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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Evolution of Asian aerosols during transpacific transport in INTEX-B" *by* E. J. Dunlea et al.

**E. J. Dunlea et al.**

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ACPD Manuscript ACPD-2008-0400 Response to Reviewer 1

Notes Reviewer comments are designated by >; Responses are designated by >>; figure numbers in responses refer to numbers in revised version

Response to Reviewer #1

> 1) Fig. 2: remove panels (a) and (c). These do not provide much useful information.

>> This figure has been modified as suggested by the reviewer.

> 2) Fig. 3: remove figure. This figure contains information that is duplicated in Figs. 4 and 5, and the time-plot format is less informative than the scatter plots and regressions in the later figures.

>> This figure has been left in the paper. We feel that showing the agreement of the various aerosol instruments in time conveys the agreement amongst the instruments in a way that only showing scatter plots does not. This figure has been updated slightly in response to comments from Reviewer #2.

> 3) Figs. 4 and 5: Combine figures into one with four panels. It is helpful to include top and right mirrored axes on such plots to help identify the value of points without having to use a straight edge.

>> This figure has been modified as suggested by the reviewer.

> 4) Fig. 6: remove figure. This information is already in Table 3.

>> This figure has been moved to the Supplemental Information.

> 5) Fig. 7: What is the intent of providing average vertical profiles for cases that are characterized by episodic layers (in the case of the Asian pollution) and boundary layer sources (in the case of the Central Valley)? Are the vertical profiles for the Asia pollution layers composites which include only those times in which the aircraft was in layers, or do they include time periods outside of layers? What is the value of these profiles to the discussion? I suggest that this figure be removed, or better described and used in the text.

>> The figure is a composite that includes the points at each altitude that were classified as belonging to each air mass category, as discussed in the text. There are significant differences between the different airmass categories (e.g. Asian vs Central Valley) which are of interest for the discussion, as well as for comparison with past and future studies and studies at other locations. However due to its lower importance this figure has been moved to the Supplemental Information.

> 6) Fig. 9: It is probably not necessary to show two sets of measurements of VOCs from different instruments that are in agreement. The fourth panel is hard to read, with little dynamic range on the organic and nitrate curves–the figure needs more



**ACPD**

8, S12371–S12379, 2009



vertical space. The width of the CO line covers approximately 20 ppbv on the scale used, and there is no evident enhancement in the OAL. MSA is not really used in the analysis, and clutters the graph. The curves are hard to distinguish, since they have the same thickness–those with color vision problems (˜10% of readers) would have a very difficult time discerning the curves.

>> The suggested changes have been made in the figure. The MSA trace was kept and a more explicit mention of it was made in the text; it is an important piece of evidence to show that the OAL is not of marine origin.

> 7) Fig. 10: Again add top and right axes. The caption does not describe the source of the data–all flights, or just the flight on 2006/05/01? Why does the "free troposphere" data look virtually indistinguishable from the "OAL" data? The text says the sulfate values < 1 ug sm-3 (about .25 ppbv at STP) are placed into the "free troposphere" category, yet there are clearly data in the "free troposphere" set that have sulfate levels well above this value.

>> Figure and caption adjusted as suggested. The “free troposphere” data does not necessarily exclude very old Asian pollution, so the idea that the OAL and free tropospheric data points look similar is reasonable. Basically, the OAL is the only older Asian pollution that we can positively identify as being from Asia. We thank the reviewer for spotting the anomalously large free tropospheric sulfate data points, it turns out that we had made a mistake in the averaging of high time resolution AMS sulfate data onto the lower time resolution SO2 measurements. Note that at the temperatures and pressures encountered by the C-130, 1 μg sm 3 can be as much as 0.5 ppbv.

> 8) Fig. 11: Again add top and right axes. How can the size distributions extend well above 900 nm Dva when the inlet for the AMS instruments has a very sharp cutoff at this

>> The axes have been adjusted accordingly. There is still some transmission above



**ACPD**

8, S12371–S12379, 2009

Interactive Comment



900 nm in the AMS; as a reminder, the transmission curves for the AMS and the inlet system are shown in Figure 1. There may also be some effect of slow vaporization of some larger particles that creates the appearance of more mass above a micron (i.e. the AMS transfer function is right-tailed). A note has been added to the figure caption to address this.

