Supporting Information of

Alteration of the size distributions and mixing states of black carbon through transport in the boundary layer in East Asia

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S1. Determination of the background mixing ratio of carbon monoxide (CO)

We assume the 5th percentile value of CO mixing ratio (138 ppb) as a threshold value to extract its background level (CO_{bg}). CO_{bg} is defined as the average of CO mixing ratios below the 5th percentile in this study, and is calculated to be 120 ppb. When we change the threshold from 5th to 10th percentiles (146 ppb), derived CO_{bg} increases from 120 ppb to 131 ppb. Figure S1 depicts the probability density function of the observed CO mixing ratio with the assumed threshold. It is suggested that the assumption of the threshold value slightly affected the estimation of CO_{bg}.

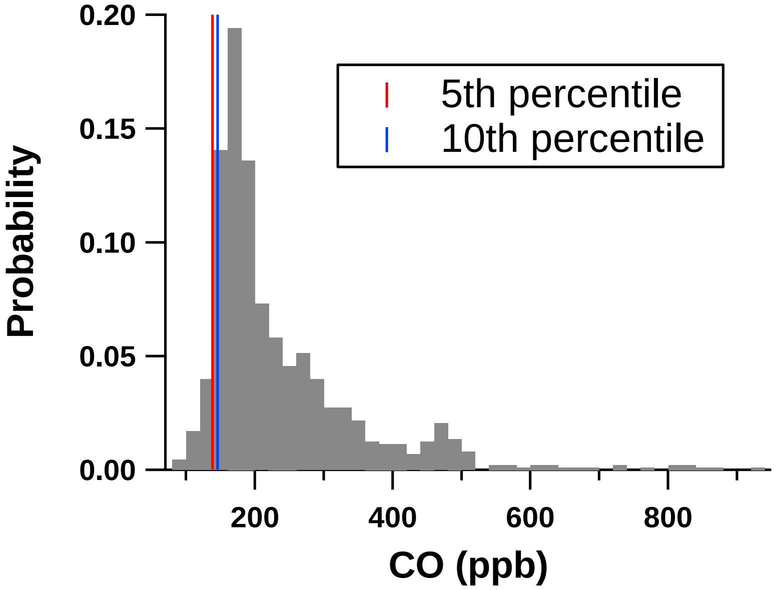

Figure S1. Probability density of measured CO mixing ratio (shaded bars). Red and blue vertical lines correspond to the 5th and 10th percentile values of the observed CO mixing ratios.

S2. Washout process

In this study, we assume that the most important loss process of BC-containing particles is their cloud droplet activation and subsequent precipitation (i.e., rainout). There is another wet removal process in which BC-containing particles are scavenged by falling rain droplets (i.e., washout). The scavenging rate depends on the particle size (d_p), raindrop size (D_p), and precipitation intensity (PI) (Seinfeld and Pandis, 2006). The accumulation mode aerosols (diameter of 0.1 – 1 μm) are less efficiently removed by the washout process, because the particle-raindrop collision efficiency has a minimum for the particle size range of the accumulation mode aerosols.

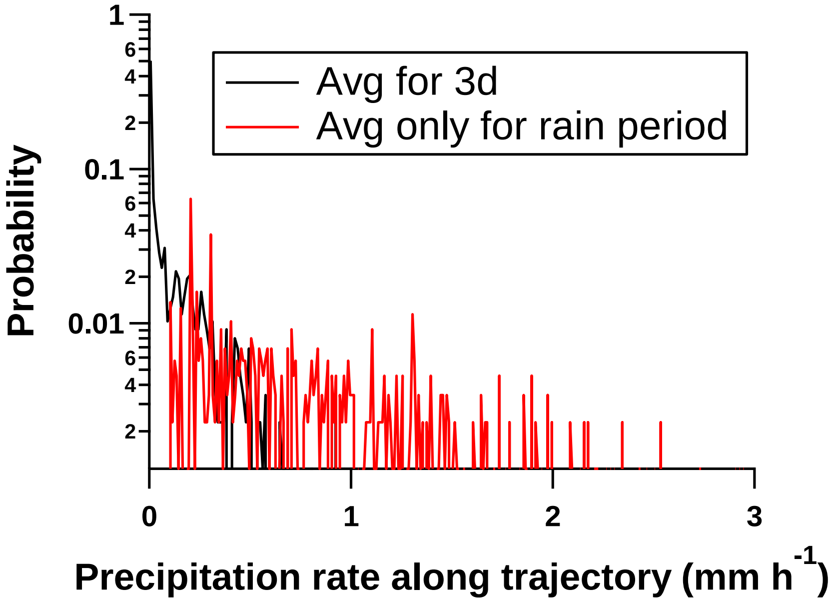

Figure S2. The probability density of the average of PI along trajectory for 3d-backward time (black line) and rain period (red line).

The average PIs along a backward trajectory were calculated for the rain period in 3d-backward time (PI > 0 mm h^{-1}). The probability of them is shown in Figure S2. They ranged from 0.1 to 2.5 mm h^{-1} (median = ~0.6 mm h^{-1}). Using the PI value of 0.6 mm h^{-1}, the scavenging rates of accumulation mode particles were estimated to be 6E-3 h^{-1} (6E-5 h^{-1}) with the assumed rain drop diameter of 0.2 mm (2 mm). The corresponding time constants are around 7 (694) days. These are longer than the typical transport time from the continent to the observation site. The raindrop diameter is not available. However, the derived time constants of the washout process for the accumulation mode aerosols are longer than those of the air mass transport from the continent to the observation site (<~2 days). On average, the loss particle mass concentration was controlled mainly by the rainout process.

In contrast to the accumulation mode aerosols, the Aitken mode (<0.1 μm) aerosols can be efficiently removed by the washout process (Seinfeld and Pandis, 2006). It is important to consider the washout process for the quantitative analysis of the loss of the number concentrations of BC-containing particles, which is beyond of the scope of this study. However, the washout process can partly account for the observed changes in the number size distributions of BC-containing particles.

References

Seinfeld, J.H., and Pandis, S. N., Atmospheric Chemistry and Physics, 2nd ed., John Wiley &Sons, New York, 2006