Answer to the Comments of the anonymous referee#1 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 #1:

A thorough, concise, and timely manuscript on the topic of contact freezing. Often suggested as one of the dominant (i.e., highest T) freezing modes, the data on contact freezing are spotty. Hoffman et al. have laid out novel experiments with an EDB setup to tackle this problem. This reads as an instrumental paper (appropriate for AMTD) but also contains a quality scientific study. There are only minor issues with this paper and I expect it to be published quickly.

Q1: Presumably a droplet (*RH*=100% wrt liquid water) is far from equilibrium with any ice that could be on the trap walls. Are drops stable? How are they maintained? If they are not (if they are evaporating) would there not be phoretic effects?

A1: Indeed, the droplet is not in the equilibrium state and is slowly evaporating. There is no ice on the walls of the EDB since dry nitrogen (dew point temperature of T_{dp} -40°C) is used as the carrier gas. Within the relevant temperature range (-30°C to -36°C) and the time scale of the typical experiments (30 s) the droplets are almost stable. The reduction of diameter was found to be at most 5%, and the cooling of the droplet due to evaporation does not change the droplet temperature by more than 0.2K. To estimate the possible role of the phoretic effect, we have calculated the thermophoretic and diffusiophoretic forces acting on an aerosol particle as a function of particle droplet separation distance under typical experimental conditions (upper panels, gas temperature -33°C, droplet is colder by 0.5K, T_{dp} = -40°C). The results are shown in the figure below, together with the Coulomb and induced dipole attraction forces (lower two panels). It is clearly seen, that at separation distance the phoretic forces are one to three orders of magnitude lower than the electrostatic forces. The corresponding difference in the scavenging efficiency with and without account for the phoretic effect was found to be below 0.5%. However, the evaporation cooling has a very pronounced effect on the droplet temperature for the higher experiment temperatures and has to be taken into account.



Figure: Forces acting on a single charged aerosol particle near the charged water droplet as a function of droplet center – particle separation distance normalized to the droplet radius.

Q2: Can the authors address the question of how charge may affect ice nucleation? Are there any expected differences from when a drop and aerosol with no charge comes in contact?

A2: We have no evidence for a possible direct effect of the charge on the freezing efficiency of aerosol particles. Recently we have addressed the possible effect of the charge on the homogeneous freezing of supercooled water droplet (Rzesanke et al. PCCP 2012) and found no effect. The absence of charge on the aerosol particle would decrease the scavenging efficiency but not the contact freezing probability. We add a short discussion of the charge relevant issues into the manuscript.

Q3: *I* believe a reference is missing at 3423-1 "with results reported by () for illite" where the () should contain the reference.

A3: The reference is indeed missing. We have added a reference to the work of (Hoffmann et al., 2013).

Q4: I recommend that section 6 be removed. It is only two short paragraphs that essentially say what the authors are going to do. Either attach this to the conclusions without a new section title or remove it altogether.

A4: We agree with the referee. The Outlook has been added to the end of the Conclusion section.

Q5: This paper will have to be edited for English. There are several grammatical errors throughout; e.g. (but not restricted to!) 3408-6: "allowing to determine" (allowing for the determination), 3424-8: "is currently performed" (is currently being performed), etc.

A5: We have carefully checked the manuscript for English grammar and removed the most obvious errors.

Our references

Rzesanke, D., Nadolny, J., Duft, D., Muller, R., Kiselev, A., and Leisner, T.: On the role of surface charges for homogeneous freezing of supercooled water microdroplets, Phys. Chem. Chem. 10 Phys., 14, 9359–9363, 2012.

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.