> 9) Fig. 12: I am particularly puzzled by the OA/CO ratios presented in the second panel. In Fig. 9, there is no evident enhancement in either organics or CO in the OAL plumes, so how can this value be calculated? Furthermore, the error bars in the first 3 panels cannot possibly encompass the uncertainty in the measurements (the AMS alone has ˜30% uncertainty), and there is undoubtedly much variability in the atmospheric concentrations of OA relative to CO and to OC relative to TOOC. (For example, see Fig. 8 of deGouw et al., 2008.)

>> Figure 5 now makes more evident the small enhancements in CO in the OAL plumes. Note that the quantity defined is OA/ΔCO, and not ΔOA/ΔCO, so a lack of enhancement in OA does not preclude calculating this quantity. We thank the reviewer for noticing the low uncertainty estimates; this was an oversight where the instrument uncertainties were not added to the variability from averaging the individual data points. This has been corrected and the different uncertainties have been added together in quadrature. While there is variability in the atmospheric concentrations of OA relative to CO, in lieu of having measurements of OA/ΔCO as a function time downwind from one single urban area, we are attempting to look at this ratio downwind of several urban source regions; this is not ideal, but the best we can do at this time.

> 10) Fig. 13: This figure is excessively detailed and is not used effectively in the discussion, and could be removed.

>> This figure has been moved to the Supplemental Information.

> 11) Fig. 14: remove figure. This information is already in Table 4.



**ACPD**

8, S12371–S12379, 2009



>> This figure has been removed.

> 12) Fig. 15: remove figure. These "example peaks" are not necessary to the discussion.

>> This figure has been moved to the Supplemental Information.

> 13) Fig. 16a: remove the panel with the 12-s measurements, since this is not informative to the model comparison. Use same scales for each of the remaining 3 graphs, and include top and right axes.

>> The 12-sec measurements provide the best information for determining the slopes. Additionally, one of the points made in this graph is to show the additional information available with high time resolution instrumentation by comparing the 12-sec measurements to the 15-min measurements. Therefore we have kept the panel with the 12-sec measurements in the revised figure. The other suggested adjustments have been made to the figure, including using only one slope to represent the Central Valley rather than a range as suggested by Reviewer #2.

> 14) Fig. 16b: as for Fig. 16b. I believe there should be one figure caption for a single figure, so this figure may need to be completely separated from 16a if a different caption is desired.

>> This has been made into a separate figure for clarity and updated similar to the previous figure.

> 15) Fig. 17. remove figure: This information is already in Fig 16b.

>> This figure has been moved to the Supplemental Information.

> 16) Table 3. Suggest splitting the "Asian Pollution" class into "YAL" and "OAL", as these are considered separately in figures, discussion, and conclusions. Suggest adding total S or SO2 to the table.

>> The suggested adjustments have been made.



**ACPD**

8, S12371–S12379, 2009

Interactive Comment



> Specific suggestions: text

> 1) Title and throughout text. There is consistent reference to "Asian" aerosols, as if this were a specific class of particles. Asia is a huge continent, with a very large diversity of aerosol sources and types, from Siberian taiga fires to the Indian biofuel "brown cloud" to Beijing sulfate/organic smog. I believe that this paper refers to aerosol particles associated with anthropogenic emissions, mostly from eastern Asia (probably mostly China). This terminology, and descriptions of the source regions, needs to be changed to be more specifically defined throughout the text.

>> A sentence has been added to the introduction to explain that for this study, the term “Asian” generally refers to emissions that originated in China / East Asia.

> 2) p. 15379, lines 17-21. These two sentences are inconsistent.

>> These sentences have been adjusted to be consistent.

> 3) p. 15380, lines 24-28. There are few experimental studies, but several modeling studies of the chemical transformations during trans-Pacific transport.

>> We have added text added to clarify that it is the limited number experimental studies that is being discussed here.

> 4) pp. 15395-6. On lines 13-15 of p. 15395, the transport of the YAL is described as occurring within a warm conveyer belt, while on lines 15-17, it is described as being processed through a "convective system". Trajectory analyses and satellite images would give strong support to the concept of wet removal of rapidly formed aerosol particles, but are not shown in the manuscript or supporting material. I recommend that they be added.

>> Two figures have been added to the manuscript, one showing relevant satellite imagery from the GOES satellite and the other showing FLEXPART trajectories for the YAL and OAL.



