Atmos. Chem. Phys. Discuss., 11, C8780–C8792, 2011
www.atmos-chem-phys-discuss.net/11/C8780/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "CARIBIC aircraft measurements of Eyjafjallajökull volcanic plumes in April/May 2010" *by* A. Rauthe-Schöch et al.

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Received and published: 12 September 2011

General comments

The study is of good scientific significance and provides some unique in situ measurements of far-field volcanic cloud characteristics. The scientific quality is good, taking into the point mentioned below. The presentation quality is good, although a small amount of reworking is required.

Specific comments

1. Suitability of CARIBIC for volcanic ash cloud characterisation: Volcanic clouds contain a mixture of solid and liquid aerosol particles, and silicate ash particles that collectively span over seven orders of magnitude in size (Durant et al., 2010). The size

of airborne ash measured as a function of transport distance from the volcano is dependent on the fraction deposited at the surface, which is influenced by particle aggregation and gravitational settling. The total fraction of tephra emitted during explosive volcanism that is finer than 5 microns is not well characterised, but it is incredibly small («5 wt%) (Mastin et al., 2009). Furthermore, the size distribution of tephra measured at the surface is often multi-modal and results from a combination of processes during initial fragmentation, transport and deposition (Wohletz et al., 1989).

The major uncertainties with the CARIBIC particle size measurements are: (1) sampling bias and particle size cut-off (changes as a function of airspeed); (2) refractive index of particle dispersion; (3) shape of particle size distribution.

Issue (1) has not been addressed with respect to airborne volcanic ash. On P. 16702 L.19-20, the authors state, "After implementing the sheath air technique, the maximum counting efficiency of the OPC unit increased from âĹij50 % (manufacturer value) to âĹij89 % (see Fig. 2b)". Please explain the "sheath air technique": does this confine the aerosol air sample within the OPC only, or is the sheath air introduced at the sampling inlet and along any tubing to the instrument? If the former, then it is conceivable that some fraction is lost before the sample air arrives at the instrument. Figure 2 appears to show that the size calibration and counting efficiency is for upper tropospheric aerosol sampled over South Africa on 14 November 2010. Therefore, it is misleading to write the efficiency of the OPC is 89 % with respect to volcanic ash particles. Furthermore, volcanic silicate ash particles are different from background atmospheric sulphate aerosol (size distribution, composition, density, phase, shape) and there are 3 separate issues with respect to volcanic ash measurement that will need to be addressed through a dedicated laboratory calibration: (1) sampling bias and cut-off introduced by the externally-mounted sampling inlet (this would include ash particle break-up caused by the inlet nozzle); (2) losses during transport from the sampling inlet to the instrument; (3) counting efficiency of the OPC. The reality is that the majority of the ash particle size distribution lies above the maximum particle size cut-off of the

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CARIBIC sampling inlet, so there is considerable extrapolation required to extend the measured size distribution to cover the full range of particle sizes encountered in ash clouds.

Assumptions surrounding issue (2), the refractive index of volcanic ash clouds, also introduces a large amount of uncertainty, which could be stated more explicitly on P. 16701. In order to infer a particle size distribution from OPC measured scattering, a refractive index is assumed. It is questionable how the refractive indices can be generalised as described on P.16700 - the instrument package collects samples so why not use these to directly determine the RIs?

Please clarify how the ash refractive index was determined equal 1.54-0.003i. The authors state that this value was compared to Schumann et al. (2011), who in turn refer to a paper by Sokolik and Toon (1999) as the source of their data for volcanic ash refractive index (S&T 1999 does not contain any data for volcanic ash – only SiO2, which is not the same thing!). More explanation and justification is needed here please, as the methodology is not reproducible as written. The refractive index calculation will need to account for the coexistence of H2SO4 droplets, silicate ash particles, and sulphate coatings on the ash particles. Further complicating the problem is the fact that the amount of ash versus sulphate aerosol present varies between eruptions, and will be heterogeneous through space and time for a given volcanic cloud. The calculated refractive index is also dependent on the mixing assumption used (see Bohren and Huffman, 1983). Data for volcanic glass refractive index are provided by Pollack et al. (1973) and Volz (1973), although these were measured on bulk macroscopic samples and may not exactly represent the RI of fine grained silicate glass suspended in the atmosphere. Therefore it is challenging to see how a single assumed RI can be applied over all the data reported in the paper simply as in-cloud "volcanic ash" versus background "ammonium sulphate aerosol" refractive indices. I would argue that this introduces large errors. Schumann et al. (2011) estimated uncertainty of up to a factor of 3 in the imaginary part of the refractive index. I suggest that a detailed explanation

