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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Long-term observations of black carbon mass concentrations at Fukue Island, western Japan, during 2009–2015: Constraining wet removal rates and emission strengths from East Asia" *by* Yugo Kanaya et al.

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Response to the Reviewer #1

We appreciate the reviewer's careful reading and comments on our manuscript. Detailed point-by-point responses are given below.

1) Page 1, line 23: Is the CO mixing ratio under standard conditions? Why not convert mixing ratio to mass concentration? This would make it easier for direct comparison with emission inventories.

The observed CO concentration is presented in volume mixing ratio. The CO emission rate (from emission inventory) is rather converted to equivalent mixing ratios under 273 K and 1013 hPa (the condition was commonly used for BC mass concentrations). This will be mentioned clearly in the revised manuscript. The same units were generally used in previous papers discussing observed ΔBC/ΔCO ratios in comparison to emission inventory.

2) Page 2, Line 10: maybe briefly mention the health, air quality effect of BC here.

In the revised manuscript, we will include the sentence below: Besides the relevance to climate change, World Health Organization warns the health effects of BC (Janssen et al., 2012).

3) Page 2, Line 23: why are downwind measurements important for constraining emissions? One may argue that measurements made in the source region can be even more useful.

The previous sentence will be modified to that below: Besides observations within the source areas, more observations from regions downwind of the source areas are needed to elucidate regional features of the atmospheric status and then to constrain the emission and removal rates, to better characterize the effects on the climate and health and establish an effective mitigation strategy.

4) Page 2, last paragraph: some of the discussion on measurement technique may be moved to other sections, for example, section 2.

As suggested, a part of the paragraph describing performance of COSMOS and MAAP will be moved to Section 2. In Introduction, we will just mention as follows:

In observations of BC mass concentrations in the atmosphere, the reliability of the instrument used is important for robust analyses. We regard single-particle soot photometer (SP2; Droplet Measurement Technologies, Boulder, CO, USA) and ECOC analyzers with optical corrections as reliable, but their use for long-term observations is challenging. Among filter-based techniques, more suitable for long-term observations, continuous soot-monitoring systems (COSMOS or BCM3130; Kanomax, Osaka, Japan) and multi-angle absorption photometers (MAAP; Model 5012, Thermo Scientific, Waltham, MA, USA) are satisfactory because the effects of co-existing scattering particles are minimized. For COSMOS, this is achieved by using a pre-heater to remove nonrefractive species (Miyazaki et al., 2008). For MAAP, multi-angle observations with respect to the particle-laden filter are made to take account of the scattering effect in the radiative transfer calculation (Petzold et al., 2002). The performances of the two instruments were certified against SP2 and ECOC analyzers as detailed in Section 2. For filter-based techniques, using a size cutoff device (PM1 or at least PM2.5) is important for minimizing interference from co-existing light-absorbing particles such as mineral dust.

The description of CAWNET and observations in Jeju and at Lulin station is kept in Introduction but moved to the next paragraph.

5) Page 4, Line 8: have the authors looked into some other emission inventories for comparison?

Although still focusing mainly on REAS version 2, we will mention comparisons with other emission inventories, CAPSS (Clean Air Policy Support System) for Korea and MEIC (Multi-resolution Emission Inventory for China) for China, in section 3.2.1.2 of the revised manuscript. For Korea, CAPSS had an even higher BC/CO emission ratio (39.2 ng $m{-}3$ $ppb{-}1$) than REAS2 (23.2 ng $m{-}3$ $ppb{-}1$), and the gap with the observation was larger. MEIC for China had values 9.5 and 9.9 ng $m{-}3$ $ppb{-}1$ for N-CEC and S-CEC, which were similarly higher than observations (5.3 and 6.4 ng $m{-}3$ $ppb{-}1$) as the case of REAS2 (8.3 and 9.9 ng $m{-}3$ $ppb{-}1$).

6) Page 4, Line 25: how does a change in the size-cut affect measurement results? The change affected the BC mass concentrations by only less than 2%, as estimated from size distribution of BC particles measured with the SP2 instrument during spring

2015 at Fukue Island (Miyakawa et al., in preparation, 2016).

7) Page 5, Line 24: 2500 m seems to be a bit high, if the purpose is to investigate emissions from the source region. Some of the trajectories may not come close to the surface at all.

Among the selected data, fractions entering below 2000 m were large, >82% for all source regions (except for area I (NE-China), 64%). Here we just intended to screen out the cases of clear descent from the free troposphere. Another criterion that ΔCO > 20 ppb, used together with the altitude criterion, helped to select cases with real influence from emissions.

8) Page 5, Line 34: how was APT calculated, and what is the source of the precipitation data? Also since some precipitation is associated with relatively small-scale processes and strong vertical motion, how reliable are trajectories when precipitation occurs?

The source of the precipitation data (in mm/h) was GDAS1 three-dimensional meteorological field data, and the precipitation rate along the trajectory was integrated over 72 hours. This will be mentioned clearly in the revised manuscript. We found that the dependence of CO mixing ratios on air mass origin areas was almost unchanged with the presence of precipitation as shown in Fig. 1 below. The CO median mixing ratio for S-CEC with APT >= 5mm was slightly larger than the other APT cases; however, this is within the variation ranges and cannot be attributed to the influence from other air masses (with smaller values). The data numbers for S-China for three groups were small (N= 9 or 10). From this analysis, the source area information was thought to be retained, even with precipitation, whose amount was not generally very large in our study.

