Atmos. Chem. Phys. Discuss., 6, S943–S949, 2006
www.atmos-chem-phys-discuss.net/6/S943/2006/


Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Some ice nucleation characteristics of Asian and Saharan desert dust" *by* P. R. Field et al.

**P. R. Field et al.**

Received and published: 22 May 2006

We thank the reviewer for their comments. Here are our point-by-point responses.

Specific comments

1. We feel that this sentence, which is now modfied, describes briefly what the CVI residuals are.

'...the dominant residue (nuclei on which the ice formed) during that event found after in-situ ice particles were captured and evaporated using a Counterflow Virtual Impactor (CVI).'

We have added nucleation definitions to introduction and additional references:

'The nucleation of ice crystals can proceed via a number of different pathways (e.g.

Vali, 1985; Pruppacher and Klett, 1997; Cantrell and Heymsfield, 2005). The main nucleation mechanisms are commonly referred to as (following Vali, 1985) i) homogeneous nucleation - the fomation of ice through the random motions of water molecules becoming arranged as an ice embryo. This mechanism is highly temperature dependent and in the atmosphere is not important for clouds warmer than -38$^o$C. There is good agreement between homogeneous freezing rates predicted theroretically and those measured in the laboratory (Mohler et al. ;Cantrell and Heymsfield, 2005). ii) Deposition nucleation - the formation of ice by vapour deposition directly onto the surface of insoluble ice nuclei. These nuclei are thought to originate mainly from crustal sources (e.g. desert dust), biomass burning or industrial processes (e.g. soot or metals). iii) Condensation freezing - this process involves the activation of a liquid droplet on a condensation nucleus that acts to freeze the droplet during the condensation stage. iv) Immersion nucleation - an insoluble ice nucleus suspended within a supercooled droplet causes the droplet to freeze. v) Contact freezing - this process again requires liquid droplets to be activated whose surface then subsequently comes into contact externally, or internally (Shaw), with an insoluble ice nuclei that causes the droplet to freeze. Pruppacher and Klett (1997) additionally state that immersion freezing occurs through the action of the ice nucleus as a cloud condensation nucleus at temperatures above 0$^o$C which subsequently causes the droplet to freeze when it is cooled below 0$^o$C. In contrast, the sequence of events for condensation freezing all occur at temperatures colder than 0$^o$C. In the experiments presented here we will never activate a droplet at temperatures above 0$^o$C, and so we will refer to events where it is believed that droplets are freezing via a heterogeneous mode as simply condensation or contact freezing.'

2. Added a more explicit statement to the introduction about what can be found in this paper and the companion paper:

'In this paper we report on a series of laboratory experiments carried out at the AIDA cloud and aerosol chamber facility in which two desert dust samples were tested under



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realistic atmospheric conditions and in relatively large numbers. This paper focuses on the results for the desert dust samples that formed ice at temperatures between -55$^o$C and -20$^o$C empahsising observations made with the Small Ice Detector that is capable of determining the phase of particles. The companion paper by Möhler et al. (2006) deals with the ice nucleation events that occurred in the -80$^o$C to -50 $^o$C temperature range. That paper contains extensive information concerning the accuracy of the various measurements within the chamber and the methods employed in analysis of the dust samples, that is precised in this text.'

3. Added some discussion of the importance of IN to the climate:

'The indirect effects of ice nuclei that include the impact on cloud albedo, cloud lifetime by changing the mean size and/or the phase of the particles have been investigated using global climate models (Lohmann and Feichter, 2005). Lohmann and Karcher (2002) find that anthopogenically produced aerosol has little effect on the formation and maintenance of cirrus for temperatures colder than -35$^o$C where homogeneous nucleation dominates. At warmer temperatures where heterogeneous ice nucleation processes dominate, the effects on cloud albedo, lifetime, and precipitation are likely to be more profound (Lohmann and Feichter, 2005). Large vertical extents of the upper troposphere are supersaturated with respect to ice.(Gettelman et al. 2006). If deposition ice nuclei become more prevalent then this could lead to increased cloudiness, although it is difficult to predict what the outcome of increasing the concentrations of ice nuclei will be in a system with cloud feedbacks of undetermined sign (e.g. Stocker et al. 2001). These results suggest that the role of desert dust in the evolution of clouds and precipitation may be important and provide motivation for the present study reported here and in a companion paper (Möhler et al. 2006). '

4. Further analysis of the dust samples has shown that less that 1% of the mass is contributed by soluble material. Therefore, we have added the following to the discussion:



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'These saturation ratios are close to what Zuberi et al. (2002) obtained for dust samples immersed in aqueous droplets ($(NH_4)_2SO_4$-$H_2O$, 10-50 $\mu$m in size). In this case, no droplets were observed with the SID, but Archuleta et al. (2005) suggest that condensation on hydrophillic sites may be important at these temperatures and humidities. Analysis of bulk dust samples used in these experiments showed that $<$1% by mass was made up of soluble material. We can speculate that if the soluble material is mainly found as surface coatings then the fraction of soluble material on the smaller particles used in these experiments is likely to be greater as the surface area to volume ratio is inversely proportional to particle size. Therefore, although we cannot definitely rule out whether the secondary nucleation events (region III) represent a further deposition nucleation mode we can allude to the presence of soluble material in the bulk sample and the good agreement with the Zuberi et al (2002) results to infer that is it likely that a condensation freezing mode is acting in region III.'

Inorganic and organic coated dust samples were the subject of a subsequent measurement campaign ata the AIDA chamber. The results are currently under analysis.

