Atmos. Chem. Phys. Discuss., 8, S6390–S6396, 2008
www.atmos-chem-phys-discuss.net/8/S6390/2008/

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 #1

Received and published: 26 August 2008

General comments

This paper is an ambitious summary of a large airborne dataset containing high quality measurements of a variety of gas-phase, particulate-phase, and meteorological parameters, in addition to chemical transport model evaluation. The focus of the work–the modification of pollutants during transport from Asia to North America–is interesting and relevant to the ACP readership. It is an important subject given the growing recognition of the significance of long-range transport to climate and tropospheric chemistry. In particular, the presence or absence of a large free tropospheric source of secondary particulate organic matter is of substantial concern, since this source could have effects on the cloud-nucleating properties of free tropospheric aerosol particles and the budgets of both gas-phase and particulate-phase organics in the atmosphere.

In general, the manuscript succeeds in supporting the conclusion that cloud processing has a significant effect on the chemical composition of aerosols transported across the Pacific, and that this mechanism needs to be better quantified. However, this conclusion is not reached in a fluent and precise manner, and the manuscript needs some modification to improve clarity and conciseness.

The strategy of the authors is to separate data acquired in layers of enhanced particulate concentration in the free troposphere into 2 groups based on estimated transport time from sources on the Asian continent. These 2 groups have estimated transport times of 3-4 days ("young Asian layers", or YAL), and 7-10 days ("old Asian layers", or OAL). The characteristics of the layers are quite different, with the YAL present with substantial evidence of gas-phase pollutants, while the OAL appear to be present without clearly identifiable enhancements in anthropogenic gas-phase species. The properties of aerosol particles with ages $<$ 2 days are estimated from measurements in California's Central Valley, the regional aerosol properties around the USA city of Seattle, and from literature values from downwind of Asia and the northeastern USA. These data are discussed in the context of a quasi-Lagrangian evolution of aerosol properties, with cloud scavenging of mixed organic-sulfate particles and soluble aerosol precursor gases being necessary to explain the observations in the YAL and OAL. Essentially, the authors argue that layers of pollution arriving near the western coast of North America are quite different compositionally than those typically found immediately downwind of large sources of pollution in eastern Asia (and elsewhere), and that scavenging in cloud systems is the most likely explanation for these differences.

The argument outlined above is quite general in scope, and requires clearly described evidence and focused discussion to make the case compelling. To achieve this objective, the manuscript needs some substantial modification to improve its precision.

Specific comments: figures and tables

There are several figures that contain information that is duplicated in tables, or that is

ACPD

8, S6390–S6396, 2008

Interactive Comment

marginally informative. Some others need modification for clarity.

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

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 scatterplots and regressions in the later figures.

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.

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

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.

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 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.

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"

ACPD

8, S6390–S6396, 2008

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.

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 size?

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.)

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

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

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

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.

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.

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

ACPD

8, S6390–S6396, 2008

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.

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.

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

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

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.

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

ACPD

8, S6390–S6396, 2008

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 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.

ACPD

8, S6390–S6396, 2008

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.

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.

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?

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.

Technical comments: To follow revision of manuscript.

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