Response to reviews of "Modelling the size distribution of aggregated volcanic ash and implications for operational atmospheric dispersion modelling" by Frances Beckett et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-254, 2021

Response to review by L. Mastin

This manuscript presents a one-dimensional plume model that solves the Smoluchowski coagulation equations to calculate particle aggregation in plumes ascending from the 2010 Eyjafjallajökull eruption, and then uses results of the plume modelling, incorporated into the NAME atmospheric dispersion model, to examine the effect of aggregation on the areal extent of the ash cloud. For the conditions considered, the results show surprisingly little effect of aggregation on cloud area. Less than 20% change or so in cloud area over the first 24 hours, when aggregation effects are considered. The manuscript thoroughly examines the sensitivity of the calculations to some key aggregation parameters and includes thorough sections discussing uncertainties and limitations.

The manuscript is clearly written, equations and assumptions are well presented, citations are thorough, and the conclusions are for the most part well supported by the results. I also think this subject will be of great interest to atmospheric scientists and volcanologists who are concerned with uncertainty in ash cloud forecasts. I have only a few comments:

Thank you for your thoughtful and thorough review of our manuscript. The changes you have proposed have helped us to significantly improve the paper. Please find below responses to your corrections and queries. We have highlighted all changes made in response to your suggestions in blue in the revised manuscript

1) The plumes you model condense water only in the upper kilometre (Fig. 1a & b), or not at all (Fig. 1c & d). I would expect many other circumstances when more liquid water would be present, and aggregation would be more important. You include some discussion of this in lines 511-524, but I think you could add a few more sentences to quantify the water content and put it into context with precipitating clouds and wet plumes. Hail-forming clouds typically contain about 0.1-10 g/m3 water (e.g., Heymsfeld & Musil, 1982), and we know that some hail-forming columns remove a lot of fine ash (e.g., Van Eaton et al., 2015). The water content of your plumes are expressed in mass fraction of the plume (Fig. 1), which is a little hard to compare with meteorological cloud water data.

Consideration of the liquid water and ice content in the plume with respect to meteorological clouds has been a valuable addition to the manuscript, and we thank you for this suggestion. We have changed Figure 1 to show the mass mixing ratios of water vapour, liquid water, and ice with height in our modelled plumes to enable this comparison (Line 230). We now show that, although water/ice is only present over a small region of the eruption column, the plume is very water/ice rich relative to typical mid-level mixed phase meteorological clouds. This is now discussed in Section 5.1, Lines 478-483. We also highlight the implications of not representing the removal of particles due to sedimentation of ash laden hailstones in the Discussion (Lines 483-486).

2) One reason, not discussed, for the small effect of aggregation on your results is that you keep the mass fraction of distal ash in the model at 5% of erupted mass in all cases (line 377). If you were to truly ignore aggregation in the control case (Fig. 8a), your ash mass used as input would have been several times greater than 5%. This assumption should be discussed (Perhaps you account for that on lines 446-469, but I don't understand some of that discussion (see below)).

We completely agree that by presenting results which also impose the 5% distal fine ash fraction restricted the interpretation of the results. Figure 7 now shows the grain size distribution used to initialize the aggregation scheme, which have diameters between 1 μ m and 8 mm, and the output aggregated grain size distribution over this size range. This shows that the mass fraction on grains \leq 125 μ m, which is reduced by just 2% at 00:00 UTC on the 05/05/2010 following aggregation. We no longer impose the 5% assumption, instead, we show the results when the mass fraction on the grains \leq 125 μ m is used. This change has very little impact on our results and the modelled area of the ash cloud with mass loadings significant for aviation is reduced by 2% (was previously 3%) when aggregation is accounted for (Figure 8). We have modified the explanation of the model setup in Section 4 (Lines 338-343, 347-352 and 357) to reflect this change and improved the discussion of the results in Section 5 Lines 423 -433 (see also response to your later comment).

3) It would be worthwhile to add a short paragraph at the end of Section 4 summarizing some of the key aggregation relationships in Figures 4-6 in physical rather than mathematical terms. The analysis is meticulous but sometimes hard to follow when relationships are expressed in terms of, for example, St_cr or q rather than words.