5. On each day the same samples are temperature and pressure cycled several times (see section 2). At cold temperatures (colder than -50C) the sizes of the ice crystals are small and not many are sedimented out before the chamber is reset for the next expansion and they are sublimated. At the warmer temperatures, the ice particles can grow bigger and sediment out. Under these conditions, it is difficult to estimate what fraction of the active nuclei have been removed and will not take part in the subsequent expansion. We have added the following text into the discussion:

'Roberts and Hallett (1968) also reported that pre-activation of aerosol leads to ice production at lower saturation ratios. Over the course of a day the aerosol samples in the AIDA chamber are cycled a number of times up to water saturation and then back to just below ice saturation. Unfortunately it is difficult to assess the fraction of nuclei lost to sedimentation that formed ice during a previous expansion. If it is assumed that aerosol that activated ice crystals are not lost via sedimentation and participate



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in subsequent expansions, then the two series of expansions (26–29, 40–42, fig. 6) colder than -50°C indicate that the condensation freezing requires higher humidity on each subsequent run. However, it should be borne in mind that the observed change in humidity at which the immersion freezing occurs is of a similar magnitude to the measurement uncertainty and could also be influenced by systematic errors due to detection time at different cooling rates.'

6. We refer to Mohler et al. (2006) to provide these details.

7. SID - University of Hertfordshire, UK. Added this information to the text. CPI - SPEC INC., is already given in the text.

Added more text to describe the SID:

'Single particles must therefore be differentiated from ice by using a threshold value for the asphericity. Using a threshold can lead to some non-spherical particles being classed as spherical and vice-versa. Field et al. (2004) assessed the choice of $Af$ threshold values by sampling stratocumulus clouds warmer than 0°C where only liquid droplets would be found and synoptically generated cirrus colder than -40°C where only ice crystals are expected to be found. These two datasets indicate that if an $Af$ value of 12 is used to threshold the particles into spherical and non-spherical then 10% are mis-classified. From intercomparison with other aircraft probes the uncertainty in 1 Hz concentration is estimated to be $\sim$20%. The lowered count rates encountered in the laboratory setting will also result in a non negligible Poisson counting error. The SID probe sampled horizontally from near the base of the chamber with an estimated flow speed of $\sim$5 m $s^{-1}$ ($\sim$1 $cm^3$ sampled per second). Intercomparison with other probes in the AIDA chamber (see Möhler et al., 2006) indicated that this flow speed may have been underestimated and so the SID concentrations calculated using the 5 m $s^{-1}$ flow speed were multiplied by 1.6. This laboratory flow speed is very different from aircraft speeds so appropriate scaling of the calibration constants for sizing were applied based on the increased time-of-flight through the probe detection volume.



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Particle sizing from this instrument assumes that the particle is spherical. Therefore, non-spherical particles will be incorrectly sized, but this 'spherical diameter' can still be used qualitatively to assess if ice particles are growing. In the following analysis, results are accumulated over 10s for estimating the activated fraction and so, given the sampling rate, this sets a concentration detection limit of $\sim$0.1 cm$^{-3}$. '

8. We have added a table the provides a summary of details described in Mohler et al. 2006. The APS and SMPS were used to characterise the aerosol and those details are reported in Mohler et al. 2006.

9. See 7, above.

10. See table 1. , point 7 above.

11. Changed sentence to this:

'This suggests that the dominant ice nucleation mode in this case is a condensation mechanism or possibly contact freezing, although the short timescales involved probably preclude the action of contact freezing and remains to be tested with parcel modelling studies. The resulting ice formation activated about 10% of the aerosol (panel f)'

12. Now covered in the introduction - see point 1, above.

13. We think that the noisiness of the signal is due to high condensed water concentrations causing strong attenuation of light in the TDL system.

14. We do not know about the morphology of the dust samples used in the chamber.

15. Added dust sample types to figure captions. 1-5

16. One step forward would be to use a CVI and look at ice crystal residual nuclei at different times corresponding to the different nucleation events.

17. We have lengthened the discussion and speculated about implications for the



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atmosphere.

Technical corrections

1. Fixed - this sentence now reads:

'Möhler et al. (2006) found that the fraction of aerosol that nucleates ice is dependent only on supersaturation with respect to ice and does not appear to be affected by the cooling rate. This suggests that if the first runs are carried out at slower pumping speeds then the dual nucleation may become apparent for the first expansion also, simply because this allows a longer temporal seperation between the nucleation events.'

2. New fig 6 caption:

'Plot shows the ice saturation ratio and temperature at which ice nucleation was observed to begin. Open symbols are the results for the Asian desert dust sample (AD1), filled symbols are for the Saharan desert dust sample (SD2). The numbers adjacent to each symbol represent the experiment number (see table 1). Downward pointing triangles denote when the fraction of aerosol forming ice crystals exceeded 0.5%. Upward pointing triangles denote when the fraction of aerosol forming ice crystals exceeded 8%. Circled symbols indicate when examination of the time series for the experiment clearly showed two distinct nucleation events. Region I: droplets form at the same time as ice (condensation freezing, or homogeneous freezing for temperatures close to -40$^{o}$C. Region II: deposition nucleation. Region III: condensation freezing (see text). Dashed line indicates the results from Zuberi et al. (2002) for freezing of aqueous droplets ($(NH_4)_2SO_4$-$H_2O$) containing dust.'

3. Fixed fig. 7 caption.

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 1509, 2006.