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Seasonal features and origins of carbonaceous aerosols at Syowa Station, coastal Antarctica

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Weingartner's correction procedure of BC mass concentrations

In an aethalometer, BC concentrations are measured by light attenuation resulting from optical absorption of BC collected on the filter tape. As earlier works have suggested (e.g., Weingartner et al., 2003; Bond et al., 2013), filter-based BC measurements have scattering and shadowing effects that can engender error of BC measurements. Therefore, we used Weingartner's procedures for this study to correct BC concentrations (Weingartner et al., 2003). In Weingartner's procedure, BC concentrations were corrected using the following process. Light attenuation (ATN) is denoted as

$$\mathrm{ATN} \equiv \ln\frac{I_0}{I}, \tag{1}$$

where I_0, and I respectively represent the light intensity incident to the filter and the light intensity after passage through the filter spot. The optical attenuation coefficient (b_{ATN}) is calculated using the following equation.

$$b_{ATN} = \frac{A\Delta ATN}{Q\Delta t} \tag{2}$$

Therein, A, Q, Δt, and ΔATN respectively represent the spot area, flow rate, time interval, and attenuation change. Then, mass BC concentrations (M_{BC}) are estimated and corrected as

$$M_{BC} = \frac{b_{abs}}{\sigma_{abs}} = \frac{b_{ATN}}{\sigma_{ATN}}R \quad , \tag{3}$$

where b_{abs}, σ_{abs}, σ_{ATN}, and R respectively denote the optical absorption coefficient, mass specific optical absorption cross-section, mass specific attenuation cross-section, and correction factor for multiple scattering of light and shadowing effects in Weingartner's correction. R is estimated using the following equation.

$$\mathrm{R} = \frac{1}{\left(\frac{1}{f}-1\right)\frac{lnATN-\ln(10\%)}{ln(50\%)-ln(10\%)}+1} \tag{4}$$

Correction parameter f is estimated using the following relation.

$$f = a(1-\omega_0)+1 \tag{5}$$

In that equation, ω_0 is the single-scattering albedo. The determined parameters *a* are 0.87 (λ = 450 nm) and 0.85 (λ = 660 nm). Because of the low BC concentrations detected at Antarctic coasts, ω_0 was found mostly as 0.97–0.99 at Syowa Station (Yabuki et al., preparation for publication). Similar values of ω_0 were measured also at Neumayer (Weller et al., 2013). Therefore, *f* values can be 1.02–1 using our measurement conditions. Here, we used 1.01 as the *f* values. Furthermore, Δt was 120 min (2 hr) in this study because of lower BC concentrations. The corrected BC concentrations were estimated for all BC data (recorded every 15 min) in this study. In aethalometer measurements using AE31, σ_{ATN} is 14,625 nm m^2 g^{-1} λ^{-1}.