We have moved the mathematics of the scaling analysis to the Appendix and re-written the Scaling Analysis section (now Section 3.1) to explain the sensitivity of the collision rate, sticking efficiency and collision rate in physical terms. Please note that we have also modified the colour-scheme used in the Scaling Analysis plots (Figures 5 and 6).

Many less important comments are listed below. Overall, I think this is a worthy manuscript and look forward to seeing it published.

Specific comments:

Line 16: change "modelled extent" to "modelled area".

Done

Line 17: At what time after the eruption start is the modelled extent of the ash cloud reduced by 3%?

Added the following explanation to Line 17:

"24 hours after the start of the release"

Line 18: add comma after "ash cloud".

Done

Line 22: change "differs to" to "differs from" (or is this an English vs. American usage thing?)

Done

Line 91: Consider adding a sentence that explains the exponents in equation 8. Most 1-D plume models do not use this exponent when calculating entrainment.

The exponents are now given the symbol f, this has been added to Table 1 and the following explanation is provided in the text at Lines 97-100:

"the radial and cross-flow entrainment terms are raised to an exponent, f, that controls the relative importance of these two terms. Devenish et al. (2010) found that f = 1.5 gave the best agreement with large-eddy simulations of buoyant plumes in a crosswind and field observations, and we adopt this here."

Line 95: add a comma after "water".

Done

line 103: change "molecular mass" to "molecular mass ratio(?)"

Corrected in the text at line 109 and in Table 1.

Line 112: what is "n_l,ice"? The sum of n_l and n_ice?

The quantity 'n_l,ice' is meant as an 'or' condition. The code computes the total amount of moisture as the sum of vapour and liquid water, or vapour and ice. For a given height there will be only liquid water or ice, according to the temperature present in the surrounding environment. We have clarified this point at Line 119.

Line 120: add a comma before "and solid phases"

Done

Line 122: Are you assuming that there is no fallout of solids from the plume? I presume that this is the case, since you are setting dn_s/ds=0. If so, change "as there is no entrainment of solids" to "as there is no entrainment or fallout of solids". In a future version, it might be worthwhile calculating particle

fallout from the column and using the remaining ash at the top of the column to initiate the NAME simulations. You may also be able to produce a physically based vertical distribution of mass for the NAME simulations.

That is correct, we assume no fallout of solids from the plume. Line 131 has been modified as you suggest.

Thank you for the very nice suggestion for development of the plume model, this is certainly worth consideration for a future version.

Line 155 (and equations 25 and 26): it would be useful to describe physically what the terms for inertial turbulence, and laminar shear are, and why they are important. These terms are not included in some other coagulation kernels, such as those by Costa et al. (2010) and de' Michieli Vitturi et al. (2021).

The turbulent inertial kernel (Equation 25) and laminar shear kernel (Equation 26) are based on Equation 36 and 38 of Folch et al. (2016), which is a modification of Costa et al. (2010). We have added this reference in the text at Line 162.

In the atmosphere particles may collide due to velocity gradients in the air. The simplest process assumes a uniform shear field and is represented by the laminar shear kernel. The turbulent inertial kernel represents coagulation in turbulent flow due to local turbulent accelerations, which produce relative particle velocities for particles of unequal mass. There is a complete description of the different coagulation processes in Pruppacher and Klett (1996) and we now refer the reader to this text at Line 162.

Line 189: add a comma after "following a collision".

Done

equation 36: Maybe I'm just dense, but I don't understand this equation. Equation 33 suggests that St_v should range from zero to infinity. In this St_cr appears to range from minus infinity to plus infinity.

In the model of Liu et al. (2000) the surface asperity cannot be zero by construction. Usually, the asperity is of the order of a nanometre. But it can also be larger than this value. The thickness of liquid water in this theory is usually of the same order of the asperity or, of course, larger. So, strictly speaking, the theory should be used within this framework. However, in the work of Costa et al. (2010), they just took inspiration from this theory to propose a reparameterization that can be used for volcanic ash in a much simpler way, and without the detailed knowledge of quantities that are usually quite challenging to constrain.