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detailing all steps in the RI calculation is needed as the reported OPC particle size "measurements" hinge on this major assumption. Ideally the RI would be determined from mineralogical examination of filter samples collected (as reported in Section 2.2) at the same time as the OPC measurements were made, and I would expect that the RI would change over different segments of a given flight through the volcanic ash cloud.

The shape of the airborne ash particle size distribution, issue (3), as a function of distance from the volcano again depends on the eruptive style (which generates a characteristic particle size distribution), and sorting during transport due to cloud microphysical processes and deposition. There are assumptions on the particle size distribution shape commonly used in satellite remote sensing: Measurements of ash particles at the periphery of the 1980 Mount St. Helens ash cloud indicated that a log-normal (or Zold) size distribution provides a satisfactory fit (reported in Newell and Deepak, 1982); while measurements of the 1982 El Chichón stratospheric aerosol layer (which had a subordinate ash fraction) indicated that a modified-γ size distribution is more appropriate for this type of volcanic cloud (Hofmann and Rosen, 1984; King et al., 1984). Fallout from the Mount St. Helens eruption was multi-modal within the first 300 km of the volcano (Durant et al., 2009), as was fallout from Eyjafjallajökull (Figure 1 in Stohl et al., 2011). On P.16702 the authors assume an exponential particle number size distribution - please explain the basis of this assumption, and how it relates to observed ash cloud particle size distributions.

In conclusion, it is not absolutely obvious that the CARIBIC OPC is a useful tool for in situ measurement of airborne volcanic ash particle size distributions – it has significant sampling bias and losses, there are large uncertainties associated with the refractive index, and a substantial amount of post-analysis is required. The statement in the abstract relating to "special mission flights" which presents the CARIBIC instrument package as a "versatile and comprehensive flying laboratory" (also repeated on P.16723 L.16) is overstated and misleading (it should probably be removed); the time required for post-analysis precludes the use of CARIBIC in an operational setting (for

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example, the background ammonium aerosol RI determination was based on data collected 6 months after the Eyjafjallajökull eruption, and it has taken over a year to take this analysis to publication). Furthermore, it is not safe to fly jet turbine-powered aircraft in volcanic ash clouds so it is unwise to advertise this methodology as a potential tool that could be implemented in a future ash cloud crisis.

Finally, the last statement in the abstract beginning L.21 could actually be turned around (and also the statement on P.16724 L.22): Lagrangian models such as FLEXPART have been refined over decades and perform well in simulating the dynamics of the atmosphere. The largest uncertainty is introduced from uncertainty in the source term; however, this can now be reduced through implementation of the inversion methodology reported in Stohl et al. (2011). It would be my conjecture that in fact the largest errors may be measurement-based due to the issues discussed above, with respect to in situ measurements versus modelling. From the modelling perspective, the ability to discriminate thin layers is dependent on the vertical resolution used; enhanced resolution is simply a trade off against increased computation time.

2. RNMI TRAJKS back-trajectory analyses used to determine volcanic origin for sample air: It may seem obvious, but the CARIBIC instrument payload measures SO2, so clearly, when elevated SO2 was encountered this could be used as an indication of a volcanic cloud encounter. The authors should also be more assertive and refer to volcanic cloud encounters when the aircraft sampled the emissions, as opposed to referring to samples with "a volcanic influence" (e.g., Figure 1 legend) . . . that kind of statement is ambiguous. The criteria for determining a volcanic origin of sampled air included: (1) an eruption height threshold of less than 9 km; (2) the back-trajectories pass within 200 km of the volcano; and (3) "To assess the uncertainty of the calculated backward trajectories, the starting point is shifted in latitude and longitude by $\pm$0.4âŮę and in pressure by $\pm$3 %. If all of these 15 back trajectories stay close together, the trajectory is viewed to be well defined".

