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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Saharan dust and ice nuclei over Central Europe" *by* H. Klein et al.

H. Klein et al.

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We thank Gabor Vali for his valuable comments. Each comment and our reply is listed below. Our proposed changes are marked in blue in the revised manuscript.

Comment:

The paper adds to the increasing but still incomplete knowledge about the role of mineral dust as atmospheric ice nuclei. In this study, the case is being made by looking at an event of Saharan dust incursion. In common with the major part of the existing literature on the subject, the focus is on ice nucleation at temperatures at and colder than -18_C.

Listed below are a number of small and some more substantial questions.

Most importantly, while it does sound plausible, the claim that the evidence presented shows dust as a 'dominant constituent' of ice nuclei isn't fully justified. In my view, the assumptions made in constructing the proof (see last comment below) need to be stated, or evidence given for their validity. Also, the findings apply only to specific temperature/superstauration regimes and that too limits their generality.

Reply:

The reviewer makes a very important point (see also the review by S. Brooks). We agree that our interpretation is not fully proven by the evidence presented in the manuscript, in that sense that it is the sole possible explanation of our data. We now try to state clearly 1.) the assumptions made, 2.) that our interpretation is one of several possible conclusions, and 3.) that the assumptions are yet unproven. As a consequence we have weakened our statement on the overall dominance of mineral dust in the abstract and in chapter 3.2. But we still keep the statement that ice nuclei at our site in central Europe are made up to a large extent by mineral dust. Our new statement now reads: "If one assumes that the different individual classes of materials (dust, soot, biogenic, ..) that may contribute to the IN throughout the year exhibit appreciably different slopes in their activation spectra in FRIDGE, then the massive presence of dust during the event of June 2008 could be expected to lead to spectra (Fig.10a) that were different from those at the other times (Fig. 10b) and would thus result in a slope of the linear regression line between both data sets (Figure 10c) that deviates from the 1:1 line. From the fact that our regression line lies within the 99.9% confidence bands of the 1:1 line one may conclude that the assumption above is invalid and that the contribution of the different nucleating materials results in average spectra of the same slope as during the massive dust event, or that the properties and composition of the nucleating materials in both data sets were quite similar, and were likely made up to a large extent by mineral dust. Although the validity of our initial assumption cannot be proven because of unknown composition and properties of the nucleating material, we consider the latter explanation more likely. It is backed by observational data (Klein, 2010) showing that the abundance of IN at the Taunus Observatory throughout

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2.5 years of daily measurement is highly correlated with local dust-specific parameters (e.g. extinction by large particles) derived from AERONET and satellite measurements. ”

The additional evidence for the initial statement is contained in the PhD thesis of Holger Klein (Oct. 2010, University of Frankfurt) and will be part of a follow-up paper on our whole 2.5 years data set. In this work we'll show that the abundance of IN throughout the year is (both for the individual data and for their monthly means) highly correlated with local dust-specific parameters (e.g. extinction by large particles) derived from remote sensing (AERONET and satellite). In addition we have some first indications (not presented in the manuscript) that our assumption on different slopes of spectra may be not completely wrong, as the activation spectrum of Snomax® in FRIDGE is indeed very different from the spectra that we record from ambient samples (see the Figure attached to our response to reviewer S. Brooks)

Specific comments (reference to page/line):

Comment:

14994/25: As Referee #1 also mentioned, ”good ice nuclei' a poor choice of words. In addition, it is also non-specific, as quality could refer to the temperature of activation, the abundance, the time lag, the probability of nucleation, per unit surface area at a given temperature, or other definitions. Most important is to distinguish between activity in large numbers and activity at lesser supercooling.

Reply:

We have replaced “good” by “efficient”, as suggested by reviewer 1.

Comment:

4995/5-6: Did Sassen (2002) really mean altucumulus evolving into cirrus? Seems unusual.

Reply:

Sassen (2002) states: " . . .this cloud resembled cirrus fibratus when overhead, but was earlier classified as a deep, water-dominated altocumulus as it approached ... In other words, this midlevel cloud glaciated with time to produce a pure ice cloud at relatively warm temperatures."

We have now replaced "cirrus" by "ice cloud".

Comment:

14995/14: What are 'cloud forming trajectories'?

Reply:

We changed the two sentences on the Wiacek et al.paper to: "Recently Wiacek et al. (2010) explored the availability of mineral dust particles as ice nuclei for interactions with clouds by analyzing the thermodynamic conditions (temperature and humidity) for the formation of liquid, mixed-phase and ice clouds along 1.8 million trajectories from the major African and Asian dust source areas over a whole year. Of those trajectories that encountered conditions for cloud formation by far the largest fraction entered conditions of mixed-phase clouds, where mineral dust can initiate glaciation."

