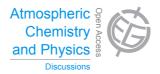
Atmos. Chem. Phys. Discuss., 15, C185–C187, 2015 www.atmos-chem-phys-discuss.net/15/C185/2015/

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15, C185-C187, 2015

Interactive Comment

Interactive comment on "Deposition and immersion mode nucleation of ice by three distinct samples of volcanic ash using Raman spectroscopy" by G. P. Schill et al.

Anonymous Referee #3

Received and published: 12 February 2015

The present work extends the knowledge on depositional and immersion mode freezing of ice on volcanic ash samples. Three selected ash samples have been chosen to study IN formation by Raman micro-spectroscopy. These samples cover the range of basaltic, andesitic and rhyolitic ash. While all three samples exhibit a similar behaviour during depositional freezing, the authors claim varying behaviour during immersion mode freezing, which might be related to differences in the mineralogy of crystalline material. In contrast to previous studies, their results indicate, that a 'simple' parameterisation of the immersion mode freezing for volcanic ashes is not legal.

After a brief and straightforward introduction, a detailed experimental section is follow-

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ing. The results of their study are presented according to IN properties in the depositional and immersion mode. The paper concludes with a discussion on atmospheric implications. The paper is well written and could be interesting to the readership of Atmospheric Chemistry and Physics. However, the paper lacks of experimental details and interpretation of data obtained from Raman spectroscopy. Further, the significance of the results has to be discussed related to the limited dataset. Therefore, publication in ACP can only be considered after addressing the following major remarks in detail.

Major issues:

- *) The title of the paper includes "... using Raman spectroscopy" however, only Raman spectra related to depositional freezing are shown in figure 1. The authors claim, that their study is the first immersion mode freezing study, therefore it is absolutely important to discuss and display details of Raman spectra obtained during the immersion mode experiments in detail. Even experimental details have to be discussed like influence of laser power on heating of the droplets and disturbing the measurements and results. Further, for confocal Raman microscopy, height of the z-resolution must be discussed related to the sample size and height. How reliable are results from a single spot measurement of 20 to 70 μ m droplets (see fig. 3)? A discussion about the legitimacy of Raman micro-spectroscopy for immersion mode measurements is absolutely necessary. For a reader, the experimental work appears in that way, that only the optical part of the microscope was used for obtaining the dataset, but not the Raman spectrometer.
- *) The authors claim that up to now only two datasets have been used to suggest one characteristic ice nucleation efficiency. They extended the dataset from two to five ash samples. There is no statistical confidence within this interpretation. How can the authors reliable conclude that immersion mode freezing is dependent on the composition of the ash by this limited dataset?
- *) How was sampling of the volcanic ashes done? Were the samples collected from

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15, C185–C187, 2015

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the nearby ash fall around the volcanos? How long were the ashes exposed to environmental conditions after the eruptions of the related volcanos? Are particles which settle close to the volcano comparable with fly-ash which is able to cover a distance of several hundreds of kilometres? Is there a general independence of the chemical composition, environmental aging from the size and the difference in exposure (atmosphere, soil, ...)?

General minor issues:

- *) Raman spectra in figure 1 (and others): These Raman spectra are displayed like they were measured in the anti-Stokes mode. Are these anti-Stokes spectra or are the spectra displayed in an unusual way for Stokes-Raman spectra. Further, assignment of the Raman excitations would improve the quality of the figure and assist a detailed understanding of the work.
- *) Sampling: Indication of the geo-coordinates of the volcanos and the sampling site as well as the distance from the sampling site to the craters would be helpful.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 1385, 2015.

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