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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric ice nuclei in the Eyjafjallajökull volcanic ash plume" *by* H. Bingemer et al.

H. Bingemer et al.

bingemer@iau.uni-frankfurt.de

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H. Bingemer et al.

Reply to Referee 1

We thank the referee for his/her valuable comments.

» Referee's Point 1. The methods section is exceptionally short. There are two major topics that need to be expanded: (1) The FRIDGE system is not adequately explained. I see there is a previous set of references but there needs to be a plot showing how this system compares to other previous instruments. Specifically there should be something like a plot of ice nucleation conditions in FRIDGE vs. some previous standard (i.e. a mineral dust or perhaps AgI aerosol) as measured in e.g. the CFDC or AIDA. «

REPLY: As requested, we have expanded the methods section by a paragraph which explains how our method compares to other methods , and to Arizona Test Dust (ATD) used as a standard in the Fourth International Ice Nucleation Workshop ICIS-2007. The comparison of FRIDGE to the continuous flow IN detection system FINCH at our own laboratory has been fully described in Atmospheric Research (Bundke et al., 2008), and we consider a replication of the figure on this issue inappropriate here. FRIDGE is furthermore compared to other independent methods of ICIS-2007 in a manuscript which was submitted to the Bulletin of the American Meteorological Society by DeMott et al. (2010) and is currently under revision. This material is subject to copyright for first publication by the ICIS-2007 community, and we have thus only cited its major statement related to the issue here.

» Referee: Second, a major point of the paper is post-FRIDGE analysis of ice IN. How were these separated from unactivated aerosol and collected? I understand this is a non-trivial step and yet this information is almost completely lacking and it is therefore impossible to judge the validity of these data. «

REPLY: We recognize that our description of post-FRIDGE analysis of IN may have been somewhat short. The FRIDGE/ESEM coupling and identification of IN by ESEM are now explained in detail in a new paragraph, which reads:

" The unambiguous identification of the analyzed particles as ice nuclei was enabled by a high precision laser engraved coordinate system on the substrates. Each substrate has three laser engraved, cross-shaped marks. The marks represent the coordinates 0/0, 100/0 and 0/100 of a Cartesian coordinate system on the substrate. In this way each nucleation event occurring during a FRIDGE measurement (and detectable by the visible ice crystal in the image of the CCD camera) can be documented by means of xy-coordinates, thus representing the position of the individual particle which has acted as ice nucleus. Those marks could easily be recovered in the ESEM, and were there again used to transfer the coordinates to the instrument by using the preinstalled "user units" software option of the electron microscope. This software enables to as-

sociate user-defined points (in this case the cross-shaped marks) with specific points of the positioning system of the specimen stage in the ESEM. Thus the position of ice nuclei could be recovered with an accuracy of approximately 10 μm. If more than one particle is observed in the field of view at the designated location those particles are not analyzed because it is impossible to identify the particle that served as the ice nucleus. Due to the relatively sparse distribution of particles on the substrate this was rarely the case."

» Referee's Point 2. The section on particle composition (page 2738, first paragraph) needs to be expanded. This is perhaps the central topic of the paper but contains little information beyond reference to other papers. IN are broken into four groups: volcanic particles, soot, sea-salt, and biological particles. Mineral dust is incorporated into the volcanic particles, but how are they differentiated? I think the paragraph says they are not and instead a non-volcanic average of mineral dust is subtracted. This 'wedge' should appear on the figure. Second, how are biological particles defined? I thought these were difficult to differentiate from other materials (humics, organics) using EM? Are there no other categories or 'undefined' particles? The pie graph seems very simplistic and clearly delineated and it is hard to believe the data so clearly break down into 4 groups and nothing more«

» Referee's Point 3. Expanding on this topic : I can not reconcile Figures 1 and 3. Looking at Figure 1 it appears that there are _700 IN / liter present during the first 'plume'. The high point of the non-volcanic periods is _100. In this case >600 would need to be of volcanic origin, meaning no more than 14% of IN would be non-volcanic. I note this is in the most extreme case, too, where one considers the HIGHEST non-volcanic abundance as opposed to the average which is an order lower. Yet in Figure 3 fully 35% and 25% of IN are in two cases in the 3 non-volcanic categories. How can this be reconciled? Are volcanoes producing sea salt and biological particles? Should not these categories be well less than 1/7th the total IN? And we haven't even considered the background mineral dust which is normally the most abundant non-volcanic IN type.

