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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.

Anonymous Referee #2

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Review of “Evolution of Asian aerosols during transpacific transport in INTEX-B” by Dunlea et al for Atmos. Chem. Phys. Discuss.

Measurements of aerosols observed during INTEX-B are described. The principal topic is a mechanism for creating the sulfate rich layers observed in the mid-troposphere off the west coast of North America. Evidence is presented that 1) the Asian aerosol was lifted in a warm conveyer belt type event which caused soluble pollutants to be removed 2) removal of soluble aerosol occurred after most of the SOA forming potential of the Asian air mass had been realized, and 3) there was gradual formation of aerosol sulfate due to SO2 which being only slightly soluble was not removed.

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Interactive Discussion
Discussion Paper

This manuscript is really long, 25K words compared with 15K works for Petltier et al (2008) which covers a similar subject. I will not impose my views on length but I will offer the advice that a paper half the length will be read (and cited) twice as often. In spite of the length, or perhaps because of it, this paper does not do justice to its main subject matter which is confirmatory evidence for the Brock et al (2004) conceptual model on transpacific transport of pollutants from Asia. My apologies for not reading the supplemental material. My rational is that if something is important to understanding the main theme of the paper it should be in the paper.

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.

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.

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. 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.

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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.

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

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.

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

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not evident from text.

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.

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?

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?

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

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.

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

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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.

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.

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.

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

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

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

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

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

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.

Wording, typos, etc.

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P 15394, line 26 “and only in specific episodes” meaning?

Figure 1 needs state boundaries

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

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

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

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

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

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

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