We have also added the following sentence and citation of a new paper: "Space observations show (Choi et al., 2009) that on the planetary scale the fraction of supercooled clouds (at -20°C) and the coincident dust aerosol frequency are negatively correlated, likely due to glaciations by dust."

Comment:

14995/19: Might want to add this paper: Hobbs, P. V., G. C. Bluhm, and T. Ohtake, 1971: Transport of ice nuclei over the North Pacific Ocean. Tellus, 23, 28

Reply:

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The measurements by Hobbs et al. were added in the text and reference list.

Comment:

14997/1: The many references to 'major' dust events should be scrutinized since no definition is given of the term. If they occur once a month, they are not unusual. So,what is being described as a 'major' versus a 'minor' dust event?

Reply:

We have added Kaminski‘s definition of “major” event (“produces visible deposits and optical phenomena”) in chapter 1. In chapter 2, when the event that we investigate is introduced, we classify it as “the strongest in 5 years” based on TSP-measurements at Hohenpeissenberg.

Comment:

14997/12 and throughout the paper: Wording like "ice nuclei observations" is a bit odd because of the use of the plural. It would be to replace it with " ice nucleus observations".The analogy for this is that "cloud observations" is better than "clouds observations".

Reply:

“nuclei” was replaced by “nucleus” throughout the text in conjunction with plural.

Comment:

14997/20: Was the loss of small particles by diffusion to the tube walls considered?

Reply:

This was investigated by measuring ambient air with a TSI 3785 Condensation Particle Counter a) for 10 seconds in front of the sampling tube and b) after 1 minute equilibration for 10 seconds after passing the sampling tube. The CN counts (at $\sim$ 6500#/cc) for b) are 1.5 % lower than those for a) as a mean over 9 switching cycles. This includes

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the environmental variability of CN.

Comment:

14998/11: The sampling rate of the deposited particles is not given. That rate, and the number of detected IN are factors in assessing the sampling error bounds, which should be stated.

Reply:

The retention efficiency of the aerosol sampler as a function of particle size and flow rate is given in Figures 4 and 5 and the text of the Klein et al. (2010) paper and is better than 96.7%. However one has to bear in mind that there are no standards available for a true calibration of ice nucleus measurements. The precision is 9%, derived from multiple sampling and analysis of test aerosol and was added to the text.

Comment:

14998/12-13: Was sampling onto the silicon wafers compared with other methods of IN detection? Is there, perhaps, a need here for a caveat that relative values of the measured IN concentrations are more definitive than the absolute values? Is linearity of the actual to detected IN concentrations assured, i.e. is the absence of any saturation or depletion effect

Reply:

Intercomparison: In the paper by Klein et. al. (2010) parallel measurements of the wafer sampling and analysis by FRIDGE and the continuous flow mixing chamber FINCH of our laboratory are shown (Fig. 6), which compare well. The wafer sampling technique did not participate in the 4th int. ice Nucleation workshop, but was developed as a consequence of it. We analyzed ATD samples from the workshop later in our lab and found good agreement with the majority of the other data of workshop participants, as shown in Fig. 4 of the paper by DeMott et al.: Resurgence in ice nuclei measurement research, submitted to BAMS, 2010. At present we have no indications

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for any such caveats against absolute values, but clearly intercomparison to other labs is required and will be done.

Linearity, saturation: we have varied the loading of the wafers over a factor of 10 by increasing the volume of air sampled from test aerosol. The maximum load of 2000 IN/wafer was the same as our highest particle load from atmospheric sampling during the dust event of May 2008. We have no indication for depletion and find a linear response over the range in which we operate the wafer/FRIDGE system. The data obtained during this test are given in the Table of the supplement.

Comment:

14999/7-8: The model data are "..related to .." or are actually what is being said?

Reply:

The reviewer is right, "related to" was taken out.

Comment:

Also, the designation, PM10dust is confusing to me, because it does not give a rapid distinction between measurement and model data. Perhaps the superscript could be changed from 'dust' to 'model'.

Reply:

PM10dust was replaced by PM10model throughout the manuscript, as recommended also by reviewer 3.

Comment:

15001/12 - Fig 6: Since the dust has been broadly distributed by the time it arrived in central Europe, why are there five distinct pulses? From Fig. 6 it appears that a diurnal patterns was superimposed on the signal with low values at night and high during the day. How can this be explained? The peaks appear (reading from the plot)

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just before noon each day, so neither greater atmospheric stability, nor mixing offer simple explanations. Does the model indicate pulses of arriving material, or changes in vertical structure? Is the daily updating of dust concentration (14999/16) a possible source of this?

