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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.

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Notes: The reviewers' comments are included and the author responses are designated by »

Response to Reviewer 1

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In my view this paper should not be published in anything like its present form.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.

» We thank the reviewers for their careful review of our paper. Both reviewers thought that the paper would be improved by a stronger focus on the scientific findings. We have extensively modified the paper with a major reorganization, eliminating some previous sections and adding some more focused discussions. This is reflected in the new title of the paper: "A Regional Scale Modeling Analysis of Aerosol and Trace Gas Distributions Over the Eastern Pacific During the INTEX-B Field Campaign". The paper is focused on our investigation of the distributions of trace gases and aerosols over the Pacific and to estimate how anthropogenic, biomass burning and wind blown dust emissions from various geographical regions impact these distributions during the INTEX-B experiment period. We also use the model to investigate the relative contributions of distant sources of pollutants relative to more local sources on the observations at the Mt. Bachelor Observatory, which obtained measurements in conjunction with the airborne observations during INTEX-B. This is a topic of growing interest due to the changing emission patterns along the Pacific Rim. We feel that the paper presents a comprehensive comparison of the model predictions with the extensive aircraft obser-

vations. We evaluated over 30 meteorological, trace gas and aerosol components as a function of altitude and present various statistics. The discussion has been sharpened. We feel that the paper has been strengthened based on the reviewers' comments. We feel that we have addressed all of the reviewers' comments in the revision. Below we address specific comments.

Specific comments:

1. Paragraphs 2-5 of Section 3.0.1 discuss 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 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.

» We have expanded the discussion of source/region contributions in the revised paper, and have presented new figures that show how emissions from various regions impact the study domain. The statement we made about European transport is discussed and it is indeed not due to Asian emissions.

- 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?

» We have added the China-CO cross sections and added discussion of the Forster et al. (2004) analysis. We show that the transport patterns are indeed similar, but that the

Foster analysis shows lower enhancements due to their restriction on CO lifetime and 1990 emissions.

- 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 C5273 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.

» The discussion of CO has been enhanced and we address the above points. The analysis of our model results are similar to the global model comparisons during INTEX-B in that the enhancements over the Pacific are observed to be 120-140 ppbv and the models underestimate it by $\sim$10%. This is well within the uncertainties in total CO emissions estimates (anthropogenic + open biomass burning). Some of the enhancement in the C130 CO is due to the fact that some of their flights did measure NA CO in the boundary layer. We estimate the NA emissions to contribute $\sim$ 10ppb in the lower 3 km based on the observations and the model predictions. Furthermore the DC8 flights sampled along 160W which has a strong gradient in CO. We also discuss through Cross section CO plots that a small latitudinal displacement in the modeled distributions would have a significant impact on the CO predictions compared to the DC8 below 3 km. While such a displacement is not apparent from the meteorological comparison, small errors after 10 day transport could be significant.

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.

» We have added further discussion of the large predicted variability in the photolysis rates. The larger variability in the predictions is due in part to the variability in the calculated aerosol fields (due to dust, biomass aerosols and sea salt) – this is shown by the large variability in the predicted particle nitrate distributions. We have added discussion around this issue and implications of impacts on other aspects of the photochemical oxidant cycle.

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 over flights? If so did they agree?

» We have added enhanced discussion and analysis of the PAN predictions. Indeed in our previous applications in the outflow of Asia we did not see these big differences in PAN. The air being sampled in INTEX-B is much more aged than in TRACE-P. We demonstrate that the overprediction is due to the high bias in the global MOZART model used as boundary conditions, and that the SAPRC99 mechanism then added additional PAN. We present results from the global model to demonstrate this. We also discuss a recent paper that shows that the more detailed chemical mechanism overpredict PAN and that this is likely due to a yet identified loss channel. The reason for the better PAN predictions over MBO is due to the fact that regional production of

PAN plays a larger role and that the emissions over NA are better characterized then over Asia.

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.

» We have added discussion and analysis. The observed sulfate in the upper troposphere is a real feature that has been also observed more recently in the 2008 ARCTAS experiment. The model predicts an enhancement at these altitudes and locations, but significantly underestimates it. The area of enhanced sulfate has significant contributions from sources in Russia, South Asia and North Asia. These are areas with highly uncertain emissions. So this remains an open question.

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 C5274 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.

» Thank you for drawing attention to this. We have analyzed this further and it is not due to NA sources, but rather occurs in a region of dust and contributions from China, Russia, and N. and S. Asia biomass burning emissions. This has been addressed in the revised paper as has the organization of material to avoid duplication of discussion. The model sees an enhancement in this region but is underpredicted.

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?

» This has been addressed clearly in the revision. North America (NA) outflow during the INTEXB is restricted to areas east of 125W and below about 3 km. We are familiar with Oltman's paper, but indeed the NA outflow around the coast does occur and is nicely captured in the WRF simulations at this resolution during the INTEX-B time period.

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.

» Good point we have not done a specific evaluation to isolate the role of stratospheric ozone over MBO during this period. There are periods when ozone and PAN are not correlated, but more periods when they are correlated showing that long range transport in the upper troposphere during this period bring together air masses of widely different origins. This is also shown by the dust distribution correlations with ozone. We know in the dust source regions the low pressure systems bring dust up from the surface and ozone down from high altitudes. We have changed the discussion in the paper.

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?

» We have removed NO from the paper, but the non zero values at night reflect the grid spacing in the model in that the grid does contain nighttime NOx emissions.

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.

» We have eliminated from the paper the lagrangian analysis to help tighten the focus of the paper. We include the THD aerosol data in a new figure that simply shows the mean and variability and not the true time series. This clearly shows that the model underpredicts the PM2.5 by $\sim$30%.

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.

» We have taken this section out of this paper and plan to include it in another paper. It is an interesting but tricky analysis.

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.

» This statement is not clear. We present the figures together and improved our discussions.

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.

» Done

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.

» Figures have been redone.

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