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## ***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.**

**A. Wada et al.**

a-wada@met.kishou.go.jp

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We would like to thank the reviewer for valuable and constructive comments and suggestions. Incorporating them into the paper has resulted in a significant improvement in the paper. We confirm that all of the co-authors have concurred with this revised version of the manuscript. Our responses to the comments of Reviewer #1 are described below in details.

For General comments: Anthropogenic emissions of carbon monoxide and other trace

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species have grown tremendously in eastern Asia during the past decades. For researchers who establish emissions databases for modeling purposes this growth is hard to keep up with, and often the primary information about polluting activities and emission factors is incomplete. Therefore, the development of alternative activities and methods to estimate the emissions, as offered by Wada et al., is very welcome. The present study uses measurements of CO and Rn-222, the latter being a tracer of continental air, to locate the CO emissions from the East Asian continent. The measurements were performed on three islands at different locations in the western Pacific, and the results are accompanied with a tagged-tracer study with an atmospheric transport model. This work and methodologies used are appropriate for ACP, and the results are interesting. I support publication of this work in ACP though have some important reservations which could be met in a revised version of the manuscript. Although the English language use is generally good, the manuscript would benefit from corrections by a native speaker. My main concern is the treatment (or better neglect) of biomass burning emissions, which are known to be an important CO source worldwide. For example, table 1 presents the results of a number of studies, of which some do and others do not include biomass burning, which seems to have overall little effect on the estimated emissions, which range from 118 to 206 Tg/yr from East Asia. Wada et al. also assume that biomass burning emissions are of minor relevance. However, I find it hard to believe that at least in some seasons, e.g., due to wild fires during periods of dryness and agricultural waste burning, biomass burning emissions can be fully neglected. Therefore, I would like to see this assumption either corrected or substantiated by stronger arguments.

Ans. In the previous studies listed in Table 1, Palmer et al. (2003) derived an estimate of 12 TgCO/yr-1 for the biomass burning emission in China, which is 6.7 % of the total emission of FF, BF, and BB. A similar percentage value of 7.9% was reported in Heald et al. (2004) for the biomass burning emission in China based on their inverse modeling. These studies support the relative minor influence of biomass burning emissions in China. However, in addressing the issue of CO emission from biomass

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burning in the context of our study, and its effect on the measurements at MNM during the winter, we carried out an additional CO simulation by using biomass burning emission (GFED ver. 3.1 from 2005 to 2010) as a driver for the CO field in STAG. Simulated CO values at MNM from November to April showed a steady “background” concentration of 7 - 8 ppb, with a couple of noticeable peaks greater than 20 ppb, one on 26 April 2008 (Fig.1) and another one on 29 March 2010. In our analysis, these two peaks were not included in our estimation because of low correlation between Rn and CO resulting from the fact that they likely have different source regions. Thus our peak selection method excludes biomass burning effect and efficiently extracted enhanced events related to anthropogenic sources. This revised manuscript has been corrected grammatical errors and awkward expressions by a native English speaker.

Specific comments: 1. p.15339, l.28/29 mentions that the radium-226 distribution and Rn-222 emissions are uniform/homogeneous. Although this assumption is reasonable, it is factually not correct. Please discuss this as a source of uncertainty in the applied method.

Ans. In response to this comment by the reviewer, we conducted an additional simulation of  $^{222}\text{Rn}$  by using the simulated results of the  $^{222}\text{Rn}$  flux density published by Hirao et al. (2010) for the region 60E - 150E, 20N - 70N, 1x1 from 2007 to 2010. The  $^{222}\text{Rn}$  flux density of other areas was set to be the same value as the original simulation and applied the same estimation method. When compared with the estimated results in our study derived by using the constant  $^{222}\text{Rn}$  flux, the difference came out to be 3.2 % for estimated emission of CO in China. We added this discussion in the revised text.

2. p.15340, l.19: replace “observed” by “reflected” Ans. It has been corrected.

3. p.14341: Sampling sites. To what extent is RYO affected by emissions from Taiwan? Can these simply be neglected? It would be useful to characterize the sampling sites based on an air mass back-trajectory study. One could, for example, plot the origin

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of back-trajectories on a map for the three stations, color-coded for different transport periods (e.g., from 1 to 5 days). Ans. In a winter season, backward trajectories for RYO pass mostly over the northeast part of Japan and China, as shown in Figure 2.