Reply:

We disagree that the dust was broadly distributed when it arrived over central Europe. The horizontal plot (Figure 3) shows filaments and small structures that upon travelling can translate at a fixed location to distinct pulses. We have added a new plot in a N-S vertical plane along 10°E (12 Z on 29 May; new figure 4) which shows these individual pulses. The same variability in time can also be seen in the in the AERONET extinction data (not shown) from Leipzig (300 km east, http://aeronet.gsfc.nasa.gov/cgi-bin/webtool_opera_v2_new?stage=3&region=Europe&state=Germany&site=IFT-Leipzig).

Comment:

15001/22: The local effect argument is not very convincing unless there were distinct differences between the first and second half of the event. Is it certain that only reductions in IN could be ascribed to these sub-scale phenomena? There are numerous reports in the literature about large upward anomalies of IN counts accompanying thunderstorms.

Reply:

We agree that one can expect as a first guess vertical mixing to increase the surface dust concentration in the present case. This is can be read from the PM10 measurements of the new Fig. 7, which begin to rise at around 11:00* (Local time) and peak at 18:00 on 29 May, and rises at 9:00 and peaks at 16:00 on 30 May, both roughly in phase with diurnal mixing. The large and rapid fluctuations of PM10 also argue for vertical mixing. The overestimate of dust by the model on both days remains unexplained.

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* The dates on the time scale of Fig. 6 were initially placed at 00:00 of the day instead of 12:00, which mislead the reviewer (we apolologize, it was corrected).

Comment:

15002/2 - Fig.7: Is the time scale on the abscissa correct? 28.05, 29.05 .. or 28.5, 29.5? Also, it should be clarified which IN points are included in the insets (it seems that 7 points) vs the those in the time series (13 overlapping the model data) vs 15 given in Table 2.

Reply:

The time scale was corrected in the new Figure 8. The IN points used in the various analysis were clarified by symbol/color-coding and explanation in the captions of Fig. 8 and Table 2 : i) The IN points included in the inserts are the filled diamonds of Fig. 8a,b (n=12) and their means (triangles, n=7), the points for correlation analysis in Table 2 were ii) the means (triangles, n=7) for correlation to model data, and iii) all data (diamonds) of 24 May through 1 June (n=15) for correlation to measurement data.

Comment:

15002/17: Does 'significantly' refer to a statistical test, or just a subjective assessment?

Reply:

"Significance" is specified two and three sentences later, where we give for the correlations investigated: a) linear correlation coefficient R; b) the number of sample pairs n; and c) the error probability α.

Comment:

15002/27-28: How were the +/- limits determined?

Reply:

The +/- are the standard error of the mean for the data on ξ during May 27 to June 1st,

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2008 (12 samples), and this information was added to the manuscript.

Comment:

15003/13: Does the total number include all bins or just 1-7?

Reply:

This comment of the reviewer led us to the discovery of an error in our data: the activated fraction had erroneously been calculated using only the APS-data for dp > 1.0 μm. This was corrected. Now we use the SMPS+APS-data for dp > 0.2 μm and in addition present the same for the model data (dp = 0.2-12 μm, bins 1-7). The resulting numbers are of course smaller, which weakens our point 3, but are within the considerably scattered laboratory data from Field et al. and the ICIS-workshop (DeMott et al. BAMs 2010). The sentence on "the high activation" had to be deleted from the summary.

Comment:

15003/22: The figure number should be 9, not 8.

Reply:

was corrected

Comment:

15003/29: How can the deviation from the 1:1 line be stated to be insignificant? Only three of the points have error bars crossing the 1:1 line.

Reply:

Our statement referred to the regression line, not to individual data points. The 1:1 line lies within the 99.9% confidence bands of the data regression line . This is now stated in the revised text.

Comment:

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15004/2-3: The analysis here given appears to be, basically, a comparison of the slopes of the IN supersaturation spectra during the dust event and at other times during 2008/9. For this conclusion to be accepted, it has to be demonstrated that most, if not all other materials that may be potential IN exhibit appreciably different slopes. It would also have to be demonstrated that changes in size distribution have negligible effect on the slope. This is a specially difficult point to establish when mixtures of material may constitute the IN, as is likely to be the most frequent situation.

Reply:

See our reply above on the first comment of the reviewer.

References: Klein, H.: Variabilität der Eiskeimkonzentration über Zentraleuropa , Variability of ice nucleus concentration over central Europe, PHD Dissertation, (in german) University of Frankfurt, October 2010.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C8480/2010/acpd-10-C8480-2010-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 14993, 2010.