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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.

C. S. McNaughton et al.

csmcnaug@hawaii.edu

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—— Both J. Kelly and R. Sullivan reviewers recommended an analysis of the uptake of Cl- by the dust. In fact this analysis proved enormously productive for all three scenarios, the INTEX-B data over the Pacific, the data collected near Mexico City and the long-term data from Mauna Loa. The relevant sections have been updated and the manuscript revised based upon these results.

I have also condensed the analysis of the UT/LS cases and included the relevant data points into the analysis over the ENP instead. Specific replies to individual comments are included below.

1. The log-normal fits to the measured size distributions do a poor job of representing the features of the coarse-particle volume distributions (Fig. 1 and 3).

The reviewer may be concerned about the “spiky” behavior evident in the volume averages in Fig. 1 & 3. This is an artifact of two phenomenon. First, the data begin to approach the Poisson counting regime making any fitting to very few particles inadvisable. Second, Mie oscillations in the data due to OPC calibration produce local minimum and maximum in the distribution. While this is partially smoothed by combining the data with the APS distributions, they can never be completely eliminated.

The least squares fitting routine simultaneously fits the 0th through 3rd moments of the size distribution. The residuals to the fitting are highest for the 0th (number) and 3rd (volume) moments. The sum of the square of the residuals for the 3rd moment average 34% of the total sum of squares (0th through 3rd moments) of the residuals, however the sum of the square of the residuals are generally low. Despite the use of multiple modes by other researchers to fit dust distributions the data presented here do not indicate that additional log-normal modes are required to fit the observations.

2. The authors conclude that the presence of Asian dust led to a 25 percent smaller number median diameter for the accumulation mode. Such a change in diameter would have significant implications for light scattering by particles.

The complexity of the origin and transport histories of the polluted versus pollution plus dust airmasses do not allow us to definitively eliminate the possibility that the dusty influenced airmasses simply were transported with accumulation mode aerosol with smaller median diameters. However, some support for our hypothesis is given by the measurements of submicrometer scattering. All else being equal, when the submicrometer nephelometer is operating behind the 1-\mum impactor, we expect that in the presence of dust, submicrometer scattering would increase due to the sub-micrometer tail of the dust distribution. In fact submicrometer scattering during dusty periods is

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only 80% the values measured in the absence of dust. Again, this does not eliminate the possibility that the accumulation aerosol are completely distinct based on emission strengths at the origin. But the independent submicrometer scattering measurements are consistent with reduction in the median diameter of the accumulation mode leading to a reduction in scattering by these particles.

2. The concept of surface-area based competition is reasonable for a condensing compound like sulfuric acid, which is effectively non-volatile, but it does not always hold for nitric acid, which is semi-volatile.

I agree with the reviewer that for accumulation mode aerosol the presence of particulate nitrate will be affected by particle acidity. But note that the equivalents ratio of ammonium to sulfate indicates that the aerosol is usually neutralized. C. Zender also points out that the Kelvin effect will favour condensation of non-volatile and semi-volatile species onto supermicrometer rather than submicrometer particles; though we have not estimated the significance of this effect. Finally, nitrate is a small component of the total mass compared to sulfate and organic aerosol. Thus the majority of the "missing mass" is likely attributable to these species rather than it being dominated by nitrate.

4. Are the source regions for the Hawaii and Alaska measurements roughly the same?

Back trajectories were examined. The trajectories arrive in Asia between the Chinese/North Korean border and the Northern tip of Taiwan and spent 5-7 days in transit. Main difference between Hawaii and Alaskan Trajectories are their sense of curvature for entrainment into the Alaskan Low or East Pacific High.

5. Evidence of HCl uptake by calcite-containing dust particles has been reported in previous studies. Did the authors find evidence of HCl reactions with dust components in the present study?

After analyzing the data we did find an increasing trend of aerosol Cl- with dust for

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the INTEX-B and MILAGRO DC-8 data. We also looked at the MLO data and found supporting evidence for this reaction. Thanks to both reviewers for pointing this out it has substantially increased the relevance of the manuscript.

p. 8474, line 7: Removed.

p. 8478, lines 7-14

Using the ammonium sulfate composition to correct the optical to geometric size distributions results in ∼80% closure between scattering calculated from the distributions and scattering measured by the TSI nephelometer. There were no measurements of organic aerosol with which to better estimate composition dependent refractive indices. The correction is a maximum of ∼4% diameter shift for submicrometer aerosol results in a maximum of ∼1.04ˆ3 = 1.12 change in volume per particle. The changes are smaller for supermicrometer aerosol and result in little difference between unheated and refractory distributions.

p. 8478, lines 15-18

Statement revised: Counting statistics and sizing accuracy of the OPC is typically poor beyond 8.0 ïĄ■m, thus data above this size range is typically eliminated from the data sets.

p. 8479, line 22: Corrected p. 8480, line 18: Deleted

p. 8481, lines 8-9: Added: The largest disparity occurs between the OPC and the LDMA over the 0.30-0.50 mm size range. OPC number agreement is 71% (R2=0.922) and volume agreement is 60% (R2 = 0.910) due to the large Mie oscillation at these sizes. The overall agreement for the total region of OPC and LDMA overlap (0.12 - 0.50 mm) is 92% (R2=0.918) for aerosol number, and 85% (R2=0.966) for aerosol volume.

p. 8484, lines 14-15: Unfortunately No. There is no independent measurement of mass for comparison.

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p. 8485, lines 8-9: To our knowledge there are no completely volatile primary aerosol save perhaps sulfate produced in volcanic vents. The heated CN number concentrations and distributions are corrected for laboratory derived thermophoretic losses which are up to 17% of total (CN > 10 nm) number.

p. 8485 line 24 and p. 8486 line 3: Same measurement of scattering at 550 nm but 0.90 refers to fine-mode dominated conditions (FMFscat>0.6 – p. 8485, line 22) whereas 0.91 refers to mixed conditions (0.3<FMFscat<0.6 – p. 8486, line 1).

p. 8486 lines 14-15: Statement revised: The Hawai'i data show the lowest scattering Ångstrom exponents and the highest absorption Ångstrom exponents while the data from the CBL near Mexico City have the lowest single-scatter albedo's.

p. 8488, line 25-26: Revised p. 8491, line 22: Removed p. 8497, line 10: Revised p. 8498, line 28: Revised p. 8533 p. 8534: Revised

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