

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

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ACPD Manuscript ACPD-2008-0400 Response to Reviewer 2

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

Response to Reviewer #2

> This manuscript uses some of the same data and reaches some of the same conclusions as Peltier et al (2008, ACP). Several authors are on both papers. This study does acknowledge Peltier et al and Brock (2004) as providing the conceptual model but the features that distinguish this study from Peltier et al are not identified. Figure 4 of Peltier et al (2008) nicely illustrates the conceptual model for the formation of Asian aerosol layers.

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>> This study is distinct from that of Peltier et al. for the following reasons: different "core data" (AMS instead of PILS) are used, together with different classification criteria. Similar conclusions are reached although with some differences as discussed below. Our separate study adds to the weight of evidence for the proposed aerosol transport mechanisms discussed by Brock and Peltier. In addition, our study presents unique data on the evolution and chemical composition of OA, as well as comparisons to two global models, neither of which were addressed in the Peltier paper.

> Paper lacks a clear statement of how much data there is for Asian plumes. Are the 2 Younger Asian Layers shown in Fig. 9 the only cases? Are the older Asian layers in Fig. 9 the only cases. Do these plumes account for 8% of the data set? If not, what are the other Asian plume like? If the plumes in Fig. 9 are the only plumes or constitute most of the Asian pollutant layer data, then the paper should be re-done as a case study with comparison to other broad categories. I would expect a case study to provide trajectories or other transport information as well as data on substances that are tracers of specific emission sources. For some of this data the reader is referred to Supplemental material. Several measurements are mentioned as being helpful in identifying Asian out flow and then not mentioned again in the main text.

>> Yes, as already stated, "Asian plumes"; as we define them account for 8% of the total data set. Text has been added to say that this amounts to intercepting approximately a dozen layers and that the two case studies account for 17% of the Asian pollution category. Trajectory information has been added for the case studies in response to Reviewer's comments above.

> I am uncomfortable about reaching conclusions on the ratio of OA to sulfate in the Asian pollutant layers when these layers are defined as having high sulfate concentration.

>> The two case studies and the MTBE evidence clearly show that Asian pollution layers are high in sulfate relative to organics, at least for the set of Asian pollution

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layers intercepted by the C-130 during this study. Our intent has not been to portray this study as an exhaustive look at all types of Asian pollution being exported over the Pacific, but rather to show that our observations show support for the Brock et al. conceptual model for organic and sulfate aerosol transport and transformation across the Pacific. It is clear (as we state) that further research is needed, particularly near the Asian continent, to definitively understand the process(es) by which aerosol is lifted out of the boundary layer.

> Thus, the Asian vertical profiles in Fig. 7 do not resemble the vertical profiles shown in Fig. 2 of Peltier et al (2008) identified on the basis of Flexpart CO source region.

>> The differences in the vertical profiles between the Peltier et al. study and this one is driven by the different definitions of "Asian"; employed in the two studies. The Peltier et al. definition is based on FLEXPART modeling information and our definition in this study is based on chemical tracer information. The two approaches are complimentary and our approach is more restrictive, with 8% of the data being considered "Asian" compared to 48% in Peltier et al. Peltier et al. included in their averaging air masses over North America for which the Asian influence was identified using FLEXPART, but which may nevertheless have been influenced by North American emissions which would result in increased OA/sulfate ratios.

> The definition of categories is confusing and I'm not sure whether there is a distinction between Asian pollutant layers with sulfate and Asian pollution.

>> In this study, "Asian pollution" means "Asian pollution with sulfate"; this is clearly defined in the paper, including the statement: "This definition may ignore less concentrated Asian pollution layers that have lower sulfate loadings, but should be restrictive enough to exclude other types of layers."

> An area of concern is in comparing the older and younger Asian pollution layers as if they have a common source. Do the FLEXPART trajectories offer any evidence of this? I have confidence that the photochemical age determinations adequately distinguish

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between old and young Asian air masses but as long as it is available, I would expect confirmatory evidence from trajectories or Felxpart.

