**Atmospheric
Chemistry
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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.

C. S. Zender

zender@uci.edu

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General Comments

The manuscript presents a detailed exposition of what can be inferred about heterogeneous chemistry during long range transport of clean and dusty air. The manuscript succinctly explains the instrumental details and purpose and proceeds logically through the well-designed number closure test, and then to the chemistry and more interpretive and integrative measurements. References to earlier measurement campaigns seem appropriate (but I am not a field atmospheric chemist). The manuscript will also have

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an important broader impact, one hopes, in large scale aerosol models, CTMs, and GCMs (which I am much more familiar with). This is because the authors quantify the extent to which typical model simplifications of heterogeneous processes lead to inaccurate representation of aerosol size distribution and light scattering (and thus radiative forcing).

I recommend publication after the minor points below are addressed.

Specific Comments

1. p. 8494, line 13: Fixing the refractive index for this sensitivity experiment means that only the optical property change due to aerosol uptake is quantified. The optical properties also change because of the conversion of Ca to CaSO4 or CaNO3. According to Long *et al.* (1993), the refractive indices of calcite and gypsum at 0.55 μm are $1.47 - 2.25 \times 10^{-6}$i and $1.40 - 1.38 \times 10^{-5}$i, respectively. That these both differ significantly from the $1.53 - 0.006$i $\times 10^{-5}$i that the authors assume is no surprise or problem. This is expected because the authors use a refractive index for the bulk aerosol, not a single, dry, un-aged component. My point here is that the conversion of, say, calcite to gypsum also changes, possibly significantly, the intrinsic refractive index of the multi-component aerosol. Depending on the conversion, this may amplify or reduction the change in accumulation light scattering due to size-competition that the authors already note.

2. Figures 1–4: The number closure (Figures 2 and 4) appears quite robust. But it is unclear from Figures 1 and 3 that only a negligible amount of mass would be missed in particles larger than $\sim 8 \,\mu$m. The increase in mass distribution from $5 < D < 8 \,\mu$m appears to be a distinct a mode of the particle size distribution. Is it real or due to poor counting statistics? If real, I would have to agree with Reviewer 1 that multiple coarse modes (or section) would better represent the

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measurements. The implied mass-related characteristics such as settling velocity and micronutrient content may be underestimated by the current fits.

3. The manuscript is exceptionally dense and meaty. Reading it gave me the claustrophobic feeling of being in an aircraft crammed with sophisticated instruments.

4. Figures 1 and 3: The measured size distribution of the refractory material varies rapidly and then cuts off suddenly near 8 μm, as mentioned on p. 8478. It should again be mentioned when introducing these figures where this feature is quite pronounced.

5. p. 8485: The increase in SSA during long range transport across the Pacific is an interesting result!

6. Figures 2 and 4: There seems to be a slight low bias in the lognormal fit number concentrations for unheated and refractory aerosols in Hawaii (Figure 2), and a slight high bias of same in Alaska (Figure 4). Why would this be the case? Are the source regions for the air masses sampled similar?

7. Figure 8: Correcting the anion imbalance of the aged aerosol to elucidate the composition of the source dust aerosol (calcite, dolomite) is clever!

8. p. 8499: These results are also consistent with Bian and Zender (2003), who used a CTM to estimate that dust reduced HNO3 by 60% in and downwind of dust source regions in summer.

9. It seems worth noting that the observed 30–70% reduction in scattering by accumulation mode aerosol in mixed dust conditions, presumably due to heterogeneous chemistry sequestration of gases, will reduce tropospheric photochemistry. This has important implications for tropospheric O3.

10. When discussing the competition for condensing secondary species, it may be worth noting that surface area alone does not determine the winner. The large particles will condense more species due to the Kelvin effect, i.e., the reduction in saturation vapor pressure over large particles compared to small. The positive feedback between growth and condensation due to the Kelvin effect is why the coarse aerosol condenses more species.

Technical Corrections

1. p. 8472: Define FT as Free Troposphere first time used

2. p. 8474 line 27: Eliminate "likely"

3. p. 8485 lines 17–20: Re-word this sentence.

4. p. 8492: Should be "alt > 1.5 km a.g.l."?

References

Bian, H., and C. S. Zender (2003), Mineral dust and global tropospheric chemistry: Relative roles of photolysis and heterogeneous uptake, *J. Geophys. Res.*, **108**(D21), 4672, doi:10.1029/2002JD003143.

Long, L. L., M. R. Querry, R. J. Bell, and R. W. Alexander (1993), Optical properties of calcite and gypsum in crystalline and powdered form in the infrared and far-infrared, *Infrared Physics*, **34**(2), 191–201.

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