Line 192: could you briefly define "surface asperity"? Most readers may be unfamiliar with this term.

Surface asperity can also be described as surface roughness, this has now been added to the sentence, at Line 202. Note though, as we have little understanding of the properties of liquid layers on the surface of volcanic ash particles that instead we use the parameterization of Costa et al. (2010) for the sticking efficiency, which does not require knowledge of surface asperity. We have also modified Line 203 to make this clearer.

Lines 197-207: this description of the state of knowledge of liquid bonding and its effect on the sticking efficiency looks reasonable to me. Although some current studies may improve our knowledge, (e.g., https://agu.confex.com/agu/fm20/meetingapp.cgi/Paper/688464), we are not there yet. Regarding eq. 38, I think that some studies (e.g., Telling & Dufek, 2013) may suggest that sticking efficiency is not a linear function of relative humidity, but the data are sparse.

Thank you for pointing out this new experimental study on particle aggregation. We agree that there is probably no physical reason why the sticking efficiency should be a linear function of relative humidity. Given the current sparsity of data, here we have taken this simple approach, capturing just the first order behaviour of the phenomenon (i.e., the higher the relative humidity the higher the sticking). We certainly look forward to new results which might help to better constrain collision rates and sticking efficiencies in our models in the future. To ensure this point is clear in the manuscript we have expanded our point in the Discussion section (Lines 472-475).

Table 1: comments:

(a) the term "relative velocity" is not defined for u_p and u_s, although I'm pretty sure that you mean the velocity within the plume minus the velocity component of the ambient wind field in the same direction (parallel or perpendicular to the plume axis).

Corrected. The term 'Relative Velocity' has been removed from Table 1 for these parameters.

(b) The units of E are shown as kg m-2 s-1, but Q_m has the units kg s-1 and E=dQ_m/ds (eq. 5). So it seems that E should have the units kg m-1 s-1.

Thank you for finding this error. Now corrected.

(c) I don't see a specific heat capacity of particles listed. Seems like it should be used somewhere.

Thank you for noting this omission. We have now added the definition for the bulk specific heat capacity (which includes the specific heat capacity of the particles) at Line 86, and updated Table 1.

Line 218: what do you mean by a "best-guess set of observations"?

Modified line 227 to provide clarity on the source of the plume height used:

"which are based on radar data, pilot reports, and Icelandic coastguard observations"

Lines 223-224: "The mass is distributed uniformly across the bins such that 50% is on grains with diameter <= 125 um and 36% of the mass is on grains with diameter <= 32 um." I didn't understand this statement until I stumbled on the GSD plots in Figure S1. Citing them here would be very helpful. It would be easier to understand if you simply said that the GSD was uniform in mass between sizes of 1 um and 16000 μ m (I think that's what it looks like in Fig. S1. Also, how many bins are you using and what are the phi intervals between bins?

Apologies for the confusion and thank you for the useful suggestion. I am keen to ensure this explanation is clear to both geologists and other scientists less familiar with the Phi Scale. I have modified this sentence as you propose (Line 241):

"The scheme is initialized with a GSD with a uniform distribution of mass across 14 bins, representing ash with diameters ranging from 1 um to 16 mm (Figure 2). Bins are defined on the Phi Scale, where the Phi diameter is calculated as the negative logarithm to the base 2 of the particle diameter in millimetres (Krumbein, 1938)."

The histograms in Figure 2 also show the input GSD used, and so we have modified the structure of this section slightly and directed the reader to Figure 2 when explaining the input GSD. We have also attempted to make this point clearer by adding a point to the caption of Figure 2:

"The aggregation scheme is initialized with a GSD with a uniform distribution of mass, as indicated by the dark blue bars."

Line 227: I'm a little confused by your use of the word "mode", here and in Table 3. To me, the mode is the peak in a histogram of size bins. But you've shown no such histograms in this paper, so it's hard to visualize. (--oops, I see that histograms are plotted in Fig. 2. Perhaps refer to Fig. 2 when describing the mode).

Apologies that this was confusing. We have restructured this section and refer to Figure 2, we hope this in now clearer (Lines 241 -245).

Line 263: add a comma after "10".

