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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of anthropogenic emissions of carbon monoxide in East Asia derived from observations of atmospheric radon-222 over the Western North Pacific" *by* A. Wada et al.

Anonymous Referee #2

Received and published: 4 August 2012

General Comments:

This manuscript analyzes 2007-2011 observations of CO and Radon at 3 sites in the Western North Pacific that often sample Asian pollution outflow events. The analysis focuses on periods of enhanced Rn to select for observations that were influenced by continental sources. The atmospheric enhancements of CO/Rn are compared with global chemical-transport model simulations of this ratio for the same time period. The differences between the observed and modeled enhancement ratios are used to adjust the anthropogenic emissions of CO for several East Asian countries. The results are

then compared to other emissions estimates for East Asia from bottom-up and inverse methods.

This work is relevant and new. The Rn tracer methodology has been demonstrated in other regions, but this manuscript is novel in its application of the approach to East Asia, where emissions are large and recent changes have been rapid. There are significant inconsistencies between official and scientific inventories for East Asia, as well as differences in these bottom-up datasets and top-down estimates inferred from observations. This manuscript follows from recent previous work of these authors, particularly the 2011 paper by Wada et al. in Atmospheric Environment. In my opinion, the current manuscript adds significant new analyses in its use of the Rn tracer methodology and the application of a chemical-transport model to interpret the observations. The writing overall is clear (although there are numerous English errors, as noted below) and the figures are good.

Some changes are needed to make this manuscript suitable for publication. While my specific recommendations are detailed below, some general issues include the following:

- More details of the methods and analysis should be included. The authors too often cite the Wada et al., 2011 paper without further explanation, but this manuscript must stand on its own without the need to read the earlier paper to understand the results.

- The authors must be more careful to qualify statements about the causes of specific events in their observations without further supporting evidence. In some cases, the Wada 2011 paper could help to provide such support.

- Some of the assumptions and simplifications in the analysis need better justification.

- Error and sensitivity analyses are critical to understand how robust the results are.

- While the writing is reasonably clear, there are many grammatical and other English errors. I've listed some of them below in the Technical Corrections, but there are too

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many for me to note here. I suggest that the manuscript undergo a thorough review by a native English speaker before final submission.

********** Specific Comments:

Section 2.1: Mention Wada et al. 2011 observations and trajectory model results that support the influence of Asian outflow events on these stations, as well as the lack of significant local sources at MNM and YON.

Section 2.2: Define the explicit measurement frequency of the Rn instrument. The hourly means are presumably averages of more frequently measured data.

p. 15342, line 12-13. State the Rn measurement period at the RYO station.

Section 2.3: STAG is run on a global domain, but this is never stated explicitly. Add this to the description of the model here.

Section 2.3: Biomass burning is not included in the model emissions. Are biomass burning events also excluded from the observational record? If so, how are these events discriminated from those that are purely anthropogenic? If not, can you quantify the possible impact of biomass burning on the observational data set and the results?

Section 2.3: CO produced photochemically from hydrocarbon oxidation is treated as a constant value in the model analysis, presumably to simplify the modeling. The amount of CO present downwind will depend on what sources influenced the sampled air masses and the transit time before sampling, and there will surely be seasonal differences in this photochemistry. What is the impact of this simplifying assumption in the modeling? Since a chemical-transport model is used here, why not calculate the photochemical CO production, instead of assuming a constant amount?

Section 3.1: Much of the findings in this section are similar to the findings of Wada et al 2011; this should be noted here.

p. 15344, line 11-14: This line should say "...are usually caused by...". It is too strong

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of a statement to say that every high CO concentration observed at these stations are caused by transport from the Asian continent.

p. 15344, line 28: Add “likely” before “influenced”. This assertion has not been proven with the evidence provided up to this point. You could cite the previous results of Wada et al. 2011 as support here.

p. 15345, line 3-6: Again, these conclusions are presented without proof that the proposed physical mechanisms actually produced the observed behavior. Reword these sentences to be less strong: “This behavior is consistent with the diurnal. . .” and “These results support a strong local. . .”.

p. 15345, line 10: See comments above. Reword as “These peaks are consistent with the transport. . .”.

p. 15345, line 24-25: Simply citing Wada et al. 2011 is not enough here to understand how the residual Rn was derived. Briefly describe the curve fitting procedure, etc.

Section 3.3: Once a ERN event is identified by analysis of delta(Rn), is it actually necessary to derive enhancements in CO? The correlation of absolute concentrations of CO vs Rn data for just the points within each Asian plume should have the same slope as the correlations of the enhancements of each species.

Figure 5. It would be useful to note the slopes for each of the lines, or else plot all panels on the same scale. This way the relative slopes at each station could be compared.

p. 15346, line 14-16. The photochemical production of CO from hydrocarbons should also vary with transport time. This would be most significant for the far-downwind MNM station. How do you know that different photochemical conditions do not contribute to the enhancement ratios?

p. 15349, line 14-16. Transport errors could still be an issue, because Rn and CO sources have different spatial distributions. Can you put a constraint on the impacts of transport errors on your analysis?

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p. 15351, line 15-17. State here the original inventory estimates for these regions, and note the relative changes represented by the new estimates.

Results section. No explicit uncertainty estimates on the observations-derived emissions estimates are given. The sensitivity to the assumptions used in this analysis, such as variability in photochemical CO production and transport errors, is not tested. Error ranges for the derived emissions should be quantified and discussed here.

********** Technical Corrections:

Just a few of the grammatical and other typographical errors are mentioned here. Many more errors need to be corrected before publication.

p. 15339, line 21. Begin sentence with “The”

p. 15339, line 23. Add “and” before “chlorofluourocarbons”, which should be lower case.

p. 15340, line 1. Begin sentence with “The”

p. 15340, line 4. Change to “Our study is the first to utilize the radon tracer method. . .”

p. 15341, line 18. Change to “. . .YON is situated immediately downwind of the. . .”

p. 15341, line 20. Change to “. . .is small, lightly populated, and has little traffic, . . .”

p. 15341, line 22. Add “the” before “northeastern”

p. 15342, line 7. Define “PIN”

p. 15342, line 20. Change “has been” to “was”.

p. 15346, line 13. Start new paragraph at sentence that begins “Delta(CO)/delta(Rn) obtained. . .”.

p. 15347, line 22. Change “special” to “spatial”

p. 15348, line 10. Change “slightly lower” to “lower”

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p. 15348, line 14. Change “underestunated” to “underestimated”.

p. 15348, line 23. Add “from the regions detected” before “at the stations”.

p. 15349, line 4-5. Change to “We applied the method to our sites in the Western North Pacific region.”

p. 15350, line 15-16. Change to “Other peaks were not well reproduced, probably because these events were relatively small.”

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 15337, 2012.