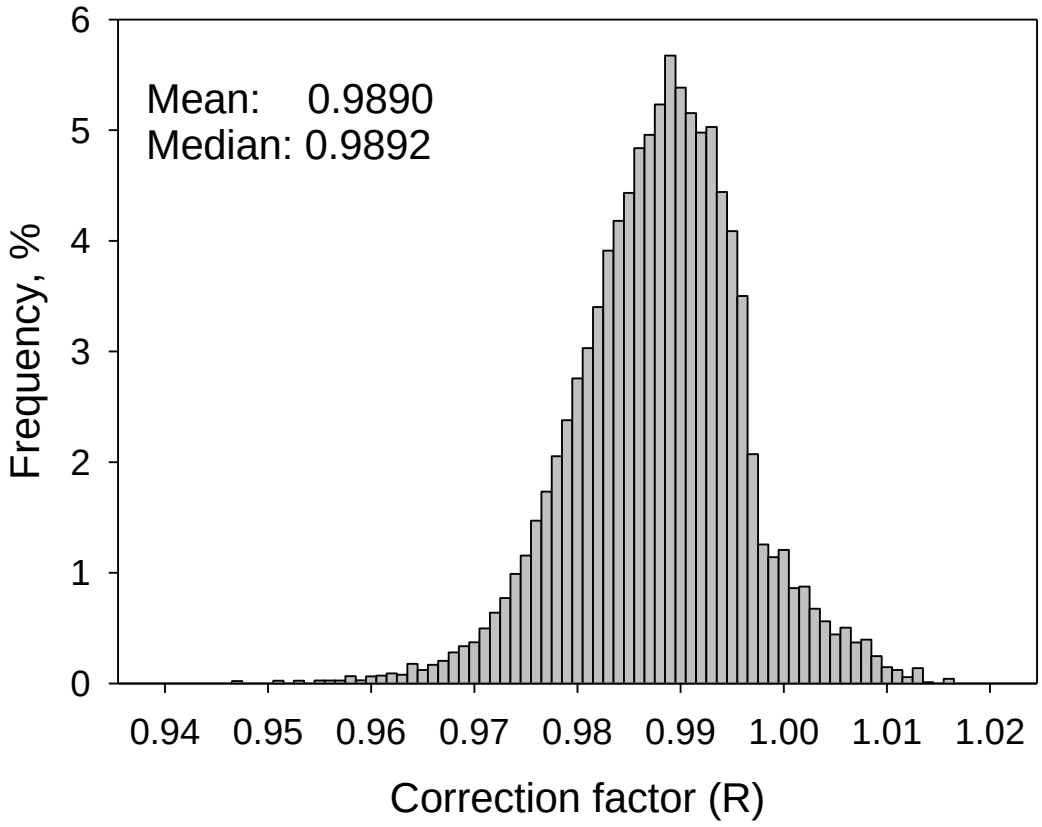

Figure S1: Histogram of correction factor of Weingartner's correction in aethalometer data measured at Syowa Station, Antarctica during our measurements.