Figure 2 of Stohl et al. (2011) shows both a priori emission (from observational sources

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such as radar and a simple plume model), and a posteriori emission profiles. There are periods when emissions reached in excess of 10 km. From the information provided on P.16708 L.10 onwards, it is not possible to determine the possible times that CARIBIC samples may have originated at the volcano. It should however be possible for the authors to relate their back-trajectories more quantitatively to the time series presented in Stohl et al. (2011) to further reduce the uncertainty. Please also clarify the text in this section: what do the 15 trajectories correspond to? Why was the emission source moved by $\pm$0.4âŮę and pressure varied by $\pm$3 %? What relevance are these amounts? This section could be strengthened by the addition of a series of figures (one for each flight) illustrating back-trajectories for some or all of the sampling locations. Also, please show the VAAC forecast overlaid on the flight trajectories (e.g., in Figure 1) and the model forecasts mentioned on P.16707. Perhaps this requires a dedicated figure. The threshold for volcanic emissions originating within 200 km of the volcano is too high. This should be 10s km maximum. Why was the threshold set so high? Why not run a forecast model constrained by reanalysis wind fields and determine if the modelled air masses coincide with the aircraft trajectories? This seems to be what the FLEXPART simulations do, so it is not clear why this section on RNMI back-trajectories has been included.

3. The structure of the paper currently reads as a chronological report of the research flight activities. The conclusions stick to this format which is not particularly enlightening. Please reword and summarise the observations in each category (particle size, mass distributions, composition, any inferred chemistry, etc.) as a function of (1) the distance from source; and (2) the age of the emissions (both parameters determined from the trajectory modelling). Then it will be possible to compare the various factors for each volcanic cloud encounter. Also please add the same information to the abstract.

Technical corrections

P.16695 L.2 The volcano is known only as Eyjafjallajökull - the abbreviated versions

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mentioned add confusion so please remove.

P.16695 L.5 There were several phases in the period 14 April to 22 May, with some notable hiatuses during that time (which separate individual phases of the eruption).

P.16695 L.8 Which phase(s) of the eruption do these estimates of particle size fractions relate to? It is not satisfactory to generalise over this whole period.

P.16695 L.20 Should this read "southeastward-propagating winds" or "northwesterly winds"?

P.16698 L.8 Please change "even to a complete blinding" to "completely opaque".

P.16698 L.17 Please remove all references to "new" or "brand new" instrumentation here and in several section headings etc. . . - it is irrelevant how old the instrumentation is. Is this mentioned because the OPC failed on the second flight? If so it is not necessary.

P.16699 L.5 Is this paper submitted or in press? Better not to cite papers in preparation.

P.16699 L.13 Please change "wrong" to "errors in".

P.16702 L.24 Are there really significant numbers of ash particles this small (137 nm)? This seems more likely to be sulphate aerosol. If this is ash, please show imagery of samples collected from the cloud to back this up.

P.16705 L.6 Check grammar.

P.16705 L.17 Start new section here on the back-trajectory modelling (doesn't belong in "other instruments").

P.16707 L.19 Please change "runs" to "issues".

P.16708 L.4-7 Check grammar!

P.16707 L.29 There is also the SAVAA resource (www.savaa.nilu.no).

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P.16708 L.4 Check grammar.

P.16708 L.15-L.26 Please reword this paragraph more clearly.

P.16709 L.3 How did the impactor samples indicate volcanic origins for the air masses? More explanation needed please.

P.16709 L.11 please be quantitative - how much did mass increase? What were the average mass concentrations for volcanic versus non-volcanic air masses?

P. 16709 L.14 Are there aerosol samples and/or SEM imagery to back up the statement that there were submicron-sized ash particles? Please provide evidence.

P.16709 L.26 2.5 wt.%?

P.16710 L.5 Contradictory statement here: the DOAS measured an "enhancement" in SO2 (again please be quantitative), yet on L.7 it is written that the signal is within the noise. Therefore, how can the signal be extracted from the noise with any confidence? Also, there are clearly particles in the cloud as measured by the OPC - how does multiple scattering impact the DOAS retrievals? Please provide a more critical analysis of the DOAS results, or remove this text.

P.16710 L.11 The CPC measurements are interesting and perhaps shed light on sulphate aerosol and capacity to act as CCN - can you please put these data in the context of background CCN? For example, is there an enhancement in the volcanic aerosol layer (please be quantitative)?

