lines indicate the dispersal axis. Sample locations for Mount St. Helens taken from supplementary material in Durant et al. (2009); for Redoubt from Wallace et al. (2013), for 911 912 Crater Peak from McGimsey et al. (2001) and for Ruapehu, from data posted online at the 913 IAVCEI Commission on Tephra Hazard Modeling database (http://dbstr.ct.ingv.it/iavcei/ 914 (Bonadonna and Houghton, 2005; Bonadonna et al., 2005)).

915 Figure 2: Illustration of the path taken by coarse aggregates that fallout in proximal sections, 916 less than a few plume heights from the source (left), and fine aggregates that fall out in distal 917 sections (right). Among distal fine aggregates, we show the path taken by those that might have 918 formed within or below the downwind cloud as hypothesized by Durant et al. (2009) (red 919 dashed line), and those that were transported downwind without changing size, as calculated 920 by Ash3d (blue dashed line). Also illustrated are some key processes that might influence the 921 distribution of fine, distal ash, including development of gravitational instability and overturn 922 within the downwind cloud (Carazzo and Jellinek, 2012), and the development of 923 hydrometeors as descending ash approaches the freezing elevation (Durant et al., 2009).

Figure 2Figure 3: Total particle size distribution for each of the deposits studied: (a) Mount St.
Helens, (b) Crater Peak (Mount Spurr), (c) Ruapehu, and (d) Redoubt. Gray bars show the
original TPSD before aggregation. Black bars show the sizes not involved in aggregation; red
bars show sizes of aggregate classes used in Figs. 1011-1314.

Figure 3Figure 4: Mass load versus downwind distance along the dispersal axis for the deposits
of (a) Mount St. Helens, (b) Crater Peak (Mount Spurr), (c) Ruapehu, and (d) Redoubt. Squares
indicate sample points within 20 km of the dispersal axis, with the grayscale value indicating
the distance from the dispersal axis following the color bar in (a). The dash trend lines represent

932 interpolated values of the mass load that are compared with modeled values to calculate 933  $\Delta_{downwind}^2$ .

Figure 4Figure 5: Log mass load versus the square root of the area within isomass lines mapped
for the (a) Mount St. Helens; (b) Crater Peak (Spurr); (c) Ruapehu; and (d) Redoubt deposits.
Also shown are best-fit lines, drawn by visual inspection, using either one line segment
(Ruapehu, Redoubt) or two, where justified (Spurr, St. Helens). Triangular markers are marked
with labels indicating the approximate percentage of the deposit mass lying inboard of these
points, as calculated using equations derived from Fierstein and Nathenson (1992).

940 Figure 5 Figure 6: (a) Transport distance versus average fall velocity, assuming a 15.1 m s<sup>-1</sup> 941 wind speed, equal to the average wind speed at Mount St. Helens between 0 and 15 km, and a 942 fall distance of 15 km. The vertical shaded bar represents the distance of Ritzville. Labels on 943 dots give the average diameter of a round aggregate having a density of 600 kg m<sup>-3</sup> and the given fall velocity. (b) Average fall velocity between 0 and 15 km elevation, versus aggregate 944 945 diameter, for round aggregates having densities ranging from 200 to 2,500 kg m<sup>-3</sup>. The 946 horizontal shaded bar represents the range of average fall velocities that would land in Ritzville. 947 Fall velocities are calculated using relations of Wilson and Huang (1979), at 1-km elevation 948 intervals in the atmosphere, from 0 to 15 km, then averaged to derive the values plotted.-

Figure 6Figure 7: Deposit maps for simulations using a single size class representing an aggregate with phi size 1.9 and density 600 kg m<sup>-3</sup>, using three shape factors: (a) F=0.44; (b) F=0.7; and (c) F=1.0. Inset figures illustrate ellipsoids having the given shape factor, assuming b=(a+c)/2.