Done

Figure 1: Are the atmospheric soundings used in these simulations listed somewhere? I don't see them.

Here we don't use soundings, rather Numerical Weather Prediction (NWP) data, from the Met Office's Unified Model. We have modified Line 227 to make this point clearer:

"Meteorological data, used by the aggregation scheme and NAME simulations, are from the Global configuration of the Unified Model (UM)"

Figure 2: the amount of aggregation shown in these figures is surprisingly modest. Only two of the four scenarios in Fig. 1 show condensed liquid water in the plume, and the condensation starts less than 1 km below the plume top (assuming the top of the plot is the plume top, which is not clear to me).

The top of the plots in Figure 1 are indeed the plume top. This is lower than the indicated maximum 'plume' height given in the plot titles (used to initialize the scheme). That is because the plots indicate the phase fractions along the modelled plume axis, and when the plume is bent over, as in this case, the maximum height of the axis is the maximum observed plume height minus the plume radius. This point was slightly hidden in the text (Lines 231-232) and so we now also note this in the caption of Figure 1:

"The maximum height of the modelled plume axis, when the plume is bent over as in this case, is the maximum observed plume height (provided in the Figure titles) minus the plume radius."

We have expanded our discussion of the water/ice content in the simulated plumes in this study, and others, in the Discussion section (Lines 465, 471-475, and 478-486).

Line 274: change "in the former case" to "in the presence of water" (assuming that's what you mean).

Following the re-write of the Scaling Analysis section, and the introduction of Appendix A (which contains the mathematics) this sentence no longer exists.

Line 275: change "appropriate for the formation of water" to "appropriate for wet conditions"

This content can now be found in the Appendix and has been corrected on Line 525 to now read:

"Since aggregation is associated with the presence of liquid water or ice and $\alpha_{k,j}$ only depends on q and St_{cr} in the presence of liquid water, we choose values of the constituent parameters in Eq. A2 that are appropriate for this case."

Figure 4: it might be worthwhile annotating one of the curves on this plot with labels that say, for example, "d_j=d_k" at the low point, and "beta_i,j@D*d_j^4" where it flattens out on the left-hand side.

Here we prefer to not annotate the plot any further as we find that the addition of the labels clutters the Figure. We hope that the re-write of the Scaling Analysis section (associated with this plot) has clarified the results shown in this Figure.

Line 333: is the "q" after "4" an exponent?

Yes, we confirm this is correct. This Line is now in the Appendix, Line 585.

Line 335: add a paragraph break before "Turning"

Done

Line 365: "This explains why the mode of the AGSD in Figure 3 shifts to larger diameters with increasing St_cr". I can't tell where the mode of the AGSD is in this figure.

Rather than referring to the Figure we now point the reader to the Table 4 which lists the mode of the AGSDs, at Line 328:

"This explains why the mode of the AGSD shifts to larger diameters with increasing St_{cr} , decreasing q or ρ_s (see also Table 4)"

Line 378: You assume that 5% of the erupted mass goes into the distal cloud. This percentage should vary depending on the amount of aggregation. But you haven't considered that here. It would be good to acknowledge that in the Discussion section.

We agree that imposing the 5% Distal Fine Ash Fraction on our results was masking some of the impacts of representing aggregation in our modelling. Instead, we now use the modelled mass fraction on grains \leq 125 µm, which as you state, varies with each timestep, (see also our response to your second comment above.) We have modified the Discussion to reflect this change at Lines 423-433.

Lines 383-384: "Model particles are released with a uniform distribution over the depth of the modelled (bent-over) plume". Uniform from ground level to the plume top?

The Devenish scheme also constrains the plume radius, model particles are released over the depth of the modelled plume. We now point the reader to Devenish (2013,2016) for further information at Line 353:

"see Devenish, 2013 and Devenish, 2016 for an explanation of how plume radius (depth) is constrained"

Lines 399-402: I'm starting to get lost here. For Figure 9, you assume that 25%, 50%, and 75% of the ash mass consists of aggregates. Aggregates of what size? Are you assuming that each light bar in Fig. 7 consists of 75% aggregates?

