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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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Reply to reviewer Adam Durant

General comments

We would like to thank Adam Durant for his thorough and extensive review of our manuscript and his helpful suggestions for improving the manuscript. In the following, we will answer his specific comments. The reviewer's comments are written in italic and our answers are given below each comment. The references can be found in the revised manuscript.

1. ... 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.

In the following, we will address these three major points:

(1) Sampling bias and particle size cut-off

We have introduced a new subsection "2.1 Aerosol particle inlet system" which explains the inlet system in more details and points out that care is taken to avoid inlet tubing wall effects as much as possible.

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 ?
We explain now in "2.1 Aerosol particle inlet system" that the sheath air is only introduced $\sim$10 cm in front of the OPC optics. However, the OPC air is extracted only from the core of the inlet tubing to avoid wall effects. This acts in a similar way as the sheath air used in the OPC.

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.
The counting efficiency gives the information on the fraction of particles detected by the OPC. It is mainly determined by the gas flow properties and the optics geometry, as e.g. the increase of the counting efficiency by introducing the sheath air technique shows. As long as the particles are clearly larger than the lower particle size detection limit (which is the case for volcanic ash particles) the particle material does not affect the counting efficiency strongly. Hence the laboratory derived counting efficiency can also be used for volcanic ash particles. However, the particle material primarily determines the particle sizing by the OPC and this is discussed and dealt with in the section about the particle refractive index.

The Flight on 14 November 2010 to South Africa was used to check the different OPC calibrations (mid latitude UT aerosol, LS aerosol, and tropical/subtropical mid-tropospheric aerosol). It was found that applying the three different calibrations to one data set gives a total change in the particle mass of $\sim \pm$10%. We did not use this flight for calibrating the OPC ! This has been made clear in the revised manuscript.

(2) Refractive index of the volcanic ash particles

Following the suggestion by Reviewer #2, we have now performed a sensitivity study using three refractive indices: n = 1.50 − 0.01i, n = 1.55 − 0.001i and n = 1.60 − 0.0001i. These values span the range of refractive indices that have been reported for ash from the Eyjafjallajökull (Schumann et al, ACP, 2011; Bukowiecki et al., ACPD, 2011; Kandler, personal communication, 2011; Shoji et al., 1994). The largest ash particle masses are calculated using the smallest real part/largest imaginary part of the refractive index. The uncertainty introduced by this range of refractive indices for the particle mass is approximately a factor 2. This has been added to the discussion of the OPC results in Section 6.1.

The instrument package collects samples so why not use these to directly determine the refractive indices ?
After discussion with Kandler (personal communication 2010) and following Reviewer #2's comments, we realised that it is very difficult to calculate refractive indices from the measured particle elemental composition as the refractive index depends not only on the composition but also on the detailed crystal structure of the particles and possible particle coatings. We have therefore chosen to use three refractive indices that span the range of expected volcanic ash refractive indices given in the literature.

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.

We agree that this would be a first step towards making a better estimate of the refractive index. However, the time resolution of the particle sampler was 90 min for 20 April and 50 min for the flights in May. This is much larger than the 180 s time resolution of the OPC which prevented a detailed matching of the few elemental composition samples with the single OPC size distribution measurements.

(3) Shape of particle size distribution

As suggested by Reviewer #1, a new figure 3 and a more detailed explanation have been added in the revised manuscript to better explain the extrapolation procedure.

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.
The exponential form of the fit is the simplest way of extending the size distribution without knowledge about the real size distribution. It is further supported by the size distributions found by Schumann et al. (ACP, 2011, their Fig. 7) in Eyjafjallajökull volcanic clouds which show a continuous extension of the size distributions from particles below 1 μm to larger particles. We have added a sentence to Section 2.2 of the revised manuscript to explain this. Using minimum and maximum slope as done in our analysis is a way to span the range of possible slopes and the change of slope for larger particles found in some of the size distributions reported by Schumann et al. (ACP, 2011).

Special comments

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);

We have softened the statement in the abstract and conclusions as well as the sentence at the beginning of the "Discussions" section as also requested by Reviewer #1: "Nevertheless, the CARIBIC aircraft conducted a comprehensive suite of measurements in volcanic ash clouds of variable age during the three flights which add some unique in situ observations to the multitude of measurements collected during the Eyjafjallajökull eruption in 2010."
We still think that CARIBIC can contribute to volcanic cloud research with its comprehensive suite of measurements. We did not claim CARIBIC to be *the best* tool for a rapid response to a new volcanic eruption. We also made it clear that these were special mission flights and not the routine CARIBIC measurement flights which are performed during routine long-distance passenger flights.

