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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Atmospheric processes affecting the separation of volcanic ash and SO_2 in volcanic eruptions: Inferences from the May 2011 Grímsvötn eruption" *by* Fred Prata et al.

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Response to reviewer 1

We thank John Stevenson for a thorough and thoughtful review of our paper. He raises many interesting and helpful issues and here we address these.

General

Many of his comments are directed towards two main areas:

1. Column collapse and aggregation.
2. Errors and their sources in satellite estimates of mass loadings.

In both of these areas he refers to his own body of published work and asks us to consider our results in the light of his own results. Our rebuttal begins with his Section 2.

Section 2: Contribution

Stevenson argues that some statements are not well supported and should be dropped. Our item-by-item response follows.

1. We agree with this statement and have added a statement along these lines in the conclusions.
2. "Separation of material". It is difficult to imagine a case in the atmosphere where wind shear would not lead to horizontal separation. Perhaps, Stevenson interpreted our findings to mean vertical separation? We have modified our statements to make it clear we are referring to horizontal separation of gas and particles.
3. We have modified our description of column collapse and acknowledge that the processes we describe may not fit well with the standard description of a column collapse. We did preface our description with use of the word "partial", but have now also explicitly described the observations more in line with Stevenson's suggestions.

4. Stevenson seems to suggest the ash cloud was a significant threat to aviation. This does not agree with the observations. There were no reported aviation incidents and the ash was not of high enough concentration when it crossed into aviation flight routes. Note that flights across the North Atlantic/North Sea typically use altitudes >20,000 ft (except at take-off and landing). The dispersing ash cloud was confined to altitudes below 20,000ft.
5. We have added the words “under the assumption of dry standard atmosphere conditions” when referring to the Mastin relationship.

Section 3: Suitability for publication

We have made the necessary revisions as described in this rebuttal.

Section 4: Areas for revision

4.1 Missing reference. We have included a reference to Stevenson et al. (2013). The first two comments in this Section need no comment because they are statements and do not seem to contradict our paper.

Ash deposited in Scotland. The third comment concerning the deposition of ash of 19-23 μm median grain size with maxima of 80 μm is interesting but largely irrelevant to our paper. Without going into a detailed analysis of Stevenson et al.’s results we simply state from his own paper: “The sizes of measured grains had modes of 25–30 μm although grains $<$10 μm were not counted, and those close to this minimum size were more likely to be missed.” This kind of sampling bias is sufficient to reconcile any differences between the two independent methods of estimating particle sizes. Stevenson et al.’s methodology counts no particles with sizes $<$10 μm and the satellite retrieval scheme detects only particles with sizes $<$10 μm. However, we stress that the satellite is measuring within the atmospheric plume and needs to make no assumptions about how the particles reached their location. While the infrared sensors discriminate ash on the basis of the BTD signal, and the sensitivity of that technique relies on small particles (effective radii $<$16 μm or so), the detection of particles relies on detecting a change in optical depth. This difference between detection and discrimination is critical. Large particles in sufficiently high concentration will cause a detectable change in optical depth and hence will be detected by the infrared satellite sensors. It is nearly always the case that these particles reside in the most optically dense part of the plume and that part is generally closest to the source. A dispersing plume loses mass by deposition (mainly) and the large particles are removed. One can imagine scenarios where large particles co-exist with high concentrations of small particles and are somehow hidden from discrimination (but not detection). We admit such cases may occur but so far there has been no evidence presented of these scenarios. The occurrence of large grains deposited in northern Scotland is consistent with the satellite retrievals that suggest only the small particles remain in the plume. The route taken by these large particles may not be the same as the route followed by the small particles. Indeed it is possible that the large particles have arrived via a different, more tortuous route, re-cycled by the atmosphere and advected by winds at different heights. In any case, we do not agree with Stevenson et al.’s assertions that larger particles exist in plumes and go undetected by a variety of different sensors (satellite IR, active lidar, in situ aircraft particle counters), which we feel are unproven. In contrast, there is evidence from independent research that is in strong support of our analysis. Moxnes et al. (2015), for example, show aircraft measurements of the particle size distribution near the plume on 22 May (see their Figure 15). The particle diameter measured (and modelled) ranges from 1–20 μm with a peak in the distribution of $\sim$5 μm. This is in broad agreement with our satellite retrievals (r effective of 5–7 μm). This leads us to conclude that the methods adopted by Stevenson et al. are largely incompatible with measurements of particles in plumes. There is a clear need for a scientific study to fully understand the causes of these disparate results and inform the interpretation of measurements of grounded particles in conjunction with satellite measurements of suspended ash particles.

