

## ***Interactive comment on “Studies of heterogeneous freezing by three different desert dust samples” by P. J. Connolly et al.***

**P. J. Connolly et al.**

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General comments

Just to clarify, the parameterisation can be use in cloud models where the cooling rate exceeds 1K / min.

Specific comments

1. Agreed, this has been changed
2. The reviewer is correct in stating that IN could have size dependent nucleation rates as has been shown by other authors. In our approach the largest particles certainly are the most active IN. For example, take the Sahara dust at -30C, if the size of the dust contained within the drops is assumed to be 0.1 micron radius then only 10% will freeze, whereas if it is assumed to be 0.3 micron radius then 100% will freeze. The aerosol size is used in the equations in the paper via the definition of surface area. As described in the paper we did this so not to introduce insurmountable complications. Additionally, our approach is different to Archuleta et al 2005, who were looking at heterogeneous freezing at humidities below water saturation. They also used the classical nucle-

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ation approach where ice formation has explicit time dependence. We used the singular hypothesis. We tried for a long time to explain the behaviour of the IN with a contact angle approach. The only way we could get anything like good agreement was to have a very broad distribution of contact angles distributed throughout the dust sample otherwise freezing occurred much too quickly when compared to the data. Again the contact angle approach is different to our approach as we used the singular hypothesis and the contact angle approach yields a nucleation rate. The singular hypothesis works well for this data (moderate cooling rates). We also feel that the contact angle approach is somewhat meaningless for irregularly shaped particles such as these and prefer to have surface sites that have different activation energies for what ever reason.

3. We are using both the small ice detector, the WELAS and the CPI for concentrations; the SID and the CPI are able to distinguish between the phase of the particles (liquid or ice) and are used for some of the dataset (as shown in Figures 6, 9 and 10). Unfortunately the SID was not available for all of the experiments so we used the CPI, with concentrations corrected using the Connolly et al algorithm as described in the text, page 469, line 15). For the experiments where we had both CPI and SID we did a comparison of the determination of  $n_s$  from both SID and CPI and found excellent agreement (see Figure 3). This is explained on page 476, line 15. Also the crystals grow into sizes observable by the CPI very quickly in all of the experiments use here.
4. This is a valid point. However, we did not mean to draw attention to the nitrate salts but the chemical ageing and modification of the mineral particles by this process. This has been clarified in the text by changing the discussion to read "It has been shown by Krueger et al, 2004 that certain Ca containing compounds such as calcite and dolomite may react with nitric acid in the atmosphere to form nitrate salts; hence potentially modifying the chemical and physical properties of the dust. However, one might expect that the nitrate salts would reduce the IN activity. Another process that may be important to increasing the IN activity is chemical aging due to oxidation of the mineral surface by ozone. More work is needed to understand the impacts of such chemical aging processes on the dusts ability to act as an IN"

#### Technical corrections

1. OK
2. OK. Don't think the word 'This' should be removed as it doesn't read well. Perhaps you made a mistake?
3. OK
4. OK

5. OK
6. OK
7. OK—this must have been a reproduction mistake? Altocumulus was defined on page 465, line 2.
8. OK
9. OK

All changes have been applied to figure captions. I would prefer to keep the crystal images as is, the reasons being (1) I think the shapes can be seen at this size; (2) by showing a relatively large number of images it gives the reader an idea of how statistically significant the shapes are, if we showed only a few that would leave a question mark on this.

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 463, 2009.

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