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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.

Anonymous Referee #2

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This paper describes the use of ground-based measurements of CO and O3 and regional chemical transport model simulations to estimate recent changes in emissions of CO from East Asia between 2001 and 2005. The study provides additional independent evidence of increasing emissions in China, and these are shown to agree well with previous studies applying different approaches. The study also explores ozone formation occurring during transport out of the region by examining observed and modeled

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ratios of O3 and CO enhancements. The paper makes a very useful contribution to the growing body of literature demonstrating emission changes over East Asia and exploring their global impacts. The first section on CO emissions is very strong, but I have a number of reservations about the O3:CO ratio analysis and these are outlined in my comments below. Overall I believe that the paper is suitable for publication once these issues have been addressed.

General Comments

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), and these studies should be cited here. 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 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 regions. The CO and O3 enhancements derived at these locations are heavily influenced by air mass origin and mixing, and it is therefore difficult to draw conclusions about Asian emissions or chemistry. The delta-O3/delta-CO ratio at these locations may be overestimated because this aged tropical marine air is more greatly depleted in O3 than CO because of its shorter lifetime. This effect influences the results shown in Figure 7, where the regression is strongly dominated by the ratios at a single site (MNM). The authors need to take these effects into account in their discussion, and to be clear in both how their enhancement ratios are calculated and what they indicate when they are addressing episodic and mean transport.

Specific Comments

p.3536 l.15-23: 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"). This weakens the paper as a whole. I

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recommend that this paragraph be rewritten either with a clearer and more specific explanation of the problems encountered, or with a general statement about why the meteorological conditions at these times was less appropriate for analysis.

p.3537 l.26-28: 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.

p.3540 l.21: How are the enhancement ratios calculated? Are they derived from episode conditions only or from some baseline? One sentence of explanation is required here.

p.3541 l.27: "Having eliminated the influence of dilution...". Unfortunately the effects 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. This should be acknowledged by replacing "eliminated" with "minimized" and by indicating that the ratios are only suggestive of net ozone formation (they do not "clearly point to"), as the influence of other processes cannot be adequately quantified here.

p.3543 l.3-6: 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.

p.3543 l.10: 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). This suggests that high ratios are driven by background air mass origin and not chemistry, contrary to the argument in section 6.2. The "overestimate" (l.16) may

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therefore reflect the boundary conditions and not processing in continental outflow.

p.3545 l.25: 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 sufficient, but this paper does not quantify this. (also in abstract)

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.

Technical Corrections

p.3529 l.1: add "the" after "improve"

p.3532 l.4: rephrase "basically based on the identical" with "based on the same" or "based on a similar"

p.3538 l.3: replace "constrains" with "constraints"

p.3543 l.10: replace "A geographical distribution the" with "The geographical distribution of the"

Figs 2,3 captions: replace "unites" with "units"

Fig 9 caption: replace "white out" with "shown in white"

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