4. p.15342, l.25: please indicate what is meant by STAG Ans. Full spelling has been added in the text. The STAG stands for Simulator of Trace Atmospheric constituent on a Global scale.

5. p.15343, l.17: biomass burning not included (see above general comment) Ans. Please see our response given above to the reviewer's general comment of not including biomass burning.

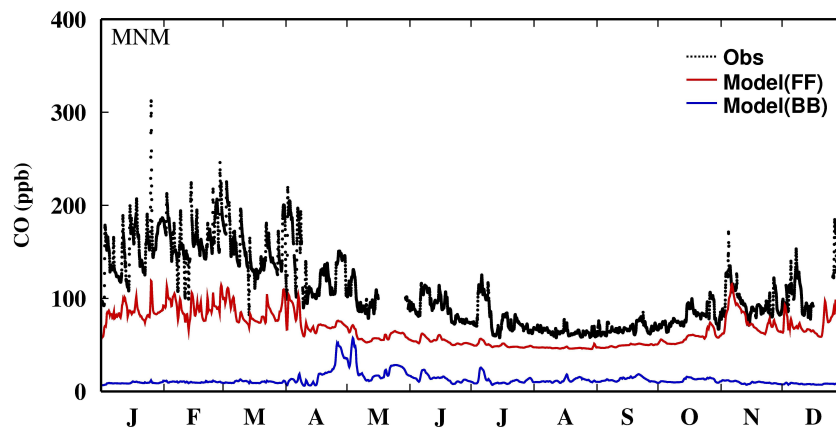
6. same page, l.19. Assuming that the contribution by NMHCs is constantly 40 ppbv is challenging. Why not relate this to the seasonal Spivakovski et al (2000) OH concentrations, as also done for CO oxidation? Ans. The contribution from NMHCs produces a nearly background level at MNM because the rate constants of NMHCs with OH radical are larger than that of CO by one or more orders of magnitude. In addition MNM is located 2000 km from any source regions. Liang et al. (2004) showed almost constant contribution of CO concentration from NMHCs in their simulation.

7. p.15350, l.18-21: Based on the tracer study by Sawa et al. (2007) it is assumed that biomass burning is a negligible source of CO. This needs to be substantiated. For example, you could use the publicly available GFED biomass burning emissions database (based on satellite observations) to show to what extent these emissions can be neglected in East Asia. Ans. We have carried out an additional experiment of CO simulation by using biomass burning emission. Please see our answer to the general comments.

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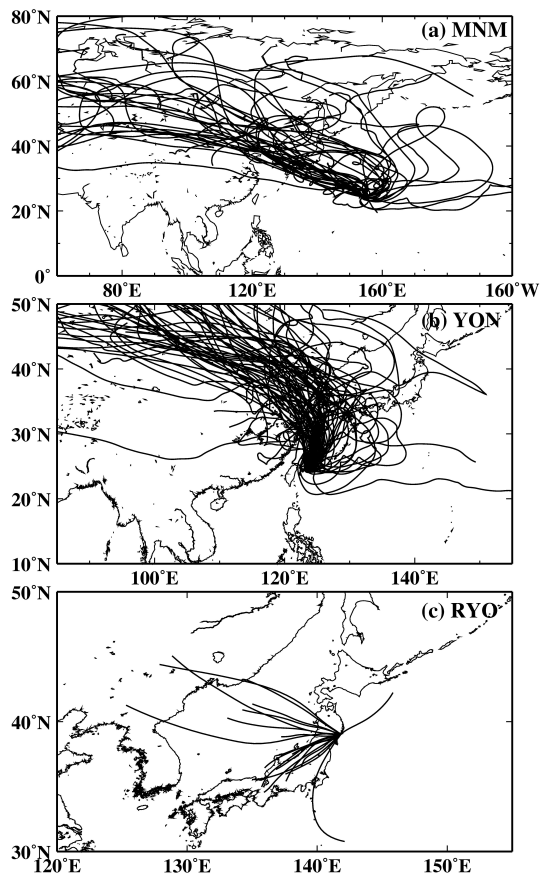
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**Fig. 1.** Fig.1. Time series of observed CO (dots), simulated CO by using anthropogenic emission of EDGAR ver. 4.1 (red), and simulated CO by using biomass burning emission of GFED ver. 3.1 (blue) in 2008.

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**Fig. 2.** Fig.2. Backward trajectory in winter season. 10-day trajectory for MNM and YON, and 1-day trajectory for RYO were shown during enhanced Rn peaks.