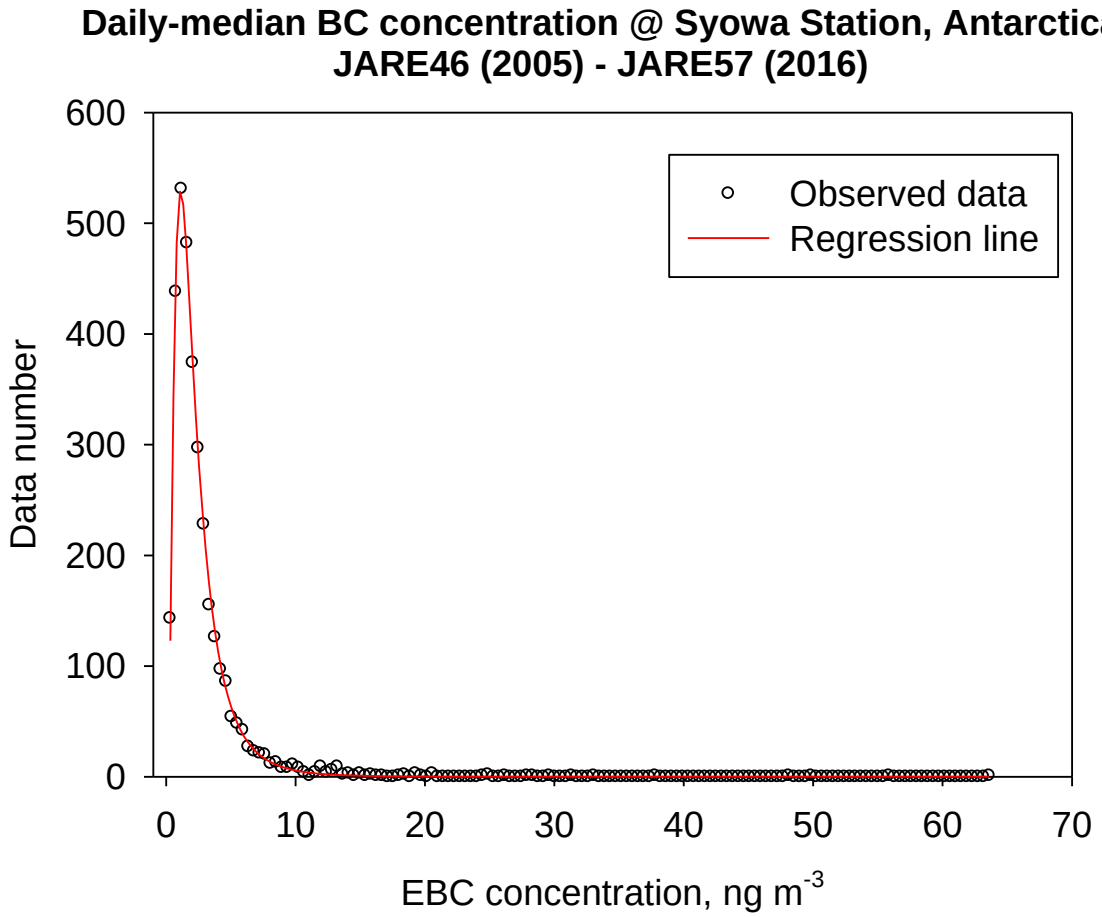

Figure S2: Histogram of EBC concentration measured at Syowa Station, Antarctica in 2005–2016.

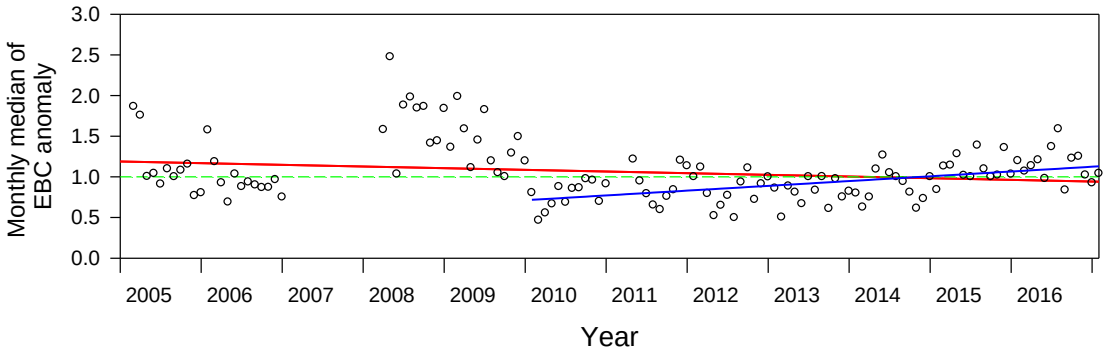

Figure S3: Variations of monthly median of EBC anomaly at Syowa Station, Antarctica. Red, blue, and green lines respectively present regression lines in all periods (2005–2016) and 2010–2016, and anomaly value of 1.

In our EBC dataset, long gaps in data exist because of instrumental troubles in 2007 and Jan. – March in 2011; short data gaps were caused by local contamination. In addition, EBC concentrations showed strong seasonal variations as depicted in Fig. 3. First, one must remove the strong seasonality before analysis of long-term trends. Daily median EBC concentrations during our measurements were estimated as normal-like values of EBC concentrations at Syowa Station. Then, the ratios of ambient EBC concentrations to the normal-like EBC concentrations were calculated and were used as anomaly values. The monthly median EBC anomaly is shown in Fig. S3. Linear regression lines were fitted during all periods (2005–2016) and were 2010–2016 for trend analysis. The regression line in all periods showed a slight decreasing trend with slope of -0.036 ng m^{-3} yr^{-1} (p = 0.0145), whereas the regression line in 2010–2016 showed a slight increasing trend with a slope of 0.105 ng m^{-3} yr^{-1} (p < 0.001).