Yes, that is correct, for each 'bin' 75% (in this example) of the mass consists of aggregates with an assigned lower density, 25% remains as particles with a density which depends on the size of the particle. We have attempted to improve this point on Lines 370-371: "Here we consider the case where 25, 50 and 75% of the mass on each size bin, for ash with diameters

 \leq 125 µm, is represented by aggregates."

Lines 399-402: Does the GSD calculated by the plume model distinguish between particles and aggregates? This isn't explained.

Apologies for this omission. Unfortunately, our scheme does not distinguish between particles and aggregates (something to work on!). This point was made (poorly) in the in the original manuscript: "As the aggregation scheme does not track explicitly the mass fraction of aggregates versus single grains, we must also make an assumption about how much of the mass released is represented by aggregates with the lower density"

To improve clarity, we have re-worded this sentence to (Line 368):

"As the aggregation scheme does not track explicitly the mass fraction represented by aggregates versus single grains for a given size bin, we must also make an assumption about how much of the mass released is represented by aggregates with the lower density"

Line 424: delete "Whereas"

Done

Line 430: add a comma after "eruption".

Done

Lines 439-440: "in this case, the modelled ash cloud is more sensitive to the input GSD of the nonaggregated ash at the source, than due to any change to the GSD or density of the ash due to aggregation". I agree, in this case. But you could have chosen other cases where aggregation was more important. Adding water at the vent in the 1D plume model for example would have increased aggregation in the plume. Higher or colder plumes produce hail, which scavenges fine ash (Van Eaton et al., 2015) We have now expanded the discussion to include the point that eruptions in the tropics can generate taller plumes and entrain more water, see Line 470. We also note that our scheme does not represent interaction of the ash with hydrometeors, and that removal of ash-laden hailstones could also represent a significant near source process which would be a useful to include in future schemes, Lines 483-486.

Lines 462-463: "our simulated AGSD still has ~30% of the total mass on tephra (aggregates and single grains) with diameters <125 um, which, given their size and density, would travel further than Iceland before depositing due to sedimentation alone." I'm not sure what this means. Thirty percent of the total erupted mass has diameters <125 um? The GSD in Fig. 7 contains much more than 30% grains <125 um.

Apologies for the confusion. We did indeed mean that ~30% of the total erupted mass had diameters \leq 125 µm. Figure 7 has now been modified to show the total grain size distribution (1 µm – 8 mm) at the source, to make it easier to interpret the fraction of the total erupted mass on a given particle size. We have also modified the text in the Discussion to clarify this point, at Line 423-433.

Lines 448-474: this is a long, rambling paragraph. I suggest breaking it up into shorter paragraphs.

We have significantly shortened this paragraph and improved the clarity of our discussion around the implications our results have for our understanding of the mass present in the distal ash cloud from the eruption of Eyjafjallajökull in 2010 (Lines 423-433).

Line 516: change "mass on smaller grains" to "mass of smaller grains" (or is this just an English vs. American usage thing?)

This sentence has been removed following the rewrite of this paragraph in the Discussion section.

Line 530: Is viscous dissipation the right term here? I think surface tension is the force that binds particles together, just as it keeps droplets from breaking up in water sprays (Hinze, 1955). I think of viscous dissipation as the process that converts viscous shear to heat (e.g. Hardee & Larson, 1977). But perhaps there are other meanings.

Here we prefer the term 'viscous dissipation', as water tension is not dissipative itself. It is the internal action of friction that at the very end causes dissipation.

References:

Costa A, Folch A, Macedonio G (2010) A model for wet aggregation of ash particles in volcanic plumes and clouds: 1. Theoretical formulation. Journal of Geophysical Research 115:doi:10.1029/2009JB007175

de' Michieli Vitturi M, Pardini F (2021) PLUME-MoM-TSM 1.0.0: a volcanic column and umbrella cloud spreading model. Geosci. Model Dev. 14(3):1345-1377Hardee HC, Larson DW (1977) Viscous dissipation effects in magma conduits. Journal of Volcanology and Geothermal Research 2(3):299-308

