**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Observations of heterogeneous reactions between Asian pollution and mineral dust over the Eastern North Pacific during INTEX-B" *by* C. S. McNaughton et al.

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—— Please see the initial paragraph of the response to reviewer J. Kelly regarding the suggestion of an analysis of the uptake of chlorine by dust. This suggestion proved enormously helpful and the manuscript has been revised to reflect this.

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P. 8489, line 21

Statement revised to:

While there is the potential that these species are simply co-emitted at the source, Sul-

livan et al. [2007] found ammonium can be associated with sulfate rich submicrometer dust leading to inaccuracy based on the Jordan et al., assumption that all ammonium is associated with submicrometer sulfate or nitrate.

p. 8494, line 7

Agreed (typo actually), I assumed 20% $Ca(NO_3)_2$ only.

p. 8495, line 23 and sub-points 1 - 4

There were no measurements of organic aerosol with which to better estimate composition dependent refractive indices. Ammonium sulfate is used here for illustrative purposes.

Sub-point #1 – 4

Laboratory measurements by Grassian indicate that the reaction with calcite is not surface limited and can occur throughout the bulk particle.

p. 8473, line 16. Citing a model study by Bauer et al. (2007) to describe the mass coating and hygroscopicity of aged mineral dust seems odd to me.

The work of Bauer et al. and the conclusions drawn from them are based on flawed assumptions regarding the hygroscopicity of dust which has reacted with atmospheric acids. It is discussed in this manuscript to identify this error and to illustrate that the dominant effect of heterogeneous reactions may be their influence on the direct effects (i.e. extinction) of the accumulation mode aerosol.

p. 8474, line 8 - "Dust particles larger than â′Lij0.2 _m are all eïnËĞAËŸ ective cirrus cloud ice nuclei (Archuleta et al., 2005)" is incorrect. The IN properties of mineral dust are well known to be a strong function of mineralogy. . .

I disagree. I believe there is insufficient data to determine the "strength" that particle size versus its composition have on their ice nucleating abilities. Chinese Loess is composed mostly of illite (51 +/- 6%) with a smaller fraction of kaolinite (19 +/- 2.8%)

and montmorillinite (8.6 +/- 3.7%). Calcium carbonate concentration varies with grain size (3.6-21% with an average of 12%), and is contained predominantly in the medium to coarse, 6-60 μm, silt grains. Suspension causes winnowing and a relative depletion of Ca:Al, while Fe:Al ratios remain effectively constant [Arimoto et al., 2006]. Keep in mind that the geometric mean diameter of our dust samples are only 0.6 ïA▪m at concentrations of 1.5 – 8 particles cm-3.

Zimmerman et al., [2008] state: Particle diameter varied mostly between 1 and 10 ïA▪m, few particles were larger with diameters up to 100 ïA▪m. However, ice nucleation mostly occurred on particles <10 ïA▪m. No effort was made to further resolve size dependencies that may affect IN efficiency and the role of specific mineralogy.

Zimmerman also state: In summary, all silicate minerals and hematite are activated (1–3%) at temperatures of _10 to _13_C. For calcite (_14_C) and gypsum (_16_C) the temperatures of 1% activation are lower. Kaolinite, illite, and hematite seem to be the most efficient IN.

Eastwood et al. did not collect atmospheric dust samples, they created them by comminution of commercially available mineral standards stating: A typical sample held between 100 to 1000 individual particles, the majority of which were between 1 and 20 ïA▪m in diameter. The average sizes of the particles used in our experiments were 7.7 ïA▪m for kaolinite, 9.0 ïA▪m for muscovite, 8.1 ïA▪m for montmorillonite, 10.0 ïA▪m for quartz and 14.2 ïA▪m for calcite based on the optical microscope images. These particle sizes and their composition differ from natural dust. They also state: [laboratory] measurements indicate that muscovite and kaolinite are very good ice nuclei with onset RHi values of less than approximately 110%, well below water saturation.

Archuleta specifically investigated the effect of size versus composition for IN using Asian dust samples and found: For a selected size of 200 nm, the natural mineral dust particles were the most effective ice nuclei tested, supporting heterogeneous ice formation at an ice relative humidity of approximately 135%, irrespective of temperature.

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Still, the point is well taken. I have significantly revised the introduction regarding the reference to Achuleta the relevant portion now reads:

Particles with diameters greater than $\sim$2 ïA▪m activate regardless of composition for supersaturations typical of continental and marine cumulus cloud ($\sim$>0.2%) [Kelly et al., 2007]. At 0.2% supersaturation CaCO3 and SiO2 with a 1% coating of gypsum, will activate if they have dry diameters greater than $\sim$1 ïA▪m. Activation of "completely insoluble" dust particles in the 0.6 – 2.0 ïA▪m size range is facilitated by the presence of slightly soluble compounds [Kelly et al., 2007]. These findings are supported by field measurements which show that calcite–containing particulate are acting as CCN [Matsuki et al., 2009; Twohy et al., 2009]. The size, composition and mixing state of dust particles will co-determine their effectiveness as ice nuclei (IN). At -10 oC to -40oC and relative humidity with respect to ice (RHi) of 110-135%, kaolinite, illite, muscovite and hematite are the most efficient IN while montmorillonite, quartz, calcite and gypsum are the poorest IN [Eastwood et al., 2008; Zimmermann et al., 2008]. [Eastwood et al., 2009] recently determined that heterogeneous reactions can inhibit the ice-nucleating ability of kaolinite. Whereas a separate study [Archuleta et al., 2005] found that Asian dust particles larger than $\sim$0.2 ïA▪m are all effective cirrus cloud ice nuclei at RHi of $\sim$135% irrespective of temperature. Given the difficulty in simulating relative humidity [Petch, 2001] and aerosol indirect effects using GCM's [Penner et al., 2006], it is difficult to have confidence in recent model estimates of the influence of heterogeneous chemical reactions on atmospheric residence times of mineral dust.

Page 8487, line 12

All samples are from the FT. There is no indication of the presence of seasalt as indicated by the molar ratios of Ca:Mg and Ca:Na.

Page 8488, line 8.

The HR-ToF-AMS used aboard the NCAR C-130 is an Aerodyne instrument and does not have the same capabilities as the K. Prather ATOFMS used aboard the Ron Brown

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during ACE-Asia. Thus no size resolved information regarding the association of acid species with Ca, Fe, and Al can be derived from the data.

p. 4893, line 5.

Thank you for the reference statement revised to:

These data points have geochemical ratios nearer to sea-salt, likely contain primary salts derived from the dry lake beds of the Central Plateau (e.g. Texcoco [Moffet et al., 2008; Moya et al., 2004]), and were visually observed (CMc) as dust sources from on board the DC-8 during INTEX-B.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 8469, 2009.