Atmos. Chem. Phys. Discuss., 10, C9950–C9955, 2010
www.atmos-chem-phys-discuss.net/10/C9950/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Airborne observations of the Eyjafjalla volcano ash cloud over Europe during air space closure in April and May 2010" *by* U. Schumann et al.

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Received and published: 18 November 2010

Response

We are grateful for the thoughtful comments of the reviewer.

The reviewer raised several issues, which we address in this response. We agree to most of them, and this will lead to improvements of the paper.

First of all, let us stress that we do not assume that the particles are non-absorbing. The derived imaginary values of the refractive index (Table 4) clearly shows that the particles, in particular the small ones, are absorbing. However, we analyzed the FSSP

data assuming two sets of refractive index values, one for absorbing (A) and one for non-absorbing material (N), A: 1.54 +0.004 i; and N: 1.54 + 0 i. Case N is used only in order to obtain a lower bound for the particle sizes in the individual FSSP channels and hence for the derived ash masses. In view of Table 4, case A should serve to provide an estimate of the upper bound results. In fact, as noted on page 22155, line 3 of the paper, for the sample of May 2, 40% of the particles had imaginary refractive index values at 630nm wavelength smaller than 0.0005i. Hence, the mean index value may be far smaller than the value 0.004 I used for case A.

From the results we get rather large particle sizes when assuming case A. The maximum particle size (see fig. 7) may reach 50 μm, and sedimentation arguments (section 4.1) tell us that such large particles, though not impossible, should be less likely. Therefore, we assume that the results for analysis N are more realistic than analysis A. The truth is still unknown and is likely located in between the results for cases A and N.

We share the skepticism on the performance of the FSSP instrument. We agree that the fact that the volume spectra tend to show maximum values often in the largest or second to largest size channels is disturbing. The instrument is a rather old instrument and certainly not the best one available for such measurements in principle today. We have better instruments for future measurements, but this instrument was the only one available for quick installation on this aircraft for the measurements after the eruption event on short notice. Since we cannot repeat the measurements, some uncertainty may be left, and that is already noted in the paper, or should be stressed even more.

We are convinced that the measurements make sense because of the following facts: 1) The FSSP signals are clearly correlated with all other signals (SO2, CO, OPC, PCASP) when entering volcanic ash plumes, see e.g., Figs. 12, 14 and 18. 2) As noted in the paper, we found that some of the lower and the largest channels showed electronic noise and these channels were excluded from the analysis therefore. Besides those channels, we do not have electronic noise because outside of clouds or volcanic aerosol plumes, the FSSP did not show any counts (as expected). 3) When

ACPD

10, C9950–C9955, 2010

plotting the mass concentration versus SO2 concentration, which are two independent measurements, the derived mass concentration would show finite values even for zero SO2 when noise induces signal artifacts. But this is not the case. 4) As the reviewer notes, preliminary comparisons with data measured nearly simultaneously onboard the BAe 146 on May 17 (Ben Johnson et al., personal communication) show that our mass-SO2 correlations for case A and N embrace the results for the BAe 146. Details are to be presented together with our British colleagues elsewhere. 5) As noted in section 3.4.1 of our paper, our results for April 19 are consistent with Lidar data of the ground based Lidars in Munich and Leipzig, giving similar ash mass concentrations for this day. The details on these studies are to be reported in separate papers in cooperation with the Lidar teams.

The reviewer several times asked for discussion of optical properties (specific extinction, asymmetry and single scattering albedo). Approximate estimates of the extinction coefficient are given in section 4.3. The asymmetry parameter and the single scattering albedo depend on scattering phase function and absorption efficiency which we could compute assuming certain particle shapes and (size dependent) refractive index values but which we did not measure, and hence are beyond the scope of this paper. Fits to log-normal modes would not be consistent with the cut-off mode discussed in Section 4.1.

The reviewer's comment concerning the effect of charging particle is very interesting, and we will note this possibility in the revised paper. However, we are not convinced that this effect necessarily causes an increase of the particle residence time for larger particles: We can imagine that also the opposite is true. The paper Ulanowski et al. (2008), cited by the reviewer, states: "In principle, aggregation of smaller particles might shift the size distribution towards larger sizes and in our context it may be relevant that aggregation of aerosol particles can be enhanced by charging under some circumstances (Clement et al., 1995)." However Ulanowski et al. (2010) do not discuss the consequences of aggregation. If charging enhances the coagulation rates between

ACPD

10, C9950–C9955, 2010

large and small-sized aerosol (Clement et al., 1995), this could result in shorter residence times because larger particles may be formed that sediment out more quickly.

We do agree to most of the specific comments and will use them to improve the paper. Some comments on critical issues follow:

Page 13: Please note that our ash mass flux refers to fine ash, not total ash. However, we agree, that the uncertainty, which was discussed in sections 3.4.3 and 5, needs to be stated in the abstract also.