Heymsfield A, Musil DJ (1982) The 22 July 1976 case study: Storm structure deduced from penetrating aircraft. In: Knight CA, Squires P (eds) Hailstorms of the Central High Plains. Colorado Associated University Press, Boulder, pp 163-180

Hinze JO (1955) Fundamentals of the Hydrodynamic Mechanism of Splitting in Dispersion Processes. A.I.Ch.E. Journal 1(3):289-295

Telling J, Dufek J, Shaikh A (2013) Ash aggregation in explosive volcanic eruptions. Geophysical Research Letters 40(10):2355-2360

Van Eaton AR, Mastin LG, Herzog M, Schwaiger HF, Schneider DJ, Wallace KL, Clarke AB (2015) Hail formation triggers rapid ash aggregation in volcanic plumes. Nature Communications 6:7860 Powered by TCPDF (www.tcpdf.org)

Response to Review by Anonymous Referee #3

The manuscript presents the introduction of an aggregation scheme for the NAME model, which is used by the London VAAC for volcanic ash transport simulations. Particle aggregation has been in the forefront of volcanological research for the past decade due to the influence it has on the deposition of fine particles and as such the introduction of the scheme in an operational model such as NAME is a significant step. Surprisingly, results suggest a second-order of magnitude impact in the case of the Eyjafjallajökull eruption (shown as a change in the cloud area).

[1] The manuscript is scientifically-valid and the description of the scheme is comprehensive. The text is well-written, but I feel that as a whole, the paper would benefit from a change in the structure, as in its current form it becomes sprawling, with lots of short, isolated sections (in the sense that the methodology, results and a short discussion is included: Section 3, 4 and 5 in the paper). There is also an over-reliance on abbreviations and the use of letters, that although introduced (Tables 1,2), make reading the manuscript very difficult (especially Section 4), unless the reader prints out Tables 1 and 2 to keep for reference. As I wrote in my short comment I believe that with its current structure the paper feels like a better match for Geoscientific Model Development as the description of the plume model and scheme are at the forefront. Still, as the editor has accepted the submission for ACP, I would suggest making some modifications towards a more 'traditional' paper structure, which I think would be easier for the reader. Specifically, I would move Section 2 to form an Appendix and add a new Section 2 as a methodology section, that introduces (in simple terms) NAME, the plume model, and the implementation of the aggregation scheme, followed by a better organized description of the different sensitivity tests carried out in the paper.

Introduction Methodology NAME and the implementation of the aggregation scheme (including only key elements of the current Section 2, with the detailed description as an appendix with Tables 1,2) Sensitivity and scale analysis (including Tables 3-5 found in Section 3, 4) Case study details (Table 5, Fig. 7) Parametric study Sensitivity tests Scale analysis Case study Discussion Limitations Conclusions

Appendix A. Detailed description of the aggregation scheme

Thank you for your comprehensive review of our manuscript. We have highlighted all changes made in response to your suggestions in blue in the revised manuscript

One of the key objectives of this study was to develop an improved approach to modelling aggregation, which could also be used by our operational atmospheric dispersion model NAME. Our new scheme enables us, for the first time, to model explicitly the change in the grain size distribution

(GSD) of the ash due to aggregation. Given the novelty of the approach we prefer that the scheme remains within the main body of the manuscript. However, to improve the structure of the paper, and justify this approach, we have merged Sections 3 and 4 (reducing the number of 'small' sections), we have moved the mathematics for the Scaling Analysis to the Appendix, and we now provide 'physical' explanations of the results in a new Sub-Section (Section 3.1). The second half of this manuscript applies the scheme to the Eyjafjallajökull 2010 eruption. We have now renamed the Dispersion Modelling Section to: 'Dispersion Modelling: A case study of the eruption of Eyjafjallajökull in 2010' following your suggestion. We have also sought to improve the clarity and readability of the manuscript by removing the overreliance on acronyms; we no longer use acronyms for aggregated grain size distribution, Fixed Pivot Technique, Smoluchowski Coagulation Equations, or ordinary differential equation.

