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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Investigations of the impact of natural dust aerosol on cold cloud formation" *by* K. A. Koehler et al.

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We would like to thank Referee #2 for the generally positive response to our work. Below we address each specific comment individually.

Response to Specific Comments (original comment above the response): ...I recommend publication of this work once the below given comments will have been addressed. Thank you for the positive response to this work.

1) page 19349, lines 9-10: Later in the paper you discuss results for particles generated after submersion in water (see page 19361 and Fig. 5). So you might not want to explicitly exclude them, here.

Yes, the reviewer is correct. We have changed this sentence to say, "Therefore, in this

study, we have primarily presented results of the ice nucleation behavior of particles generated from bulk powders without submersion in water, using a fluidized bed."

2) page 19353, line 26: "(Delta-RHi=1.5% ...)" - between 200 to 400nm?

Yes, this is correct. We have changed the sentence to explicitly state this now, "Homogeneous nucleation is only a weak function of size and thus the homogeneous freezing representations for 200-400 nm particles would fall nearly on top of one another for the RHi scale on these figures (ΔRHi = 1.5% between 200 and 400 nm particles at T = -60°C)."

3) page 19356, line 2: In connection to the size dependence, please also stress that due to doubly charged particles, it could be the case that those particles that activated ice were largely the larger, doubly charged ones (you say earlier, that about 30% of the particle might be doubly charged). It could be the case that you always observed freezing of the larger doubly charged particles, only, and this should be discussed somewhere.

This is certainly true. In this section we changed this sentence to read, "As for ATD, ice nucleation conditions for dry-generated CID show very little dependence on RHi for T<-40°C and, despite the broader size cuts (and larger influence of multiply-charged, larger particles), a strong size dependence was observed." We also remind readers in Section 2.2 that, "Thus low activation fractions may result if only the largest, multiply charged particles in the aerosol stream activate."

4) page 19359, lines 6-9: "It also appears ..." - I guess you refer to the fact that you only got one datapoint for each of the two smaller sizes, at about -35_C, while you got more datapoints for the 400nm? If so, then maybe stress here that for T>-35_C and for 200 and 300nm particle sizes freezing was not observed (as otherwise one could think that you just did not measure in this range.)

The reviewer is correct, the lack of points for the smaller sizes is because freezing was

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not observed, not because measurements were not completed for these temperatures. We state this explicitly now, “It also appears that the larger particles have ice nucleation activity via condensation/immersion freezing at warmer temperatures than do the smaller particles (no freezing was observed for particles less than 340 nm at temperatures warmer than -35 °C), a trend also suggested for ice nucleation by uncoated dust particles during the ICIS campaign.”

5) page 19360, line 21: Isn’t SD shown as blue circles (not green)?

Thank you for catching this error, it has been corrected.

6) page 19360, line 24: Either also give the type of symbol used for coated ATD, or delete these mentions for the other dust-types, too (as they are given in the caption again).

We have added that this type is denoted by black triangles.

7) page 19360, line 25: Isn’t OLD green squares (not circles)? (This also seems to be wrong in the caption of Fig. 5.)

Thank you, we have fixed this in both places.

8) page 19361, paragraph starting at line 15: I find this whole paragraph very confusing: 1) The sudden appearance and discussion of wet generated aerosol seems to be somewhat out of place, in this paragraph (at its beginning and at its end), particularly as you said in Chapter 2.2 that you would not treat wet generated particles.

Since several studies in the literature have examined the ice nucleation behavior of atomized dust samples (e.g. Archuleta et al.), we felt it was important to discuss the large difference in the behavior or wet- and dry- generated dust. Size resolved ice nucleation behavior of wet-generated ATD and CID as a function of temperature are available in Koehler et al. (2008), however, since this data is not likely to be relevant to the atmosphere, we did not feel it was a worthwhile to present all of this data. Instead, we show for one size the change in behavior in Figure 5.

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8-2) Also no tests were run with CID in aqueous suspension, however, you only say that explicitly for SD.

The filled red circles are for the CID in aqueous suspension.

8-3) There is a break in the topic of the sentences ending and starting in line 19 that is confusing.

We do not feel this is a break in topic. In the beginning of the paragraph we say that we are going to explain the difference in behavior for wet- and dry- generated aerosol. At line 19 we start to describe the behavior of the dry-generated particles. The next paragraph describes the behavior of the wet-generated particles in contrast to the dry-generated particles.

8-4) The size dependence of dry generated particle types is not shown in Fig. 5. And also, this is very hard to see from data for 200, 300, and 400nm in general.

The size dependence in not repeated in Figure 5, as it would be simply too much data for one figure. However, the size dependence at temperatures colder than -36°C is very clear for the CID in Figure 2 and SD in Figure 3, although somewhat less so for ATD in Figure 1. At warmer temperatures, we agree that the size dependence is most apparent for the ATD. We have changed this discussion to make this point more clear.

8-5) There is again a break in the topic of the sentences ending and starting in line 25.