Section 3.2, Fig 5b: The correct caption is: Fig. 5. Example of ammonium sulfate particles (left), a silicate particle with droplets of ammonium sulfate (middle) and an agglomerate of large silicate particles with iron oxide (small particles).. . “

Section 3.3. The size distributions shown in the manuscript are a composite of PCASP and FSSP-300 data. In order to calculate total volume etc. we construct the composite so that there is no overlap. Higher channels of the PCASP (corresponding to sizes roughly above 1 μm) are not shown; for the FSSP-300, the lower channels are not included. Furthermore, the channels 1 and 2 of the PCASP instrument cannot be used due to electronic noise which occurs when the probe gets cold. For the PCASP we show the original resolution of the remaining channels. For the FSSP-300 the reviewer is correct. We choose to re-bin the channels into larger size bins for a couple of reasons: 1) to account for Mie ambiguities, 2) to smooth the spectra, because there was considerable channel-to-channel variability, 3) to obtain better counting statistics for larger size bins.

The sampling area of the FSSP-300 is an instrument property which may change over time according to our experience. The sampling area is difficult to determine in the lab. We have always used in the past the method to match the FSSP-300 sampling area so that we obtain the best possible overlap with the PCASP data for particular situations with dry ambient conditions and then assumed this sampling area to be constant for a measurement campaign. We are not aware of potential problems related to a change

ACPD

10, C9950–C9955, 2010

of instrument sensitivity with particle size.

The 40 % uncertainty for mass concentrations relates (only) to the uncertainty in the sampling area. That's why we wrote "resulting". It refers to the previous sentence on the sampling area uncertainty. Other uncertainties (e.g. due to choice of refractive index) are discussed elsewhere in the paper.

Finally we note that the eps-file version of Fig.7 presented in our ACPD-paper missed the micro-symbol in the axis legends. The corrected version is enclosed. As suggested by the reviewer, it now contains better adjusted scales for volume and surface.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 22131, 2010.

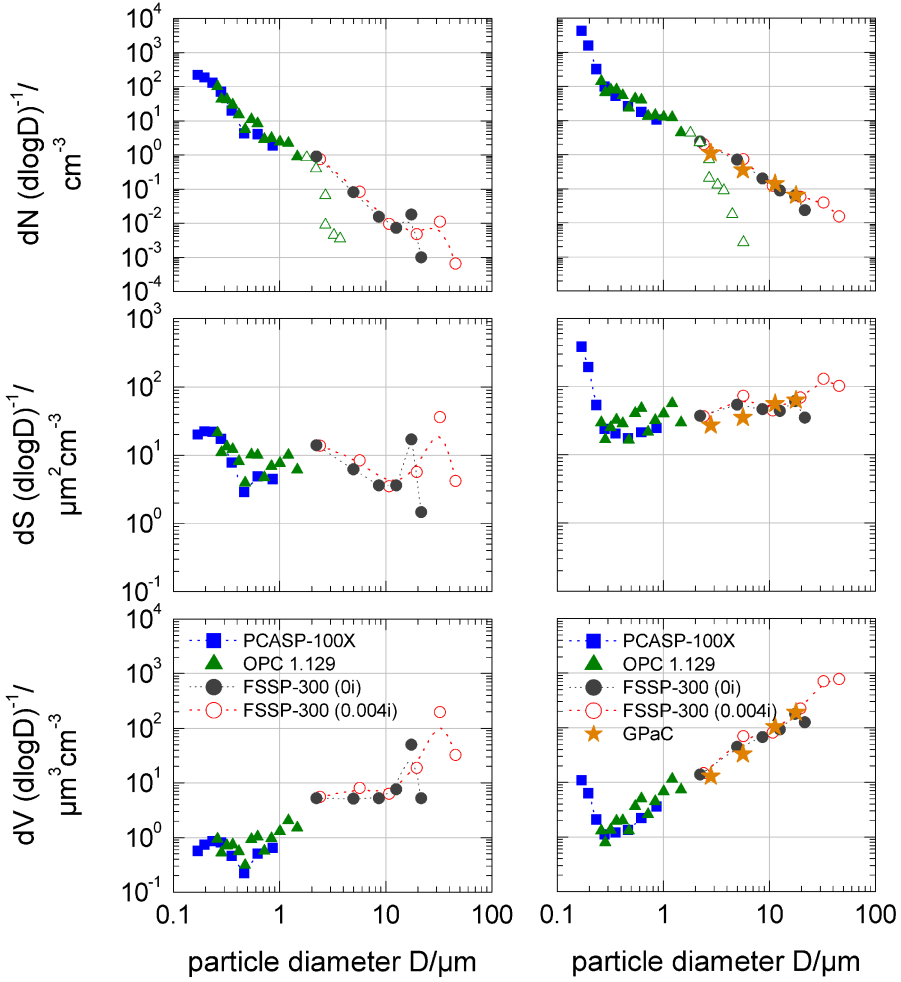

Fig. 1. Fig. 7 revised: Particle number N, surface S, and volume V per unit size interval and unit ambient volume versus particle diameter.

ACPD
10, C9950–C9955, 2010