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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trans-Pacific transport and evolution of aerosols and trace gases from Asia during the INTEX-B field campaign" *by* B. Adhikary et al.

Anonymous Referee #1

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General comments:

In my view this paper should not be published in anything like its present form. My primary concern is that no significant scientific findings are presented. The paper discusses the model calculations that have been conducted, compares the model results with measurements, and reaches conclusions regarding the performance of the model. There is little here that advances our understanding of tropospheric chemistry. I suggest that the authors completely reassess what scientific findings can be gleaned from the great deal of work that they have performed, and write a paper that objectively, rigorously and clearly conveys these findings. As the paper stands, the

model-measurement comparison discussion is often quite subjective and not rigorous. It is also largely conducted without referring to previous work that can help to put the present study into perspective. Following are some of the specific shortcomings of the present manuscript.

Specific comments:

1. Paragraphs 2-5 of Section 3.0.1 discusses the distribution of CO. This discussion should be conducted from a wider perspective and in a more objective manner. Specifically:

- In Fig. 2a the tongue of CO in the upper left corner is identified as entering the domain from Europe, but no justification is given for this identification. It looks to me to simply be Asian emissions transported toward the upper left corner. I assume that the authors' interpretation is based on the MOZART-4 global model that provided top and lateral boundary conditions for this study. This should be discussed fully, and perhaps Fig. 2a modified to show the STEM model domain within the larger CO field derived from the MOZART-4 model.

- The left-hand panel of Fig. 2c is a latitudinal distribution at the same longitude and season as shown in Fig. 2b of Forster et al. (2004), who presented a transport climatology for transport of Asian CO emissions across the Pacific. However, there is no apparent similarity between these two plots. This difference should be discussed. Was 2006 an anomalous year, are there systematic differences in the underlying transport models, or is there another explanation?

- The discussion of Fig. 2b requires improvement. How can the model agree much better with the C-130 observations than the DC-8 observations, when the DC-8 is generally operating upwind (at least from a trans-Pacific perspective) of the C-130? Does this indicate that the C-130 is primarily sampling North American emissions? If so, why are these data included in a paper discussing Trans-Pacific transport? It seems to me that the model-measurement difference (20-30 ppbv) is remarkably large for the DC-8

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comparison in the lower 4 km, when one considers that the background concentration at this season is something like 90-100 ppbv, and the average modeled concentration is only about 120 ppbv. Such a remarkable difference should be investigated and discussed in detail.

2. The discussion of Fig. 5 d and e should investigate and discuss the reason that the model overestimates the magnitude and greatly overestimates the variability of jNO2. This discussion should be extended to jO3 (1D) in Fig. 7, which might be expected to be more difficult to estimate and to have larger implications for the photochemical environment.

3. The vertical profile panels of Fig. 6 need improvement, as the features are not distinguishable as presented. Perhaps log scales for the abscissa will help. It appears that the model overestimated PAN by much more than a factor of two, at least at low and high altitudes in the DC-8 profile. This should be discussed more clearly and objectively. In particular, Tang et al. 2004a (referenced in paper) present calculations with basically the same model for the same region and season. There PAN measurement and model seem to be in reasonable agreement. Why was there agreement in the earlier study, but disagreement in the present? This question requires discussion. Further there appears to be agreement between the model and the PAN measurements at Mt. Bachelor. This contrast must be fully discussed. Did the aircraft compare measurements with the surface during overflights? If so did they agree?

4. The discussion of the sulfate data in Fig. 9 is very unsatisfying. Is it possible that the observations themselves have problems? Were the instruments on the DC-8 and C-130 shown to yield comparable results? Higher sulfate at high altitudes at the downwind locations sampled by the C-130 compared to the DC-8 seems unphysical. This discussion needs a great deal of improvement.

5. In the discussion of Fig. 10 reference is made to a “second sulfate peak at similar altitudes associated with the North America plume located around 130W”. I do not

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believe that this second peak is really from North American sources, is it? If this is the assertion of this paper it must be supported by a clear discussion of the mechanism responsible for transport of North American emissions to an altitude of 3 to 7 km this far to the west of the continent. Lines 8-15 on pg. 16399 seem to return to this feature. The discussion needs to be better organized and less repetitive.

6. In the discussion of Fig. 10 it is stated: "A clear delineation between North America outflow and long range transport for (sic) Asia is seen at 130W." It should be discussed whether this North American outflow is actually present in reality, or an artifact of the model. Transport analysis usually finds that even at the surface in springtime, onshore flow dominates and that North American influence is largely absent in the large-scale flow (see e.g. Fig. 8 of Oltmans et al., 2008). There are mesoscale flow features that can transport North American emissions off shore at low altitude, but it is doubtful that the relatively coarse scale WRF model that defines the transport in this modeling study can resolve these flow features. This issue needs a full discussion. Fig. 2 does not show such a feature in CO. Is the model offshore SO2 feature perhaps due to ship emissions, rather than North American outflow?

7. At the top of page 16402 it is stated: "The ozone curtain shows strong influence of stratospheric ozone throughout the INTEX-B period." What is the justification for this statement? How is it excluded that the high O3 at higher altitudes is not transported O3 produced from anthropogenic precursors? There seems to be significant correlation with PAN.

8. In Fig. 12 it appears that the model does not give NO equal to zero at night. What is the problem that leads to this non-physical behavior?

9. In the discussion of Fig. 16 it is stated; "The results show that the model is able to capture the magnitude of the total PM2.5 mass at THD with some underprediction during the first week." To me it appears that the underprediction is throughout the period with little correlation. A more objective discussion is required.

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10. The quasi Lagrangian event discussion beginning on page 16404 is highly subjective, and incomplete. Are the authors really arguing that "the sulfate aerosol increased from 0.3 μg/m3 to 0.6 μg/m3, to 1.5 μg/m3, respectively" over a 2 day period? Is there enough SO2 to support this increase? Why is the sulfate increase on the second day 3 times as large as on the first day. Why was the sulfate so low at the start, when the air mass had already been transported for days in the marine environment. What is the mechanism responsible for the oxidation? Much clearer and complete discussion is required.

11. One paragraph in the Summary states: "The modeling results show that during the INTEX-B campaign, transport pathways of gaseous species and aerosols were different. While most of the long lived trace gas concentrations showed a north/south gradient over the Pacific, the aerosol transport pathway was mostly between 30–40N latitude." What does this mean? Do the aerosols and gases separate during transport? A clearer discussion is required.

Technical Corrections:

1. Figure 5 – The abscissa of the vertical profile graphs need to be adjusted so that the features discussed can be discerned. The panels also need letters identifying the figure panels.

2. Better quality (or larger) figures are required in many cases. Even when the image is magnified on a computer screen, many of the details are difficult or impossible to discern.

References:

Forster, C., et al. (2004), Lagrangian transport model forecasts and a transport climatology for the Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) measurement campaign, J. Geophys. Res., 109, D07S92, doi:10.1029/2003JD003589.

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Oltmans, S.J. AS. Lefohn, J.M. Harris and D.S. Shadwick (2008), Background ozone levels of air entering the west coast of the U.S. and assessment of longer-term changes, Atmos. Environ., 42, 6020–6038.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 16381, 2009.