We feel this is continuing the same discussion of the nucleation behavior of the dry-generated dust, but agree it is repetitive and we have removed the sentence. We also moved the discussion of the behavior at temperatures warmer than -20°C to the end of the previous paragraph for clarity.

8-6) I'm don't agree that the RHi required for onset of ice nucleation is nearly invariant (it varies by at least as much as 25%.

The reviewer is correct, invariant is not the correct word, as it does vary, it is simply not

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an obvious function of temperature.

The rewritten version of that paragraph and the next is below:

Figure 5 highlights the clear distinction between the behaviors of dry-generated particles and those generated from atomization of aqueous suspensions (so called "wet-generated", filled symbols, from Koehler (2008)) at temperatures colder than -40°C. Due to the small sample size of the SD available for these studies, no tests were run with SD in aqueous suspension. At temperatures colder than -20°C, the dry-generated particle types generally showed a size dependence. For example, the CID showed distinct RHi values for ice nucleation as a function of size for temperatures below -36°C. This result is consistent with results found by Archuleta et al. (2005) for an Asian desert dust sample. At warmer temperatures this point is most apparent for the ATD. While the particles smaller than 400 nm lost ice the ability to nucleate ice rapidly for increasing temperature, the polydisperse samples containing larger aerosol exhibited ice nucleation near water saturation. A similar behavior is noted for the SD. For all dry-generated dust types, the freezing mode transitioned from deposition to condensation freezing at $\sim$ -36°C. Wet-generated CID and OLD and coated dust particles, however, generally initiated ice formation at conditions similar to those required for homogeneous freezing of sulfates at temperatures colder than -36°C. We expect that this indicates that when the particles contain sufficient soluble material to cover the particle surface, active sites are prevented from serving as deposition nucleation sites. Nucleation was observed at temperatures warmer than $\sim$-36°C, above the limit of homogeneous freezing, indicating that condensation freezing may still proceed, but may depend on the insoluble and soluble material (Möhler et al., 2008). It appears that soluble material, either present as a component of the dust, or deposited on the dust surface through gas phase reactions, acts to inhibit the active sites on the dust, so either the solution coating freezes homogeneously or the dust initiates freezing through immersion or condensation freezing at much higher RHi than was observed for the uncoated ATD. This higher activation RHi in some cases is indistinguishable from con-

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ditions for homogeneous freezing of the coatings. In contrast, the wet-generated ATD exhibited similar behavior to the other dry-generated dusts, although a higher RHi was required for freezing of the wet-generated ATD than when dry-generated for temperatures colder than -36°C. This may be due to the very small soluble content of this dust type and the impact of its redistribution following atomization.

9) page 19362, lines 10-12: Sullivan et al. (2010, still an ACPD-paper) found that for ATD particles coated with sulphuric acid the IN fraction in the deposition freezing regime was greatly reduced (compared to untreated particles), while the IN fraction was reduced to a lesser degree for the condensation freezing regime. Would this mean that coating with SOA and coating with sulphuric acid can do comparable things to an IN? It could be worthwhile discussing this in your work.

While these two ideas are possibly related, we do not feel that we can necessarily make this claim based on our data. In our study, we focused on the conditions for which activation occurs, trying to determine the conditions for which the first particles initiate the ice phase. We did not examine the total fraction of particles able to initiate the ice phase for either case. In Section 3.4 we did compare our results to results by other researchers with other coatings, but we don't feel it is appropriate to claim that SOA and sulfuric acid coatings impact the ability of mineral dust particles to serve as IN in the same way at this time.

10) page 19366, line 15: I think you mean "ice formation on 1% of the particles at temperatures warmer than -25°C was only observed ..." (i.e. move the "was only observed")

We have changed this for clarity.

11) Figures 1-5: Datapoints in the region that is shaded in the lighter grey differ from other datapoints of the same type (i.e. you made the interior somewhat lighter). However, I found this not explained anywhere. You might want to think about excluding these points, anyway. Or you have to explain at least that and why they are somewhat

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lighter .

This was simply an artifact of the plotting program. We have fixed the figures so the points do not look different, as they are not intended to be different.

12) Also, black and the blue that you used are VERY hard to distinguish, so I suggest you use a lighter blue or cyan, instead.

We have changed this to a lighter blue on all figures.

13) Figure captions 1-5: In captions of Fig. 1-3 and in the main text you quote ICIS-data as being for ice nucleation on 1% of polydisperse particles, while in caption to Fig. 5 you quote them as 0.1%. Please correct.

In Figure 5 it should read 1%, this has been corrected.

14) Also I suggest you only explain the general layout of the figure once (in the text or in the caption of Fig. 1) and then refer to this. (E.g. explaining the grey areas, the Koop-line,...) Also, for all captions, the data is NOT shown as a function of sizes, but as a function of temperature and for different sizes.

This information is before all the figures in the beginning of section 3 because it applies to all the figures and is not repeated in each section for brevity. We have changed the captions to read, “as a function of temperature for sizes:. . .”

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 19343, 2010.