>> FLEXPART trajectories have now been included; see description in response to Reviewer #1 comments. The two layers do appear to originate from the same region in East Asia.

> What is unique in this study: Reasons for publishing a revised version of this paper. 1. Identification and characterization of Asian layers relying on AMS. Figs 4 and 5 indicate high quality data sets. 2. O/C ratio from high resolution mass spec. 3. OA/Delta CO 4. A potentially good case study, though not developed in this manuscript. 5. Age related changes in Fig. 12 subject to caveats that different starting conditions confuse comparison. Many good ideas are presented in this study but for the reasons given above and in the Specific comments, I believe that a much better paper could be produced after revisions.

> Specific comments > P 15392, line 20-21. Definition of Asian aerosol layers in Section 3.1. "All of these factors were used in enough test cases (see Sect 3.11 for example) to establish that elevated levels of sulfate were also indicative of Asian pollutants."; I don't know what was done and I don't know how many test cases there were or what a test case consists of.

>> The two case studies provide clear examples of what a "test case" means; the choice was made to only discuss two examples in depth in this paper, to limit the length of the paper.

> P 15393 line16-17 referring to Asian pollution levels that contain sulfate “used only data for this category above the MBL” P. 15394 line 18 “Asian aerosol in the MBL” This appears to be a contradiction. Is there a difference between Asian pollution layers that contain sulfate layers and Asian pollution? This is not evident from text.

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>> This is discussed above in response to this reviewer's comments.

> P 1593, line 22-23 The use of data west of -125 longitude excludes North American pollution layers as is confirmed by back trajectory analysis. Marine boundary layer experiments in this region show North American influence. How do you exclude the possibility that some part of the pollution may be due to North America? This is probably more serious at low altitude.

>> Figure 4 showing MTBE vs aerosol sulfate makes this point. MTBE is only found in Asian gasoline, not in that from the U.S.; if our criteria allowed North American influence on what we have declared as Asian pollution, there would be data with elevated sulfate levels that had no elevated MTBE. This is not the case. This point has been made more clearly in the text.

> P 15399, dilution rates. Isn't there an implicit assumption that the younger and older layers started out with the same organic aerosol concentration after the lifting event? How is this justified? However this rate of dilution would decrease the sulfate aerosol by significantly more than the observed decrease. Is this still true if you multiply the sulfate by 0.9/0.5 to take into account the observed conversion of SO₂ to sulfate?

>> The dilution rate discussion has been removed.

> P 15408- P 15409 Delta CO and Delta TOOC used in ratios. Are CO and TOOC concentrations given anywhere in paper? Some of this data can be read off of Fig. 9 but that gets back to the question of whether Fig. 9 contains all or most of the Asian plume data. Also, it is hard to get average values over plumes from this graph. What are the background values and how are they justified?

>> As discussed, Figure 5 (formerly 9) contains data for the two Asian layer case studies, for which OA/CO values are calculated and then used in Figure 10 (formerly 12). The revised version of Figure 5 more clearly shows the enhancement of CO in the

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OAL, and makes more obvious the background CO levels on either side of the layers. The following sentence has been added to clarify how background CO values were estimated: "background CO values were taken from average CO concentrations on either side of enhancements in Asian pollution layer case studies."

> P 15410 lines 3-8. Higher dilution rate needed to explain OA/Delta CO Without concentrations it is not possible to follow argument.

>> This paragraph has been significantly shortened in line with this reviewer's suggestions below.

> P 15414 line 24-25. Since the OA/Delta CO does not increase, this suggests that some carbon is being lost from from the aerosol in during the process; This statement results from comparing two air masses which have different histories and different sources. I don't believe that the lack of a change in OA/Delta CO has a statistical meaningful implication on carbon loss.

>> As discussed above these two airmasses appear to originate from the same region in Asia. This is already qualified as a suggestion and is consistent with findings of several other studies.

> Figure 2. Why is the transmission efficiency of AMS rounded down to 100%? Values greater than 100% are physically allowed.

