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Discussion paper

Interactive comment on "Global impact of mineral dust on cloud droplet number concentration" *by* Vlassis A. Karydis et al.

Anonymous Referee #2

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Anonymous Reviewer #2. Review of manuscript "Global impact of mineral dust on cloud droplet number concentration" by V. Karydis et al.

This work uses a suite of models (including the atmospheric chemistry model ECHAM5/MESSy, MECCA, aerosol thermodynamics with ISORROPIA-II, and a series of other aerosol micro-physics subroutines) in order to explore, through numerical experiments, the potential global impact of wind-blown mineral dust in the number concentration of activated cloud droplets. Three mechanisms are explored in the paper: adsorption over insoluble dust particles, classical activation on particles with soluble coating, and a second order effect which involves interaction of the mineral cations in the dust particles with other inorganic aerosols. These mechanism are explored through sensitivity simulations in which the model is run with/without the process under consideration. The paper is relevant and well written. The material presented is novel since it attempts to quantify potential impacts of mechanism not previously considered. However, I think the discussion of the implications of these mechanism should be performed in much more depth than what is done in the paper, and some substantial modifications in technical details are needed for the paper to be published. The difference between the conclusions found in this study and a previous work (Karydis et. al 2011) should be made explicit.

General Comments:

- No description of the cloud scheme utilized in the model is done. Therefore, it is not clear under which conditions is the activation parameterization triggered. Very little or no mention of cloud microphysics is done in the paper. The distribution of low level cloudiness in the model is not presented, which would be crucial to determine the actual extent of global impact of CDNC on aerosol-cloud-radiation interactions.

- Although the paper is mainly focused on the impacts of dust on CDNC, no mention is done regarding the impact of dust on number concentration of aerosol particles that could activate. It would help in the interpretation of the results to know what the impact of switching dust emissions off is on the number and size of aerosol particles. A figure showing the changes caused by dust on the aerosol particles should be shown next to Figure 5.

- There is no mention in the paper about the geographic distribution of soluble and insoluble fractions in the dust modes predicted by the model. This would definitely help with the discussion and interpretation of the results. A map showing this distribution would help understanding the underlying processes.

- The paper does not explain how the CDNC shown in the maps is calculated. Are those grid-cell averages? Are those in-cloud values? Is this the value only after activation subroutine is called? Or are these values produced by the full cloud-microphysical scheme?

- No indication of the frequency of occurrence of liquid clouds at the level in the model is mentioned, nor that of the climatological cloud cover in those regions. If this is somehow included in the manuscript, the overall importance of dust on CDNC globally could be better assessed. The specifics of the annual average CDNC shown in the paper should be discussed and described in detail.

- Some fundamental issues with the unified theory should be discussed by the authors in this manuscript. In particular the potential oversimplification of the activation process for insoluble particles with small soluble coatings (as could potentially be the case for dust particles). See specific comments.

- I suggest modifying some of the conclusions of the paper, since they can be over-reaching. It doesn't seem that the paper actually "demonstrates" that the biases are substantial, or that this treatment is indeed correct. In fact, the authors acknowledge almost no sensitivity of CDNC to massive cuts in dust emissions, or hydrophilicity parameter, or on dust chemical composition. For example, I quote "By assuming drastic differences in the dust source and the dust hydrophilicity we find only small ($\sim$5%) changes in the average CDNC".

Specific Comments:

Section 2.2. Line 265. It is not clear from the equations nor the references cited in the document, how can an exponent x = -3/2 be obtained from equation (3) when there is insoluble material but no FHH terms. The -3/2 exponent arises from the fact that the whole volume of the particle contributes to the soluble material during the activation process. It is not explicit from the document what is the expression relating critical diameter and critical supersaturation when there is a substantial fraction of insoluble material (i.e., in equation 3, with no FHH terms, but a small amount of soluble material). The relations between dry aerosol size and critical supersaturation are severely modified when an insoluble core is present (see for example, Pruppacher and Klett, chapter 6, equations 6-37 to 6-42). Therefore, there is a possibility that one could see sub-

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stantial changes in CDNC by simply improving the description of the relation between critical diameter and critical supersaturation for cases where there is an insoluble core (no FHH terms). This issue should be explored and discussed in the paper.

- It would be convenient for the readers to see average values of CDNC, or average fractional changes printed in the global maps of figures 1, 2, 5, 6, 7 and 8.

- The difference between results shown in Figure 5, and Figure 8c are not entirely clear to me. So figure 5 has no mineral dust emissions, and Figure 8c, was performed with 50% aerosol emissions compared to base case? So in the case of no emissions, there is a net decrease in CDNC, but when there is only a 50% decrease in the emission load there is an increase in CDNC?

- Line 712. Should it read “insensitive”?

- Figure 2. Are these values grid-cell averages? Or are they in-cloud values only?

- From figure 8, it seems that BFHH parameter has a larger (or at least comparable) impact to reducing mineral dust emissions by 50%? This should be discussed in much more detail. As mentioned above, perhaps showing the net impact that the 50% reduction in dust emissions has on aerosol number concentration would be helpful in the interpretation of the results.

- Similarly, the paper shows very little sensitivity of CDNC to dust chemical composition, but relatively high sensitivity to the BFHH parameter. However, it is reasonable to believe that the FHH parameters are linked to the chemical composition of the mineral dust particles. Therefore, some discussion should be included regarding the relationship between the FHH theory parameters and dust chemical composition, and the potential impacts it could have in the simulations.

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