In text of the revised manuscript, we will include the following sentences: When precipitation occurred, trajectories might become less reliable. Nonetheless, we found that the dependence of CO mixing ratios on air mass origin areas was almost unchanged with the presence of precipitation. Therefore, origin area information was used for further analysis of wet removal.

The figure will be included in the supplementary material.

9) Page 8, Line 7: are the two ratios for Cape Hedo significantly different?

This analysis was from Verma et al. (2011) and there statistical difference was not studied.

12) Page 9, Line 14, the REAS2 Korean BC/CO ratio is greater than that for the domestic sector?

10) Page 8, Line 13: It is interesting (and surprising) that Korea has a higher BC/CO ratio than China, given my impression that Korea is in a more advanced stage of economic development than China. Any reason why?

15) Page 15, Line 3, any measurements in Korea that may shed light on the BC/CO emission ratio from that country?

The BC/CO emission ratio for domestic sector (15 ng m^{-3} ppb^{-1}) mentioned in the previous manuscript was for China, not Korea. The BC/CO emission ratio for domestic sector in Korea was estimated to be rather low, 2.8 or 4.1 ng m^{-3} ppb^{-1} for REAS2 and CAPSS, respectively. The sectors that raised the BC/CO emission ratio in Korea were industry and transportation (42 and 27 ng m^{-3} ppb^{-1} for REAS2 and 357 and 29.5 ng m^{-3} ppb^{-1} for CAPSS). In the revised manuscript, we cite past two studies on short measurements in Korea, Sahu et al. (2009) reporting the ΔBC/ΔCO ratio from Korean Peninsula of 8.5 ng m^{-3} ppb^{-1} and Park et al. (2005) reporting 4.2–6.2 ng m^{-3} ppb^{-1} measured in Gwangju city, Korea. Both supported our observed values for Korea rather than those from emission inventories. The discrepancies indicate inappropriateness of the assumed high BC/CO ratio for industry and transportation sectors. This point will be included in the revised manuscript. One sentence in Abstract of the previous manuscript falsely indicated that high emission ratio in Korea was influenced by domestic sector. In the revised manuscript, this sentence will be rewritten as follows:

The estimated emission ratios (5.2–6.9 ng m^{-3} ppb^{-1}) varied over the six air mass origin areas; the higher ratios for South Central East China (30–35°N) than for North Central East China (35–40°N) indicated the relative importance of domestic emissions and/or biomass burning sectors.

11) Page 9, Lines 7-11. Northern China may have more centralized space heating that uses relatively large, more efficient boilers with smaller BC emission factors (compared with southern China).

Upon suggestion, we confirmed the prevalence of central heating systems in N-CEC rather than in S-CEC in China Statistical Yearbook 2014. In the revised manuscript, we will mention that prevalence of central heating in N-CEC than S-CEC (China Statistical Yearbook, 2014) might be a cause.

13) Page 13, Line 6, Figure 8b should be Figure 8a? Gray squares are not very easy to see in the figure. May consider using a different color. Correction is made (Figure 8a). Gray squares will be changed to light blue.

14) Page 14, Line 18, is the decreasing trend for Japan statistically significant? Upon comment by Reviewer #2, Figure 12 and discussion on the long-term trend using corrected BC mass concentrations will be removed, as the uncertainty is not small enough.

16) Figure 1: may consider using inventory BC/CO emission ratio for the map.

Although considered, we concluded that BC emission rates were better to show here, for readers to get impression of source regions first. In the supplementary material we will show the map of the BC/CO emission ratio.

Again we thank the reviewer for providing important comments that improved the manuscript.

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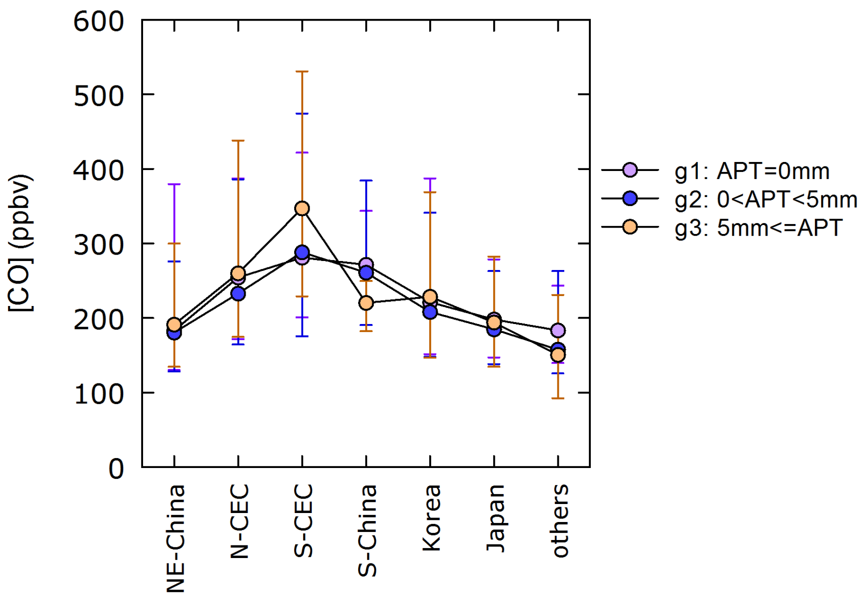

Fig. 1. Median and the 10–90 percentile ranges of the CO mixing ratios for individual air mass origin areas. The data were categorized into three groups, with APT = 0 mm, 0–5 mm, and >= 5mm.