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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Diagnosing recent CO emissions and springtime O_3 evolution in East Asia using coordinated ground-based observations ofO_3 and CO during the East Asian Regional Experiment (EAREX)2005 campaign" *by* H. Tanimoto et al.

H. Tanimoto et al.

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We thank the reviewer for positive comments and valuable suggestions. Please see our detailed responses below.

Comment 1: The use of delta-O3/delta-CO ratios to identify O3 production from continental emissions was introduced in previous studies (e.g., Parrish et al., Science, 1993). The enhancement ratio studies of Parrish et al. focused on ozone formation in continental outflow where the air mass origin was largely the same, so that CO changes

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could be attributed to local emissions and O3 changes to subsequent chemistry. This may hold for the sites close to Gosan used here, but not for remote sites such as MNM which are dominated by air from marine region...

Reply: Parrish et al. (1993) and (1998) papers are now cited. Although we generally agree with the reviewer's comments, there are a couple of points to argue.

(1) We would like to note that Parrish et al. (1998) discussed springtime O3 production from North American anthropogenic precursors by using O3-CO correlation at Azores, a remote island far away from the U.S. east coast. The distance from the N. American continent to Azores is comparable to that from the Asian continent to MNM. We think that discussions using O3-CO correlation are still useful at such sites under prevailing wind conditions, if we carefully choose transport episodes, while we do agree that the air masses are not completely the same, but largely the same.

(2) We think that the air masses observed at individual sites are largely the same from GSN to MNM when we focus on pollution events I, II, and III. In other words, we select these events so that we can track the air masses originated over the Asian continent and transported by cold fronts to MNM in the boundary layer. The air masses are, of course, subject to dilution with maritime air masses during transport over the time-scale of days, but these effects can be canceled by taking ratios, if the dilution ratio is the same for both O3 and CO. As the reviewer suggested, the dilution ratios for O3 and CO could be different, because of faster depletion of O3 in maritime air masses. This point could be uncertainty in our discussion.

We completely agree with the reviewer on his/her point to more clearly distinguish O3-CO relationships between episodic and mean transport, since they are not exactly the same things to discuss. This point was confusing in the original manuscript submitted to Atmos. Chem. Phys. Discuss. In this revised paper, we define different terminology for these diagnostics; delta-O3/delta-CO for high episodes and O3/CO for monthly means. These changes are reflected in text and Figures 7, 8, and 9.

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We still think that "major" factor driving delta-O3/delta-CO (not O3/CO) for the "events" I, II, and III is chemistry. However, we are not able to completely exclude the impacts of meteorology, especially at MNM. Since this could be uncertainty in quantitative estimates, we reduce tone about en-route photochemical O3 formation, and rephrase several sentences accordingly (Abstract, Concluding Remarks). For monthly mean O3/CO ratios, we add the importance of meteorological factors in driving O3/CO ratios and distributions.

Comment 2: The justification for not looking at these earlier events is poor, as model problems are alluded to but are not addressed specifically ("complicated transport mechanisms", "unknown local sources").

Reply: The sentences in the original manuscript were somewhat misleading. The main reason why we do not look at the high-CO events observed at Gosan during 25-27 March (DOY 84-86) and 31 March-2 April (DOY 90-92) is not the model's problems. This is because these events are not typical cases for pollution transport associated with cold fronts, and are different from the events I, II, and III. We replace rewrite this paragraph in section 4.1

Comment 3: The problems associated with under and overestimation of CO when modeling plumes have been examined with a range of models at different resolutions by Kiley et al., JGR, 2003.

Reply: Thanks for this information. Kiley et al. (2003) paper is now cited.

Comment 4: How are the enhancement ratios calculated? Are they derived from episode conditions only or from some baseline?

Reply: The delta-O3 and delta-CO values are calculated as "peak height" (additional enhancement above the baseline levels around the peak). One sentence is now added (section 6.1).

Comment 5: "Having eliminated the influence of dilution...". Unfortunately the effects

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of dilution have not been eliminated here because the background air is not constant, and therefore the increased enhancement ratio cannot be attributed to chemistry alone. Separating the contributions of transport, mixing and chemistry would require a more detailed analysis than is made here...

Reply: As described in Reply to Comment 1, we still think that the major contributing factor is chemistry for delta-O3 in "individual plume". However, we rephrase the sentences since the analysis presented here has uncertainty and we do neither "eliminate" nor quantify other effects, as the reviewer mentioned.

Comment 6: The tight correlation at MNM is likely to be driven by differences in air mass origin, not directly by aging, although clearly lower peak CO is affected by mixing during the longer transport time. If the measurement-based enhancement ratios are derived from the episodes alone then they are not directly comparable with these origin driven mean O3-CO ratios.

Reply: We see that the discussion on the enhancements of O3 and CO was confusing in the original manuscript, since the enhancements were discussed on both "event" basis and "monthly" basis. The baselines are subtracted for individual peaks for event-based ratios, but not for monthly-based ratios. In this revised version, we rephrase delta-O3/delta-CO (event basis) (section 6.1, Figure 7) and O3/CO (monthly basis) (section 6.2, Figures 8 and 9) to clearly distinguish the differences.

Comment 7: Figure 9 is new and interesting, but needs better interpretation in the text here. The main outflow regions are dominated by low enhancement ratios, and high ratios clearly pick out meteorological features (e.g., the steep gradient towards MNM).

Reply: We agree with the reviewer about interpretation for monthly O3/CO ratios. We add a sentence to explain the importance of meteorological factors.

Comment 8: Rephrase this last sentence: "en-route photochemical O3 formation" is not needed to make Asia an important source of O3: source-region formation may be

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sufficient, but this paper does not quantify this. (also in abstract)

Reply: We add the word "events" for en-route photochemical O3 formation. We keep this sentence but reduce the tone to say that "this feature is suggestive" (Abstract and Concluding Remarks)

Comment 9: Figure 8: for a cleaner comparison between the observed and modeled data it would be better to use 3-hourly observational data in place of hourly data.

Reply: Figure 3 and Figure 8 are modified to compare 3-hourly data.

Comment 10: Technical Corrections

Reply: All typos and technical errors are now corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 3525, 2008.