P.16710 L.20 The impactor sample compositions are interesting – can you please tabulate all data for each flight? This would make it easier to observe trends.

P.16711 L.6 How much SO2? Are the authors suggesting that there should be a general correlation between SO2 and BrO?

P.16711 L.23 "...were affected by volcanic emissions" - ambiguous statement.

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P.16712 L.6 Please show the predicted ash localities on the relevant figure.

P.16712 L.15 "before the double peak" - please reword as it is not very clear what is meant.

P.16713 L.22 The increase does not appear to be that "strong" relative to the background curve - they look similar; the volcanic distribution is offset to higher masses. Perhaps the curves look similar because both are based on the same exponential function on P.16702 (except using different fitting parameters). On Figure 7 please write "measured" on the upper plot and "simulated" on the lower plot.

P.16714 L.23 This paragraph and the next should come earlier in the methods section.

P.16716 L.29 Please reword this section - it probably isn't necessary to go into detailed description of the second derivative of the size distribution curve in qualitative terms.

P.16717 L.1 Ash particles with sizes of 12-14 microns are not very large; in fact this is very fine ash (which by classification can reach 2000 microns diameter). It would be more appropriate to cite a different paper here that focuses on the sedimentology of the Eyjafjallajökull eruptions.

P.16717 L.18 This paragraph should be written in the past tense.

P.16718 L.25 Please provide a quantitative comparison, i.e., tabulate the data from the current study and Sigmundsson et al. (2010).

P.16719 L.8 "there is a hint" - please reword. Surely all air in volcanic clouds is a mixture of ambient atmosphere and emitted volatiles?

P.16719 L.21 Please define "small" ash particles. The size distribution changes are related to changes in eruptive style . . . a more explicit way of stating this.

P.16719 L.24 Ash particles tend to aggregate and settle collectively at a rate faster than single particles. It is misleading to suggest ash with diameter 20 microns will sediment at that rate. This does however work in favour of the argument that much of the fine

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ash (<63 microns) will fall out close to the volcano. However, there will be some larger particles carried much further from the volcano due to aerodynamic drag resulting from irregular particle morphologies and aggregate "rafting" (Sorem, 1982). Therefore the >20 microns fraction cannot be ignored.

P.16720 L.12 this paragraph is confusing (please reword), e.g., "second ash plume" . . . should this be "second volcanic cloud encounter"? Also, are the 3 curves really that different? What is the uncertainty on the measurements? (not shown on the figure) New particle formation may be inhibited by a lack of SO2 and/or water (Bekki et al., 1996).

P.16720 and elsewhere Constant references to volcanic plumes; the aircraft were 1000s km from the volcano. These were not plumes (attached to surface). Please use "volcanic cloud".

P.16721 L.1 This paragraph begins by comparing CO measurements made at fumeroles (at source) in USA and Antarctica with airborne concentrations 1000s km from source (extreme distal) from an Icelandic volcano (all different compositions). The volcanic systems are not comparable and neither are the sampling locations. Why does the measurement of 80 ppb "seam reasonable"? There is also brief reference to measurements of halogen species and CO in the Eyjafjallajökull cloud in relation to the impact on ozone chemistry. Please discuss the CARIBIC measurements in the context of Rose et al. (2006) and Millard et al. (2006).

P.16722 L.19 Is there any published evidence that SO2 causes "enhanced corrosion" of aircraft?

P.16723 L.2 Please also remind the reader of the many uncertainties associated with the in situ measurements. Was the source term well constrained in this study? If so by what method? The largest errors in modelling come from uncertainty in the source term, not simulated transport. Also, models tend to over-predict far-field ash concentrations because aggregation is not accounted for. Therefore, forecast ash cloud hazard

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areas will be overly conservative by nature.

P.16723 L.12 Please change "was not particularly designed" to "is not optimised".

P.16724 L.26 Could add reference to Prata and Thomas (2011) who chronicle phases of the eruption where ash and SO2 were collocated, and visa versa. Also, please remove or back up the "corrosive" statement.

Figures 9 and 10 Peak CPC derived number concentrations are in the last few minutes of the flight; please discuss the origin for this in the text. This dwarfs the "in cloud" measurements.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 16693, 2011.

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