**ACPD**

8, S12371–S12379, 2009

Interactive Comment



> 5) pp. 15398-15400 (discussion of "older Asian pollution layer") This section describes the attribution of layers of sulfate that are not substantially enhanced in other species typically associated with anthropogenic emissions. As stated in the text, "Based on the combination of higher sulfate concentration, back trajectory modeling, and gas-phase tracer information, we conclude that this layer originated in Asia 7-10 days prior..." This statement alone is not adequate support for measurements that provide some of the critical findings of this manuscript, such as the relative dominance of sulfate over organics and the importance of cloud scavenging in controlling free tropospheric aerosol chemistry. In the example flight (Fig. 9), there are not clear enhancements in any anthropogenic signal that can be associated with the layers of enhanced sulfate. The trajectory analyses are not shown, nor are meteorological analyses that would convince the reader that these layers originated from anthropogenic sources in eastern Asia and passed through a wet scavenging event prior to detection over the eastern Pacific. There needs to be much clearer and more thorough documentation of the origin of these layers and the processes they have encountered during transport. In addition, the sulfate values reported for the OAL grouping appear to be also typical of the "free troposphere" grouping, and do not stand out in Fig. 10 (although they certainly stand out in Fig. 9). The argument is made that the OAL samples represent a dilution of ˜18x relative to the YAL samples (3%/hr dilution, 4 days). Based on Figs. 9 and 10, where the maximum total S in the OAL is approximately 1 ppbv, this dilution rate would seem to suggest a YAL total S of 18 ppbv–3x the observed value. After an extensive discussion of dilution rates, the statement is made (p. 15400, lines 10-14), "... the difference in concentrations of these two layers is (not) necessarily representative of the change in concentrations of these air masses during transpacific transport. However, if we assume that the differences in concentration are broadly consistent with the dilution of a single layer during transpacific transport, the implication is that there is a much larger production rate for sulfate than (for) organic aerosol during transpacific transport." This may be a valid conclusion, but it is not supported by the discussion of dilution rates (which are then not quantitatively used, as in



**ACPD**

8, S12371–S12379, 2009

Interactive Comment



the quoted sentences above). Furthermore, layers in the free troposphere do not dilute at a constant rate as suggested by this analysis. For example, Neuman et al. (JGR, 2006) report on layers advected above the marine boundary layer in the lower troposphere from the northeastern USA across the Atlantic with minimal dilution over time periods of a few days. In sum, evidence has not been presented that the layers identified as OAL originate from anthropogenic sources in eastern Asia, as is assumed in the subsequent analysis. This claim needs to be confirmed by meteorological analysis, transport modeling, and specific evidence from gas-phase species.

>> See response to last comment regarding additional figures that show back trajectories leading to Asian continent for OAL. Adjustments to Figure 9 as suggested by this reviewer improve ability of reader to see enhancements in CO in OAL. One of the conclusions of this study is that aerosol sulfate is a better tracer for Asian origin than gas phase tracers, thus the Asian origin of the OAL is shown with the back trajectories and confirmed by the presence of sulfate. The dilution rate discussion has been removed.

> 6) Organic mass spectra: Section 4.2.3 describes the organic mass spectra and evidence for greater shifts in fragment ions in particles found in the aged layers and free troposphere. This information presented is both qualitative and extremely detailed, and the conclusion that aged organic aerosol shows more oxidized fragment ions than does fresh is unremarkable. This section tends to detract from the main points of the manuscript, and should be eliminated or moved to supporting materials.

>> This section has been shortened to a summary and the bulk of it has been moved to the Supplemental Information.

> 7) Page 15417, lines 24-26: Has the GEOS-chem model evolved since the findings of Heald et al (2005) to produce more organic aerosol production in the free troposphere to better align with the ACE-Asia measurements, and might this explain the current over-prediction of OC?

>> Heald et al., 2005 did not include SOA from isoprene photooxidation, where the



**ACPD**

8, S12371–S12379, 2009



version used in this study did. This source approximately doubles the SOA burden (Henze and Seinfeld, GRL, L09812, doi:10.1029/2006GL025976, 2006) but would not "fix" the OA discrepancy during ACE-Asia off of the coast of Asia. There are also "research" versions of GEOS-Chem that include updated aromatic SOA sources and uptake of glyoxal/methygloxal, but these were not part of this study.

> 8) p. 15419, lines 9-11: Equally striking to the under-prediction of OA by MOZART is the over-prediction (per CO) by GEOS-Chem (Figs 16, 17), especially for the Asian layer cases.

>> What appears to be more of an issue is the under-prediction of CO by GEOS-Chem; text to highlight this has been added.

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 15375, 2008.