953 Figure 7Figure 8: Deposit map for simulations using a single size class representing an 954 aggregate with F=1.0, phi size 2.4 $\phi$  and density 600 kg m<sup>-3</sup>. Figs. 7<u>a8a</u>, b, and c, illustrate the 955 deposit distribution using Suzuki k values of 4, 8, and 12, while Fig. 7Fig. 8 d illustrates the 956 deposit distribution resulting from release of all the erupted mass from a single node at the top 957 of the plume. Inset plots schematically illustrate the vertical distribution of mass with height 958 in the plume for each of these cases. Simulations used other input values as given in Table 1. 959 Colored dots represent sample locations with colors indicating the sampled mass load, as in 960 Fig. 1a.

Figure 8<u>Figure 9</u>: Results of Mount St. Helens simulations using a single size class of round aggregates in each simulation:  $\phi$ =1.8, 2.0, 2.2, 2.4, and 2.6 in (a), (b), (c), (d), and (e); (f) shows

the mapped mass load, digitized from Fig. 3Fig. 438 in Sarna-Wojcicki et al. [1981]. Markers
in each figure provide the sample locations, with colors indicating the mass load measured at
each location, as shown in the color bar. Lines are contours of mass load with colors giving
their values. The mass load values of the contour lines, from lowest to highest, are 0.01, 0.1,
0.5, 1, 5, 10, 20, 30, 50, 80, and 100 kg m<sup>-2</sup> respectively.

Figure 9<u>Figure 10</u>: Contours of  $\Delta^2$  (left column),  $\Delta^2_{downwind}$  (middle column), and  $\Delta^2_{area}$  (right 968 969 column) as a function of  $\sigma_{agg}$  and  $\mu_{agg}$  for deposits from Mount St. Helens (top row); Crater 970 Peak (Mount Spurr, second row); Ruapehu (third row), and Redoubt (bottom row). The values 971 of these contour lines are indicated by the color using the color bar at the right. Maximum and 972 minimum values in the color scale are given within each frame. The best agreement between 973 model and mapped data is indicated by the deep blue and purple contours; the worst is indicated 974 by the yellow contours. Regions of each plot where agreement is best is indicated by the word 975 "Lo".

976 Figure 10Figure 11: Results of the Mount St. Helens simulation that provides approximately 977 the best fit to mapped data ( $\mu_{agg} = 2.4\phi$  and  $\sigma_{agg} = 0.31\phi$ ). (a) Deposit map with modeled isomass 978 lines and dots that represent field measurements with colors indicating the field values of the 979 mass load, corresponding to the color bar at left. The black dashed line indicates the dispersal 980 axis of the mapped deposit whereas the solid black line with dots indicates the dispersal axis of 981 the modeled deposit (the latter lies mostly on top of the former and obscures it). The modeled 982 dispersal axis was obtained by finding the ground cell in each column of longtitude with the 983 highest deposit mass load. (b) Log of modeled mass load versus measured mass load at sample 984 locations. Black dashed line is the 1:1 line; dotted lines above and below indicate modeled 985 values 10 and 0.1 times that measured. Gray dots lay outside the range of downwind distances 986 covered by trend lines in Fig. 6 Fig. 4 and therefore were not included in the calculation of  $\Delta^2$ . 987 (c) Log of measured mass load (black and gray dots), and modeled mass load (black line with dots) versus distance downwind along the dispersal axis. The black dashed line is the same 988 989 trend line as in Fig. 7Fig. 4. Gray dots were not included in the calculation of  $\Delta^2_{downwind}$ . (d) 990 Log of mass load versus square root of area contained within isomass lines. Black squares are 991 from the mapped deposit, red squares from the modeled one.

992Figure 11Figure 12: Results of the Crater Peak (Mount Spurr) simulation that provides993approximately the besta good fit to mapped data ( $\mu_{agg} = 1.82.4 \phi$  and  $\sigma_{agg} = 0.31 \phi$ ). The features994in the sub-figures are as described in Fig. 10Fig. 11. "CP" in Fig. 11Fig. 12 a refers to the Crater995Peak vent.