Pollen traps and trajectory analysis. It is hardly surprising that small (tiny?) particles with sizes of 3–5 μm were detected in raindrops, after all these are the size range for particles that the satellite IR sensors detect (and detected in this case). The trajectory analysis is however, suspect. Figures 1a and 1b of Stevenson *et al.*'s paper seem to show that the southern trajectories that bring air over Scotland begin at low altitudes. The air that passes over Greenland stays there or moves north, west or east (Stevenson *et al.* comment that trajectory analysis is consistent with smaller particles arriving in the UK via Greenland on 27 May, but no data were provided). We feel this needs no further comment as our rebuttal is not concerned with the interpretations of Stevenson *et al.*'s body of research. On the point about why the IR retrievals did not detect ash over Greenland – we simply note that they did detect some ash and this has been reported elsewhere (Prata and Rose, 2015; see Figure 52.11).

4.2 Discussion of plume "collapse". We agree that our interpretation is speculative and have tidied up some text (including terminology) as recommended by Stevenson. We have also added a citation to Jude-Eton *et al.* (2012).

4.3 Uncertainty estimates for ash retrievals. Stevenson goes into some detail to suggest that satellite retrievals are much less certain than we have stated. We think in general this may be true and there are many assumptions relied upon, which might not be appropriate in all cases and could be improved with further study, and these contribute to the uncertainty in the retrieval. However, this paper deals with one case, the 2011 eruption, where we are more certain of the error estimates used. Moreover, rather than requiring a higher error, the validation data for suggests the satellite

retrievals have a lower error than the conservative estimates we have adopted. Prata and Prata (2012) report air quality validation data for the SEVIRI satellite retrievals (see Figure 18 of that paper). Four of the five validation points show the satellite retrieval is higher than or close to the PM10 measurement. Only one point is lower. It is also the case that validation data for Eyjafjallajökull using the same retrieval scheme gives results within $\pm$0.2 mg m^{-3} of independent data. For a retrieval with concentrations of 0.2 mg m^{-3}, the accepted lower limit of detectability for current IR broadband imaging sensors (hyperspectral sensors may do better), the error is $\pm$100%. But for concentrations that matter to aviators ($>$2 mg m^{-3}) the error is $\pm$10%. Stevenson *et al.* (2015) relies on simulated imagery, which is subject to greater error than actual observations. Here he repeats the common misconception of the way the IR sensors are used to detected ash clouds. Clouds (ash or otherwise) with high concentrations of large particles are always detected by the IR sensors. The retrievals cannot be confidently performed when there is no BTD signal, but satellite retrieval practitioners and experts do not claim to retrieve mass loadings in these optically thick parts of the cloud. The mass loadings are provided on a pixel-by-pixel basis and vary across the plume. There is no underestimation (or overestimation) within the error bounds provided for these retrievals, and for the optically thick parts or where meteorological cloud interferes, no retrieval is made. So as with any measurement system, when the conditions under which the retrieval methodology is met, an error estimate can be provided. In optically thick clouds there is no means to provide an error estimate. This comment also needs to be stressed with regard to other assumptions, for example dense spheres and the presence of meteorological clouds. In the case of the dense sphere assumption, the "sphere" part is used for Mie calculations and the "dense" part is a scaling factor. If the ash density assumed is incorrect by 50% then so is the mass loading. The retrieval methodology used is state-of-the-science and further study is required to improve the assumptions. Information on odd shaped particles, shards, asperities, bubbles, density, compositional uncertainty are not readily available and we use what is currently accepted. One could assume all ash particles are plates and shards and large (grain size >32 μm), but the retrievals would not agree with the validation data. We note also that dispersion models rely on similar assumptions concerning the density, shape and size distribution of particles in order to get accurate transport.

With regard to the last comment by Stevenson in Section 4.4, we see no discrepancy here. As explained earlier, the ash deposit measurements sample a different portion of the size distribution to that sampled by the IR sensor. In fact we would argue that the ash fallout is irrelevant to our study as necessarily this is ash that is not in the plume (it has fallen out – recall if it were still in the plume then the IR optical depth would be affected).