2. KNMI TRAJKS back-trajectory analyses used to determine volcanic origin for sample air: It may seem obvious, but the CARIBIC instrument payload measures SO_2, so clearly, when elevated SO_2 was encountered this could be used as an indication of a volcanic cloud encounter.
This is certainly true and we have also done this for the CARIBIC flight on 16 May 2010 as detailed in Heue et al. (ACP, 2011). Unfortunately, as stated in the paper, on 20 April the DOAS instrument did not detect a SO_2 signal, on 16 May when DOAS detected SO_2 the OPC did not function and on 19 May when the OPC detected volcanic ash the DOAS system did not function. So apart from these teething problems that meanwhile have been resolved, this certainly can be done in further CARIBIC measurements of volcanic clouds.

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.
This section gives the transport times from the volcano to the CARIBIC flight path. As such it simply means that for the flight on 20 April 13:47 – 17:33 UTC, the air was 15 – 29 hours earlier over Iceland, for the flight on 16 May 08:08 – 13:49 UTC the air was 34 – 53 hours earlier over Iceland and for the flight on 19 May 07:43 – 15:38 UTC the

air was 19 – 24 hours earlier over Iceland.

Please also clarify the text in this section: what do the 15 trajectories correspond to ? Why was the emission source moved by $\pm 0.4^\circ$ and pressure varied by $\pm 3\%$? What relevance are these amounts ?
The main idea is to calculate a trajectory ensemble and then to assess the spread of the ensemble to get a handle on the uncertainty of the trajectories. The 0.4° in latitude/longitude and 3% in pressure are arbitrary shifts to create the trajectory ensemble. The text has been reworded to explain this more clearly.

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 ?
In our opinion 200 km is a reasonable threshold as trajectories may vary by as much as 1000 km after 1 day of backward calculation when using two different trajectory models. Another source of uncertainty is the meteorological data used: ECMWF or GFS, time resolution of 6 hours or 3 hours, horizontal resolution of 1° or 0.1°. Changing this threshold e.g. to 50 km reduces the whole air samples with "volcano contact" from 11 (200 km threshold) to 4 (50 km threshold). Such a small number of volcano air samples would not agree with the chemical signatures measured in the whole air samples by Baker et al. (GRL, 2011). Therefore we think that the threshold should be larger (we keep 200 km).

Technical comments

The manuscript has been changed to incorporate the Reviewer #2's comments regarding wording and grammar.

P.16695 L.2 The volcano is known only as Eyjafjallajökull - the abbreviated versions mentioned add confusion so please remove.
Although the full Icelandic name is Eyjafjallajökull, the shorter versions have been used in the literature (e.g. Eyjafjalla in the title of the Schumann ACP paper, Eyjafjöll in the

title of an ESA/EUMETSAT Workshop report). So we prefer to keep this disambiguation in the manuscript.

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).
This has been clarified in the revised manuscript.

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.
This were samples from the first two days (14/15 April) where the overlying glacier increased the strength and explosivity of the eruption and created more fine ash particles than during later stages of the eruption. This has been added in the text.

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.
This paragraph discusses the *technical* properties of the OPC which are independent of the real particle composition. The OPC does not discriminate between the particle composition but merely counts the particles in different size bins. However, the main focus of the paper is the particle mass concentration. Since mass scales with the volume of the particles, the smallest particles counted by the OPC do not contribute significantly to the total particle mass (see also Fig. 8 of revised manuscript). Kandler (personal communication, 2011) found a few ash particles as small as 90 nm in samples collected by the DLR Falcon.

P.16707 L.29 There is also the SAVAA resource (www.savaa.nilu.no).
Thank you for this hint. This enumeration is only listing resources used for the CARIBIC flight planning. SAVAA has not been used and is therefore not mentioned.

P.16708 L.15-L.26 Please reword this paragraph more clearly.
We have reworded this paragraph. See also replies above concerning the trajectory ensemble.

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.
This introductory statement does not mention submicron-sized ash particles explicitely. The time-resolution of the CARIBIC particle sampler (90 min for 20 April and 50 min for the other two flights) is not high enough to correlate particle samples with peaks in OPC particle mass concentrations. Because of the inertial impaction, it is also unsure how good the original size distribution is retained in the sample. So we can only determine the average elemental composition of all collected material. The sentence has been changed to:
"The aerosol impactor samples indicated volcanic origins of the probed air masses through marked changes in the elemental composition for all three measurement flights as discussed below."

