



# Supplement of

## 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

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#### **Supplementary material**

#### 1. Dependence of CO mixing ratios on air mass origin areas insensitive to the precipitation amount

When precipitation occurred, trajectories might become less reliable. Nonetheless, we found that the dependence of CO mixing ratios on air mass source areas was almost unchanged with the presence of precipitation. Therefore, source area information was deemed still usable for further analysis of wet removal rates.



Figure S1. 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 accumulated precipitation along trajectories (APT) = 0 mm, 0-5 mm, and > 5mm.

#### 2. Difference between the frequency distributions of the observed $\Delta BC/\Delta CO$ ratios

The observed average ratio for area II [N-CEC,  $5.3 \pm 2.1 (1\sigma)$  ng m<sup>-3</sup> ppb<sup>-1</sup>] was smaller than that for area III (S-CEC,  $6.4 \pm 2.2$  ng m<sup>-3</sup> ppb<sup>-1</sup>), as discussed in Sect. 3.2.1.2. Figure S2 shows the difference between the frequency distributions of the observed  $\Delta$ BC/ $\Delta$ CO ratios for the two air mass types (black and red lines). A significant difference between the distributions is observed, particularly for the data with a  $\Delta$ BC/ $\Delta$ CO ratio smaller than 4 ng m<sup>-3</sup> ppb<sup>-1</sup>. The difference was therefore statistically significant (p < 0.01) when Welch's t-test was applied to the two data sets. Similarly, the distribution for type V' (Korea only), shown in blue, was different from that for type II, with a statistical significance (p < 0.01), mainly because the fraction of data with a  $\Delta$ BC/ $\Delta$ CO ratio higher than 8 ng m<sup>-3</sup> ppb<sup>-1</sup> was larger.



Figure S2: Cumulative frequency (%) of observed ΔBC/ΔCO ratios for selected air mass types (II, III, and V'). Only cases without precipitation are used.

### 3. Geographical distribution of BC/CO emission ratio in REAS2

In section 3.2.1.2, the observed  $\Delta BC/\Delta CO$  ratios were compared with the area-averaged BC/CO emission ratios from REAS2 inventory. The geographical distribution of the BC/CO emission ratios from REAS2 inventory for January 2008 (with a  $0.25^{\circ} \times 0.25^{\circ}$  resolution) is depicted in Figure S3, together with the borderlines used to classify air mass origin types, as supplement information.



Figure S3. Geographical distribution of the BC/CO emission ratios (in ng m<sup>-3</sup> ppb<sup>-1</sup>) from REAS2 inventory for January 2008.