4.4. Discussion of failure to detect ash grains in the upper plume.

This is a curious comment because the IR sensors do detect ash in the northern part of the plume. Stevenson *et al.* (2013) do not demonstrate that pollen sensor data show that ash was present in the upper part of the plume. At best it is speculative based on a trajectory analysis (that was not shown). Their own analysis shows it was the southern part of the plume that crossed Scotland, and this is also shown in our paper. With regard to general mechanisms for lack of detection of ash by IR sensors, there are many, of which Stevenson suggests some. We do not agree with the suggestion that "low Earth surface temperature" will limit the potential for BTDs. It is not the surface temperature that matters; it is the difference between the background temperature (in the case of satellites it is usually the Earth's surface, but could be a cloud deck), and the temperature of the ash cloud or plume. If the surface is colder than the ash cloud, a BTD is still detected but has the opposite sign (see Becket *et al.*, 2017 who make use of this for remobilized ash). When the thermal contrast between the ash and the underlying background are the same, the BTD vanishes and no ash discrimination or

quantification is possible. In any case this northern branch of ash was detected and the signal soon dissipates – probably because the ash was of low concentration and soon falls below the detection limit of the sensor.

5. Plume height time series.

Time series of plume height are already published by Petersen *et al.* (2012) Figure 5 and Moxnes *et al.* (2015). It is difficult to justify repeating that data in our paper, when it is already clearly presented, accessible and referenced in our paper.

Line-by-line comments.

2:20 We have added a comment on this.
5:20 Thanks.
7:20 Both methods have shortcomings. The radar has a vertical resolution of between 2–5 km at a range of 75 km (see Figure 2 of Petersen *et al.*, 2012) and the satellite method depends on pixel resolution and of course solar and satellite viewing geometry, and can be ~±1 km. The differences between the two methods are within their respective error uncertainties.
8: table 1 Done.
9: figure 3 We agree that a new plot of the height time series would be nice but as this is already published (see Figure 5 of Petersen *et al.*, 2012 and Figure of 3 Moxnes *et al.*, 2015) we find it difficult to justify.
10:5 We have moved some to the Figure caption as suggested.
11: figure 5 (a) We can't see the relevance of the direction of travel of the satellite, but have added that information in the caption. The paper by Prata *et al.* (2016) describes the AIRS BTD technique (several bands are used). (d) the peak at ~5 km is also ash

(see Figure 5(c)). This is ash over the sea ∼64 °N – it is not over Greenland and is not sulphate. The black lines show the height range over which the parameters were calculated. We have added an explanation in the caption.

12:5 Comment added in text.

14:5 Agreed. We have defined our meaning of “fine”.

14:12 Agreed. This has been added to the text.

14: figure 7 This is not an underestimate.

15:5 References added.

16:9 We disagree and have left this unchanged.

16:15 References added.

16:17 The causes of positive differences are mostly associated with water vapour and thermal contrast effects. There is no other literature that we are aware of discussing positive differences in this product. The point of the sentence is to emphasize that the strongly positive anomaly over is unusual.

16:27 This is very difficult to state without including a large amount of explanatory text. Since there are more than 2000 sounding channels, there are over 2000 level peak contributions. We have added a general reference on this topic which concerns remote sounding and information content.

18:13 Yes we were sloppy in our use of terminology and have changed to PDC and added a reference to a discussion of the different uses of these terms.

18: figure 10 It is difficult to unequivocally assign a source to the ash that is transported in the atmosphere at low altitudes. We hypothesise that the source of the ∼3 km ash that eventually found its way to Scandanavia is a combination of ash from the partially collapsing column and some ash fallout from above. We note also that the image of the eruption column in Figure 1 shows ash separating from the main column above the ground but at low altitudes. There is no strong evidence of PDCs in the deposited material, suggesting much of the ash separating from the column did not feed PDCs on the ground. Unsteadiness of the column and the source could also contribute ash at low levels.

19:1 Of course we do not mean to say that the ash is static during this period, and we expect that atmospheric transport processes (e.g. buoyant transport, advection by the low level winds, particle settling) are acting on this ash cloud. We note that the winds were not strong and so the ash moved slowly. The cloud would also have been fed by new ash from the on-going minor eruptions. It can only be advected away if there are winds to do the advection.

19:5 We agree that the term ‘collapse’ is commonly related to total column collapses, but note that we preface this with partial collapse or even collapses. We have added a sentence to clarify this picture.

19:10 We have modified the sentence to read “The photograph shows a short vertical section dug into the deposit with evidence of hail that is collocated with ash, and suggesting removal of water.”