P.16710 L.5 Contradictory statement here: the DOAS measured an "enhancement" in SO_2 (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.
The DOAS results are discussed in more detail in Heue et al., ACP, 2011. This sentence is meant to complement our findings that the aircraft flew through air with volcanic ash and gases at that time. Since the signal is close to the noise, it is not possible to give a number. We have changed the text to make this point.

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) ?
The numbers are already in the text: background $=$ 1500 particles/cm^3, volcanic enhancement up to 18000 particles/cm^3 for particles larger than 18 nm (black line in Fig. 5

of revised manuscript). We have reworded the text.

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.16718 L.25 Please provide a quantitative comparison, i.e., tabulate the data from the current study and Sigmundsson et al. (2010).
We have added a new Table 2 which lists the elemental composition of all aerosol samples collected during the three flights as well as the elemental ratios discussed in the text. It also lists the corresponding values for the data from Sigmundsson et al. (2010).

P.16711 L.6 How much SO_2 ? Are the authors suggesting that there should be a general correlation between SO_2 and BrO ?
In the paper by Heue et al. (ACP, 2011) we focused on the detailed analysis of the two major SO_2 peaks. Therefore the aerosol properties for the third peak haven't been estimated. If we use the same air mass factor as for previous SO_2 peak the concentration is roughly 10 ppb. However, besides the different AOT also changes in the flight altitude and the flight direction have been neglected here. As this estimate is not precise this number is not in the revised text. Instead, we have changed it to speak of "a much smaller increase in SO_2 and no increase in BrO".

While SO_2 is emitted directly by the volcano, BrO is only produced later in the volcanic plume from precursor species emitted by the volcano. So we do not expect a correlation due to chemistry but due to the fact that both gases are only found inside the volcanic clouds and are transported together.

P.16711 L.23 "...were affected by volcanic emissions" - ambiguous statement.
This has been changed to: "which were traced back to the Eyjafjallajökull".

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.
The word "strongly" has been removed. We stand by our conclusion that the volcanic size distribution (red bars in Fig. 8 of revised manuscript) increases with size for parti-

cles larger than 450 nm while the background size distribution (blue bars) stays more or less constant.

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 ?
This has been reworded to point out that the long sampling time of this aerosol sample spanned a period of air mixed with volcanic emissions (first part of sampling) and a period of pure background air without volcanic emissions (later part of sampling).

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 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". Therefore the $>$20 microns fraction cannot be ignored.
We have changed the text to mention aggregation and particle rafting, inserted a reference to Sorem (1982) and softened the statement.

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".
We have changed this in the entire revised manuscript including the title.

P.16722 L.19 Is there any published evidence that SO_2 causes "enhanced corrosion" of aircraft?
Yes, e.g. Casadevall, Volcanic hazards and aviation safety: Lessons of the past decade, Flight Safety Foundation – Flight Safety Digest, May 1993. This review cites crazing of acrylic windows, corrosion damage to plastic and rubber seals, corrosion of metal components of the airframe as well as engine corrosion due to volcanic gases (mainly sulfuric acid from converted SO_2). We are now mentioning this conversion in

the text.

And Peter W. Purcell reported on the ash@eufar.net mailing list on 18 May 2010 about the status of the ARSF Do228:
"In the course of inspection, it was found that there was discernable signs of fine ash on the engine impellers, and various engine controls and levers were stiff and binding (since freed up with lubrication) - this was attributed to oxidation due to SO_2 exposure. Further signs of oxidation and more severe corrosion were found on undercarriage components, and have been inhibited. ...
The corrosion is of some concern as the instruments indicate that SO_2 exposure totalled on 19 April 240 seconds at between 2 and 5ppb; and on 21 April 40 seconds at greater than 10 ppb with 8 seconds greater than 100 ppb (spiking at 160 ppb)."

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. ... Please discuss the CARIBIC measurements in the context of Rose et al. (2006) and Millard et al. (2006).
We have removed the fumeroles and Antarctic volcano reference and rewritten this paragraph as requested with a reference to Rose et al. (2006). The ozone loss through halogene activation reported by Millard et al. (2006) has been incorporated in the paragraph discussing the (missing) ozone losses.

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.
We have inserted a sentence on the uncertainty of the in situ measurement and mentioning the source as uncertainty for the modelling. The source term in the FLEXPART model run has been constrained by inverse modelling as described in Stohl et al. (ACP,

2011).

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.
This is in both cases due to tropospheric clouds and pollution in the lower troposphere while approaching Frankfurt Airport. We have added a note in the text for the two figures.

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