Figure 12<u>Figure 13</u>: Results of the Ruapehu simulation that provides approximately the good best fit to mapped data ( $\mu_{agg} = 2.4\phi$  and  $\sigma_{agg} = 0.31\phi$ ). The features in the sub-figures are as described in Fig. 10Fig. 11.

999Figure 13Figure 14: Results of the Redoubt simulation that provides a reasonable fit to mapped1000data ( $\mu_{agg} = 2.4\phi$  and  $\sigma_{agg} = 0.31\phi$ ). The features in the sub-figures are as described in Fig. 10Fig.100111.

Figure 14Figure 15: Modeled mass load of the Mount St. Helens eruption for four cases using  $\mu_{agg} = 2.4\phi$ ,  $\sigma_{agg} = 0.31\phi$ , and different diffusion coefficients: (a) D=0 m<sup>2</sup> s<sup>-1</sup>, (b)  $3\times10^2$  m<sup>2</sup> s<sup>-1</sup>, (c)  $1\times10^3$  m<sup>2</sup> s<sup>-1</sup>, and (d)  $3\times10^3$  m<sup>2</sup> s<sup>-1</sup>. Other inputs are as given in Tables 1 and 2. Lines are isomass contours of modeled mass load and colored dots are sample locations. Colors of the dots and lines give the mass load corresponding to the color table.

Figure 15Figure 16: Modeled mass load of the Ruapehu eruption for four cases using  $\mu_{agg}$ =2.4 $\phi$ ,  $\sigma_{agg}$  =0.31 $\phi$ , and different diffusion coefficients: (a) D=0 m<sup>2</sup> s<sup>-1</sup>, (b) 1×10<sup>2</sup> m<sup>2</sup> s<sup>-1</sup>, (c) 3×10<sup>2</sup> m<sup>2</sup> s<sup>-1</sup>, and (d) 1×10<sup>3</sup> m<sup>2</sup> s<sup>-1</sup>. Other inputs are as given in Table 1. Lines are isomass contours of modeled mass load and colored dots are sample locations. Colors of the dots and lines give the mass load corresponding to the color table.

1012 Figure A1: Illustration of the path taken by coarse aggregates that fallout in proximal sections, 1013 less than a few plume heights from the source (left), and fine aggregates that fall out in distal 1014 sections (right). Among distal fine aggregates, we show the path taken by those that might have 1015 formed within or below the downwind cloud as hypothesized by Durant et al. (2009) (red 1016 dashed line), and those that were transported downwind without changing size, as calculated 1017 by Ash3d (blue dashed line). Also illustrated are some key processes that might influence the 1018 distribution of fine, distal ash, including development of gravitational instability and overturn 1019 within the downwind cloud (Carazzo and Jellinek, 2012), and the development of 1020 hydrometeors as descending ash approaches the freezing elevation (Durant et al., 2009).

- 1021 Figures S001-S004: Figures analogous to Figs. <u>1011</u>, <u>112</u>, <u>1213</u>, and <u>1314</u>, respectively, but
- 1022 with no particle aggregation.
- 1023 Figures S005-<u>S046S056</u>: Figures analogous to Fig. 10Fig. 11, but for different values of  $\mu_{agg}$
- 1024 and  $\sigma_{agg}$  given in their labels.
- 1025 Figures <u>S047S057-S088S108</u>: Figures analogous to <u>Fig. 11Fig. 12</u>, but for different values of
- 1026  $\mu_{agg}$  and  $\sigma_{agg}$  given in their labels.
- 1027 Figures S089S109-S130S160: Figures analogous to Fig. 12Fig. 13, but for different values of
- 1028  $\mu_{agg}$  and  $\sigma_{agg}$  given in their labels.
- 1029 Figures <u>\$131S161-\$172S212</u>: Figures analogous to <u>Fig. 13Fig. 14</u>, but for different values of

- 1030  $\mu_{agg}$  and  $\sigma_{agg}$  given in their labels.
- 1031 Figure S173: Figure analogous to Fig. 12, but using