

Reply to interactive comment on „Ice nucleation properties of fine ash particles from the Eyjafjallajökull eruption in April 2010“ by Referee #2

First, we would like to thank referee #2 for thoroughly evaluating our manuscript and giving constructive feedback.

Major comment 1:

Larger ash particles sediment near the source region from which the sample was taken. Smaller ones are transported over longer distances and may have a larger impact on cloud microphysics. Therefore a discussion dealing with the atmospheric relevance of the investigated volcanic ash particles is necessary since the chemical and physical properties of the emitted volcanic ash particles, and hence their IN ability, may vary over the original size distribution.

Reply to major comment 1:

Measurements of the volcanic ash particle size distribution find that the fraction of particles smaller than 2 μm is enhanced in the initial volcanic plume, bulk samples collected in the vicinity of the volcano and in air masses transported towards Central Europe (Schumann et al. (2011), Gislason et al. (2011), Schleicher et al., (2011)). For phase II of the eruption it is assumed that aggregation in the water-rich plume enhanced the sedimentation of the small particles (Ilyinskaya et al., (2011)). During our experiments the aerosol is dispersed with a rotating brush generator which causes the particle agglomerates to (at least partially) break up, setting free the smaller particles. At the same time particles larger than a few μm are eliminated from the aerosol population by an impactor stage. Thus, with the volcanic ash sample used in our experiments the atmospherically relevant particles smaller than 2 μm can be investigated. We have added a note on this aspect to our manuscript (see p.10).

Major comment 2:

The derived parameterizations are based on the concept of ice-active surface site density, i.e., the singular approach. Following the long term discussion whether the heterogeneous ice nucleation process has a stochastic or singular nature a discussion concerning the validity of the approach applied would be desirable.

Reply to major comment 2:

The ice-active surface site density approach is used for describing the ice nucleation properties of our volcanic ash sample even though it does not capture the stochastic nature of the single ice nucleation events. However, this approach appears to be a reasonably good representation for the ice nucleation efficiencies observed in our experiments since it avoids introduction of a complex set of parameters which would be needed for a thorough description of the polydisperse aerosol population by a distribution of contact angles (Marcolli et al. (2007)). There are also stronger indications that ice crystal concentrations observed in AIDA experiments depend on temperature (and RH_{ice} for deposition nucleation) rather than on time (e.g., Möhler et al., 2006). This has been clarified in the text (see section 3.3).

Minor comment 1:

For completeness of atmospheric volcanic ash observations in connection with the eruption of the Eyjafjallajökull volcano in 2010, please cite the LIDAR observations performed by several European groups (e.g., Ansmann et al. (2010) and Mona et al. (2011)). (17667/20-24)

Reply to minor comment 1:

We agree that lidars can provide valuable information about the dispersion and composition of volcanic ash layers in the atmosphere. Accordingly, in our introduction we added “lidars” to the list of instruments which were used in order to monitor the dispersion of the Eyjafjallajökull ash cloud. We also added a reference to the study by Seifert et al. (JGR, 2011).

Minor comment 2:

Immersion freezing requires an insoluble or undissolved aerosol particle. (17668/14-15)

Reply to minor comment 2:

We changed “aerosol particle” to “undissolved aerosol particle”.

Minor comment 3:

The differentiation between droplets and ice crystals with the WELAS instrument is done by particle size whereas ice crystals are larger than droplets. Is there an overlap region between droplets and ice crystals where the droplet signal can not really be separated from the ice crystal signal? How large are the measurement uncertainties when deriving ice fractions? According to the size distribution measurements, you also have very large unprocessed aerosol particles inside the AIDA chamber, is it possible that these particles interfere with the droplet and ice measurements?

Reply to minor comment 3:

There is definitely an overlap region between droplets (resp. large aerosol particles) and freshly nucleated small ice crystals. However, ice crystals grow rapidly to sizes much larger than the initial droplets/large aerosol particles and the assumed size threshold. For immersion freezing, the resulting ice crystals quickly grow via the Bergeron-Findeisen process, whereas for deposition nucleation ice crystal number densities are much larger than the number concentration of the large aerosol particles. Thus, in particular for deposition nucleation large aerosol particles contribute to the measurement uncertainty, but only to a minor amount. Therefore, it is assumed that ice crystal number densities in immersion freezing mode can be determined with an uncertainty of $\Delta n \approx 20\%$, whereas for deposition nucleation we find $\Delta n \approx 25\%$.

Minor comment 4:

Why did you choose different particle diameter thresholds to distinguish between aerosol particles/droplets and ice crystals for the immersion freezing and deposition nucleation experiments? Please add an explanation. (17673/8-10)

Reply to minor comment 4:

The most suitable threshold is found for each experiment individually since variations in the aerosol size distribution need to be taken into consideration. For immersion freezing also the droplet size distribution is influenced by these aerosol size distribution variations. Therefore, the thresholds might vary slightly between experiments. Also, in deposition nucleation mode ice crystals typically do not grow to as large sizes as for immersion freezing. Thus, the threshold is sometimes set at lower values in order not to underestimate the small ice crystals. We

modified the sentence about the deposition nucleation to “For deposition nucleation, this ice crystal threshold is more difficult to find and set to a slightly smaller value $d = 15 \mu\text{m}$ in order not to underestimate the number density of small ice crystals.” (see p.8)

Minor comment 5:

What measure has been used for comparing the surface properties of the ash particles investigated in this study to those observed in the atmosphere? (17677/1-4)

Reply to minor comment 5:

For our analysis we used ESEM images of the volcanic ash particles: similar to the particles observed by Schumann et al. (ACPD, 2011) we also saw shard-like, silicate-rich particles with sharp edges. However, we only investigated a small number of particles with this method.

Minor comment 6:

What is the physical concept underlying this statement? Please add something like: "... assuming that ice fraction is related to the available ice nucleus surface area ..." (17678/25-27)

Reply to minor comment 6:

We have added a sentence addressing this issue: “These large particles can be assumed to have increased freezing probabilities at higher temperatures due to the larger available ice nucleus surface.” (see p.14)

Minor comment 7:

Why is the ice crystal number density used? According to DeMott (1995), Connolly et al. (2009) and Niedermeier et al. (2010) the ice active surface site density has the dimension $\# \text{m}^{-2}$. Or is A the total aerosol surface density? Please clarify the units. (17679/14)

Reply to minor comment 7:

The ice crystal number density n is given in $[\text{cm}^{-3}]$ and the aerosol surface density A is given in $[\mu\text{m}^2 \text{cm}^{-3}]$. This has now been also clarified in the manuscript.

Technical comment 1:

Delete “However,” (17666/3)

Reply to technical comment 1:

We deleted “however” as suggested.

Technical comment 2:

Fig. 5 instead of Fig. 4, and similar reference mismatches (17677/22 and else)

Reply to technical comment 2:

The references are now correct.

Technical comment 3:

To illustrate the uncertainty of the relative humidity with respect to water and ice of the measurement the uncertainty range could be shaded in Fig. 5. So that shaded area of the immersion freezing results would lie on the water saturation line. (Fig. 5)

Reply to technical comment 3:

We have now added a grey shaded area which illustrates the uncertainty in relative humidity over water and over ice.

Technical comment 4:

Delete the brackets. (17684/7)

Reply to technical comment 4:

The brackets have been deleted.

Technical comment 5:

Would “aerosol surface area density” be more appropriate than “aerosol surface area”?
(17690/1)

Reply to technical comment 5:

In table 1 “aerosol surface area” has been changed to “aerosol surface area density”.

References

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