More than anything this point needs to be explained before publication. «

REPLY to points 2 and 3 : A detailed explanation of the particle classification (including a new Figure 3) and a discussion of the abundance of these particle groups in the IN were added to chapter 3.

The reviewer argues that, based on the most extreme plume/background ratio of IN, the volcanic category should be around 85% of IN particles. In addition our statements in the revised manuscript some more reasons support the argument for volcanic IN: First, it has to be considered that because of the limited number of activated ice nuclei in FRIDGE, the complex FRIDGE/ESEM coupling technique and the time-intensive manually EDX analysis the total number of analyzed ice nuclei was not very high (in total 250). Therefore the confidence intervals for the individual particle groups (given in Table 1) are rather large. Therefore even in the sample with the lowest volcanic abundance (sample #906, 64.7%) the upper limit of the confidence interval is as high as 80.3% (Table 1). To decrease this uncertainty significantly a much higher number of particles would have to be analyzed.

Second, some of the particles, which were classified as biological, sea salt or soot, may also contain volcanic material, if these particles are complex internally mixed particles.This complex mixing state of the IN is not visible by the classification as "sea salt" or "soot", which is based on the detected main component of the individual particles only.

At some of the volcanic particles an internal mixing with sulfates and/or chlorides was observed. These particles were classified as "volcanic". Mixtures with small contents of volcanic material and dominant sea salt contents are classified as "sea salt". The presence of a silicatic component within individual sea salt particles cannot be verified in the IN samples, as the background signal from the silicon wafer substrates hinders the detection of small silicon contents within the sea-salt particles. In contrast to externally mixed sea salt internal mixtures of sea-salt with other components are reported

to be active as IN (Ebert et al., 2011).

» Referee's Point 4. As the salient reference on ice nucleation by volcanic materials the work of Durant et al. 2008, which is later referenced, correctly deserves a place in the introduction. «

REPLY: This reference was already present in the 4th sentence of the introduction (P. 2734 L. 23), which might have been overlooked by the referee.

» Referee: Furthermore, how do the activation points of Durant et al. compare to the activation points in this paper? This needs to be clearly explained since it is the previous data that most closely compare to what is presented in this work. «

REPLY: There were already two sentences (P2740 L1-5) on this, but we agree that this issue deserves some expansion. Our freezing temperatures indeed compare well to those reported by Durant et al. (2008). The new paragraph on this reads: "The highest temperature at which we processed samples in FRIDGE was -8°C. The growth of ice was observed in each of the samples analyzed at this temperature and at water saturation. We cannot exclude the existence of ice nuclei active at temperatures even warmer than -8°C. At -8°C the number of IN was on average 34% of the number counted at -18% and water saturation. Under the volcanic plume IN were enhanced against the seasonal background of 2009 by approximately a factor of 9 for the IN active at -18°C, and by a factor of 6.5 for -8°C, respectively, suggesting a major volcanic contribution. Such a high freezing temperature compares well to data reported from laboratory measurements of bulk volcanic ash from Kilauea, Hawaii and Cerro Hudson, Chile, by Durant et al. (2008) and from Mount. St. Helens (Schnell et al., 1982). From a synthesis of their own laboratory data and historical data Durant et al. (2008) conclude "that volcanic ash particles initiate freezing in a relatively narrow temperature range of approximately 250 K to 260 K (or perhaps up to 265 K)". However, in comparing these activation conditions to ours one has to bear in mind that some of these previous data (e.g. from Durant et al. (2008) and Schnell et al. (1982)) were obtained

for the immersion and contact freezing modes, whereas our measurements address the deposition and condensation-freezing modes."

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