>> The thought was that many readers would be confused by this. Efficiencies of > 100% have been put back in.

> Figures 3 and 4. There are SMPS peaks in Fig. 3 which are significantly higher than the AMS+BC data. I can't find these points in Fig. 4. The SPMS axis in Fig. 4 does not go to high enough values to show all of these points. Overall the agreement is quite good.

>> The SMPS peaks that are significantly higher than the AMS+BC data clearly align with changes in altitude, indicating a further problem with the improperly seated o-ring

described in the text. This text describing the SMPS has been augmented to mention this. These points were simply off-scale in Figure 3(a) and 3(b) (formerly Figure 4); this figure has been changed to show these points now. Additionally, it was discovered that the SMPS data presented in Figure 2 (formerly Figure 3) included by mistake points above 5 km, where those were known to be problematic as described in the text; those points have now been removed from the figure.

> Fig 16a caption “For the Central Valley there was no central grouping of points indicating a single slope.” It looks to me that the high slope line fits the data and the low slope lines fits a much smaller subset.

>> The two lines have been replaced by a single line that fits the higher sloped group of data points; this has been described in the figure caption.

> Length Most everything in the articles is relevant, but some things are more relevant than others. Here are some suggestions for material that I considered to be less relevant to the central message. I have not attempted to be exhaustive.

> P 15399 Dilution rates Hard to see how a single rate measured some where else, that is a factor of two higher than the INTEX rate adds to paper.

>> This discussion of dilution rates has been removed.

> P 15409 – 15410. Discussion of other mechanisms for loss of OA 1 or 2 citations plus conclusion should suffice

>> This paragraph has been significantly shortened to include only the most relevant conclusions.

> P 15411, lines 13-18 introduces the topic of organic aerosol vs. Ox and refers to supplemental material.

>> This material has been moved to its own short paragraph, the OC/TOOC discussion has been streamlined slightly.

> P15413 lines 7-14. Reasons why PMF was not used.

>> We disagree with the reviewer in this point. This is an important point to make, especially for the AMS community, as other groups are applying PMF to similar datasets and often interpreting the results erroneously.

> P 15414, line 25 – P15415 line 15. Discussion of heterogeneous oxidation. More of an overview than relevant to measurements.

>> As discussed above we prefer to keep this discussion. Although the data has limitations, it is the first published data of its kind for highly aged OA and this discussion is of high interest for the OA community. E.g. our paper has already been cited for this point by Hallquist et al. (ACP 2009) and Kroll et al. (PCCP, 2009, in press).

> Figure 1 is almost identical to Fig. 1 of Peltier and can be removed.

>> This figure has been moved to the supplemental information.

> Figure 15. High resolution spectra. If I am not mistaken, the details of these graphs are not used in the analysis: only the fact that you can add up O and C peaks. Reference to published high resolution spectra would suffice.

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

> Wording, typos, etc. > P 15394, line 26: and only in specific episodes” meaning?

>> This wording has been clarified.

> Figure 1 needs state boundaries

>> This figure was moved to the Supplemental Information at this reviewer’s suggestion.

> Fig. 2. What direction is air moving? Arrow has two heads.

>> Arrows with two heads show physical dimensions; arrows with one head each

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show directions of flow.

> Fig. 12 caption. TOOC appears in caption before it is defined.

>> Definition of TOOC has been added to figure caption.

> Fig. 14 Legend Intex-B C-130 Which layer or category?

>> This figure has been removed at the suggestion of Reviewer #1.

> Fig. 13 next to last line of caption. hydrogen (CH) should add. in the older layer.

>> This has been updated in the figure caption (now in the Supplemental Material).

> Fig. 16a There are gray data points that are not identified. Perhaps a resolution problem.

>> As stated in caption, gray data points do not fall into one of the four classified air mass types. The air mass types were intended to provide insight into the processes affecting aerosols being transported across the Pacific; they were not intended to classify all data points, e.g., air masses over the North American continent not in the Seattle or Central Valley are not classified.

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

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