

**Answer to the Comments of the anonymous referee#2 on the manuscript “Experimental quantification of contact freezing in an electrodynamic balance” by N. Hoffmann, A. Kiselev, D. Rzesanke, D. Duft, and T. Leisner**

We highly appreciate the referee’s comments and suggestion. Below we answer the referee comments and questions (our answers are marked with a bold **A**), along with the comments themselves (marked with bold **Q** and *italics*).

**Anonymous Referee #2**

*Received and published: 3 June 2013*

*This paper discusses the experimental quantification of contact freezing using an electrodynamic balance. Contact freezing is one of the 4 heterogeneous freezing pathways in mixed-phase clouds. Contact freezing is the most challenging freezing pathway experimentally because it depends on the collision rate that needs to be determined as well. In this paper the authors study contact freezing by charging the droplet and the aerosol particle. They first compare collection efficiencies and then discuss contact freezing. The paper is well written, the method is sound and the results are novel. Thus I recommend this paper for publication after the following comments have been taken into account:*

**Major comments:**

**Q1:** *I’m missing that the authors compare their results to earlier results of contact freezing. This is crucial to put your results into perspective and to compare different methods of studying contact freezing. Please add that.*

**A1:** The main objective of this article is to present a novel method of measuring the contact freezing probability. We have shown that the only possibility to characterize the probability of freezing on a single collision event is to treat it separately from the rate of particle – droplet collisions. As it has been recently shown in the review article of (Ladino et al., 2013) it was rarely done in the previous studies, where either the rate of the collisions, or size of the ice nucleating particles were unknown (although the approach was attempted by Svensson et al., 2009). It immediately follows from our study that contact freezing experiments cannot be compared if only fraction of frozen droplets is reported or the value of onset temperature is given (see also the answer to the next comment). The only work using a similar approach that we are aware of (Bunker et al., 2012) reports values of contact freezing probability measured at much higher temperature, so that the comparison is not possible. Given the technical nature of this contribution, the recently appeared review article on contact nucleation (Ladino et al., 2013), and our own paper addressing the issue of size dependency of contact nucleation probability (Hoffmann et al., 2013), we refrain from engaging into the comprehensive discussion of comparison issues; however, we do include the reference to the recent review article in the revised manuscript.

**Q2:** *p.3422: Why do you only investigate a rather narrow temperature range between 237.9K and 240.3K? I recommend extending that so that the onset freezing temperatures can be seen as well.*

**A2:** The limitation is of a technical nature: as it can be seen from our Figure 8, the probability of contact freezing at temperature above -33°C for kaolinite is approaching 1% and decreases almost exponentially with increasing temperature, meaning that we either need to investigate a much higher number of droplets or very high collision rates to achieve statistically significant number of freezing events per temperature set point within the time frame of a single-droplet experiment (30s). Increasing the number of droplets is very time consuming, the collision rate is limited by the aerosol particle number concentration (typically on the order of  $100 \text{ cm}^{-3}$ ). The contact freezing probability cannot be related unambiguously to the onset temperature (defined as a supercooling temperature at which some fraction of droplets – typically 1% – are frozen), because the fraction of frozen droplets is also a function of collision rate. For this reason the onset temperature cannot be used for comparison of contact freezing experiments unless the collision rate is known (see our answer to the previous comment).

**Minor comments:**

**Q3:** *p.3413, line 27: PSL is used here for the first time but explained further back, please change that.*

**A3:** We agree. In the revised manuscript the abbreviation PSL is explained on the first appearance.

**Q4:** *p. 3415/3421: How is the introduction of PSL spheres biasing your collection efficiency results? Couldn't you obtain collection efficiency results without adding PSL spheres given that they cause problems for the PSL experiments? And if so, how do the results differ?*

**A4:** We agree that this point was not clearly described. Only liquid water droplets were actually used for counting the particles collected by the liquid droplet in this study, so that no "collecting droplet" and therefore no additional PSL particles for marking the limits of the droplet footprint after evaporation were employed. We have added this sentence to the manuscript on the page 3421. We argue that the discrepancy between the expected and measured value of scavenging efficiency for the PSL particles was caused by the strong agglomeration of the PSL particles during the droplet evaporation.

**Q4:** *p.3423, line 1: Reference missing.*

**A4:** Corrected, see answer A3 to the anonymous referee #1.

## Our references

Hoffmann, N., Duft, D., Kiselev, A., and Leisner, T.: Contact freezing efficiency of mineral dust aerosols studied in an electrodynamic balance: Quantitative size and temperature dependence for illite particles, *Faraday Discuss.*, Accepted manuscript, DOI: 10.1039/C3FD00033H, 2013.

Ladino, L. A., Stetzer, O., and Lohmann, U.: Contact freezing: a review, *Atmos. Chem. Phys. Discuss.*, 13, 7811-7869, doi:10.5194/acpd-13-7811-2013, 2013.

Svensson, E. A., Delval, C., von Hessberg, P., Johnson, M. S., and Pettersson, J. B. C.: Freezing of water droplets colliding with kaolinite particles, *Atmos. Chem. Phys.*, 9, 4295–4300, doi:10.5194/acp-9-4295-2009, 2009.

Bunker, K. W., China, S., Mazzoleni, C., Kostinski, A., and Cantrell, W.: Measurements of ice nucleation by mineral dusts in the contact mode, *Atmos. Chem. Phys. Discuss.*, 12, 20291–10 20309, doi:10.5194/acpd-12-20291-2012, 2012.