[2] I feel that after the detailed discussion presented in Section 3, 4, the results in Section 5 are a bit underwhelming. Of course, results are what they are, but I was expecting a larger impact (also given the Egan et al. 2020 paper that focuses on the same eruption). It might also have to do with the quantity chosen (ie the cloud area), but since the model choices reflect operational VAAC simulations this is still a valuable result. One of the reasons I am suggesting a more traditional format also has to do with being able to better evaluate the model choices (when they are all presented at the same place). In any case, I think that this should be discussed in the 'Limitations' section.

We now include the point that our dispersion model setup in this study reflects the choices used by the London VAAC, and that our results focus on the implications for the modelled extent of the ash cloud with mass loadings of significance to the aviation industry (Lines 405-407).

Some minor comments have been added in the attached PDF document.

Overall, I feel that the inclusion of aggregation is NAME is an important step forward for the community and I suggest for publication of the paper after revisiting the paper structure and discussing the case study results in more detail. I would like to wish the writers the best of luck with the revisions.

Please also note the supplement to this comment: https://acp.copernicus.org/preprints/acp-2021-254/acp-2021-254-RC2-supplement.pdf

Comments from this are:

1. I think that the word 'together' is not necessary

Corrected

2. What is surface asperity?

Surface asperity can also be described as surface roughness, this has now been added to the sentence, at Line 202. Note though, as we have little understanding of the properties of liquid layers on the surface of volcanic ash particles that instead we use the parameterization of Costa et al. (2010) for the sticking efficiency, which does not require knowledge of surface asperity. We have also modified Line 203 to make this clearer.

3. The discussion here gets a bit too abstract... I would suggest revisiting the section to adding two short paragraphs in the beginning and ending of the section to guide the reader.

We have re-written the Scaling Analysis to describe the results in physical terms, which help to explain the behaviour of the aggregation scheme. The mathematical treatment of the scaling has been moved to the Appendix. Please note that we have also modified the colour-scheme used in the Scaling Analysis plots (Figures 5 and 6).

4. I would suggest adding a one-sentence description of PC3 instead of just a citation

Corrected, Line 364 now reads:

"However, it is expected that porous aggregates, specifically cored clusters which consist of a large core particle (> 90 um) covered by a thick shell of smaller particles (Brown et al., 2012; Bagheri et al., 2016) may have lower densities than single grains of ash of equivalent size"

5. This seems like an important model limitation that should be addressed ("As the aggregation scheme does not track explicitly the mass fraction of aggregates versus single grains)".

We agree that this is an important model limitation which should be addressed during future model development. We have strengthened our point on this in the 'Limitations Section', such that it now reads (Lines 492-496):

"Our 1-dimensional treatment of the SCE does not allow us to represent the change in density of the simulated aggregates or track explicitly the mass fraction of aggregates versus single grains within a given size bin. Our scheme could be significantly improved by using a multi-dimensional description which represents the fluctuation in the density of the growing aggregates and retains information on the mass fraction of aggregated particles. This would also require a better understanding of the structure (porosity) of aggregates."

6. It's very difficult to make out differences in the plots... I would suggest trying collapsing the columns as a single panels as shown in the sketch (Panels a, b) and then adding two new panels showing the cloud area against percentage as Panels c, d.

Thank you for your suggestion. As you note, the introduction of the aggregation scheme has had little impact on the modelled ash cloud from the eruption of Eyjafjallajökull in our case-study. When the total column mass loading plots shown in Figure 9 are collapsed into a single panel, such that all the plume contours with varying aggregation fractions are shown together, it is very difficult to interpret the results. This is shown in the plots below. For this reason, we generated Figure 10, as presented in the original version of the manuscript. This shows the relative areas of the plumes presented in Figure 9 which we think provides a better visualization, which is easier to interpret.



Figure 1R: Modelled 1-hour averaged total column mass loadings of the Eyjafjallajökull ash cloud at 00:00 UTC on the 05/05/2010 when 25%, 50% and 75% of the mass is on aggregates with density 1000 kg m⁻³ and 500 kg m⁻³.

7. I would suggest adding a couple of paragraph breaks, for example at line 457 and 465

Thank you for your suggestion, we agree that this paragraph was long and rambling. We have shortened the text here and worked to improve the clarity of our points, lines 423-433.