19:16 Yes this is a good alternate term and we have added it to the text.

It is difficult to estimate the altitude of the skirt as the brightness temperatures can’t be used and there is no shadow. It is unlikely to be higher than ∼4 km, although there could be a lower concentration of ash above (undetected and likely undetectable). Again there is not sufficient data to state the lower height limit of the “skirt” and it may extend to the ground.

20:5 Yes we have moved this part.

20:10 Figure 1 is very direct evidence that separation is occurring from the convective column.

22:figure 13 We have added a shaded region to each panel indicating the presence of liquid water. It is probable that aggregation can also occur above this region, where all condensed water is expected to be ice, but we expect the rate of aggregation to be higher when liquid water is available.

23:11 We are discussing separation of gas and particles. We have rephrased this as: “The vertical separation of gases and particles in volcanic eruption columns occurs frequently and if it occurs in the presence of wind shear it is inevitable that this results in a lateral separation of gases and particles distally. Wind shear is ubiquitous and significant when eruption columns extend to the tropopause and consequently it should be expected that some separation will occur."

23:23 Agreed Sentence changed to: "The plume model that we use here to analyze the transport of particles in the eruption column highlights the importance of multiphase processes, particularly the role of water in vigorous eruption columns."

24:figure14 No. The VAG shows clearly that ash was forecast over the Norwegian sea up to FL200 (20000 ft or 6-7 km). There is no evidence of ash at that location in the observations. The VAG does not agree with the observations. This is not a trivial academic point. Closure of airspace there prevented helicopters from reaching oil-rigs and had an unnecessary economic impact. Stevenson's comment highlights an important concern, that scientists looking at the same graphic can make completely opposite inferences. This demonstrates the (intentionally) low information content of the VAG.

24:1 Three references provided. But actually they can be generalized because the ash was detected in Norway, Scotland, Sweden, Holland and northern Germany. So everywhere else there was less concentrated ash or none.

24:4 The wording implies northern Europe and needs no modification. Stevenson's argument about ash concentrations contradicts the evidence. The PM10 stations show concentrations $<$500 μg m^{-3}. Why assume a thickness of 1 km? Why not 3 km? Then the values are more like 300 μg m^{-3}, which agrees with the observations. Tesche et al. (2015) also report lidar measurements in the range 150–340 μg m^{-3}. Moxnes *et al.* (2015) report values $<$ 100 μg m^{-3} based on aircraft data and modeling. These data, our data, previous measurements from Eyjafjalljökull (lidar, airborne and ground-based air quality) all provide adequate support for the assumptions we use in satellite-based IR retrievals. Our statements match closely to what was observed. Our error estimates for the eruption are robust and should not be extended to all ash retrievals for any other eruption.

24:15 Thanks.

25:2 Agreed.

25:14 It is less likely to cross hemispheres or even reach low latitudes from a high

latitude eruption, but zonal dispersion is likely. Hemispheric spread (in this context meridional and zonal in one hemisphere) is more likely, the longer the aerosol remains in the atmosphere. This was the point of the sentence.

25:15 Sentence added.

27:equation 1 sgn signifies the mathematical signum function. It is standard notation, -1, if x <0.

27:26 Agreed. We note that the variation in density of solids (from ~700 kg m^{-3} for vesicular pumice to ~3200 kg m^{-3} for glass shards) does not greatly alter the critical fall-out velocity calculated using our reference density (1200 kg/m3) with changes in the value by a factor of 0.76 to 1.6.

28:1 The rest of the paper does not disagree with the radar heights.

28:16 Arason *et al.* (2013) have presented this data at a scientific conference and have made this presentation available through their personal website. The ash in ash-infused hail ranges in size from 2 microns to 10 mm with a peak for 0.2–1 mm. This might form a useful later study.

S1:6 Because we are only modeling the "ashy" part. There was very little, if any, ash from the upper part of the plume that was transported southwards.

S2:2 It is a unimodal particle size distribution.

S2:5 Standard parameters were used as specified in the FALL3D documentation, but as mentioned many runs were used.

S7:figure S4 There isn't just one geometric standard deviation assumed. The scheme generates a large number of brightness temperature simulations based on Mie calculations that assume different effective particle radii and geometric standard deviations for the lognormal distribution. The procedure finds the best fit between the observed and simulated brightness temperatures.

S8:figure S5 As above.

References

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