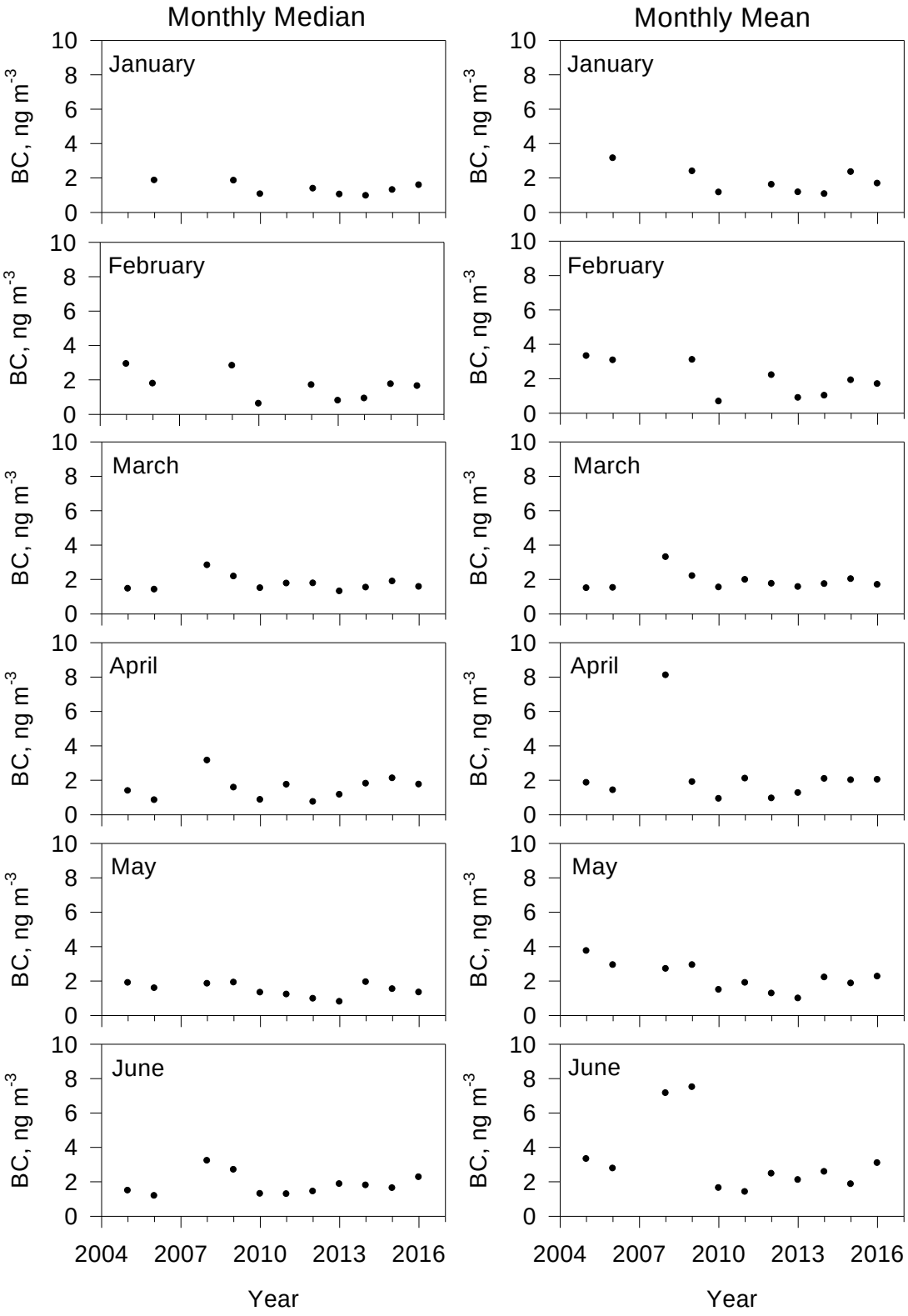

Figure S4: Monthly long-term of EBC concentrations at Syowa Station, Antarctica in 2005–2016.

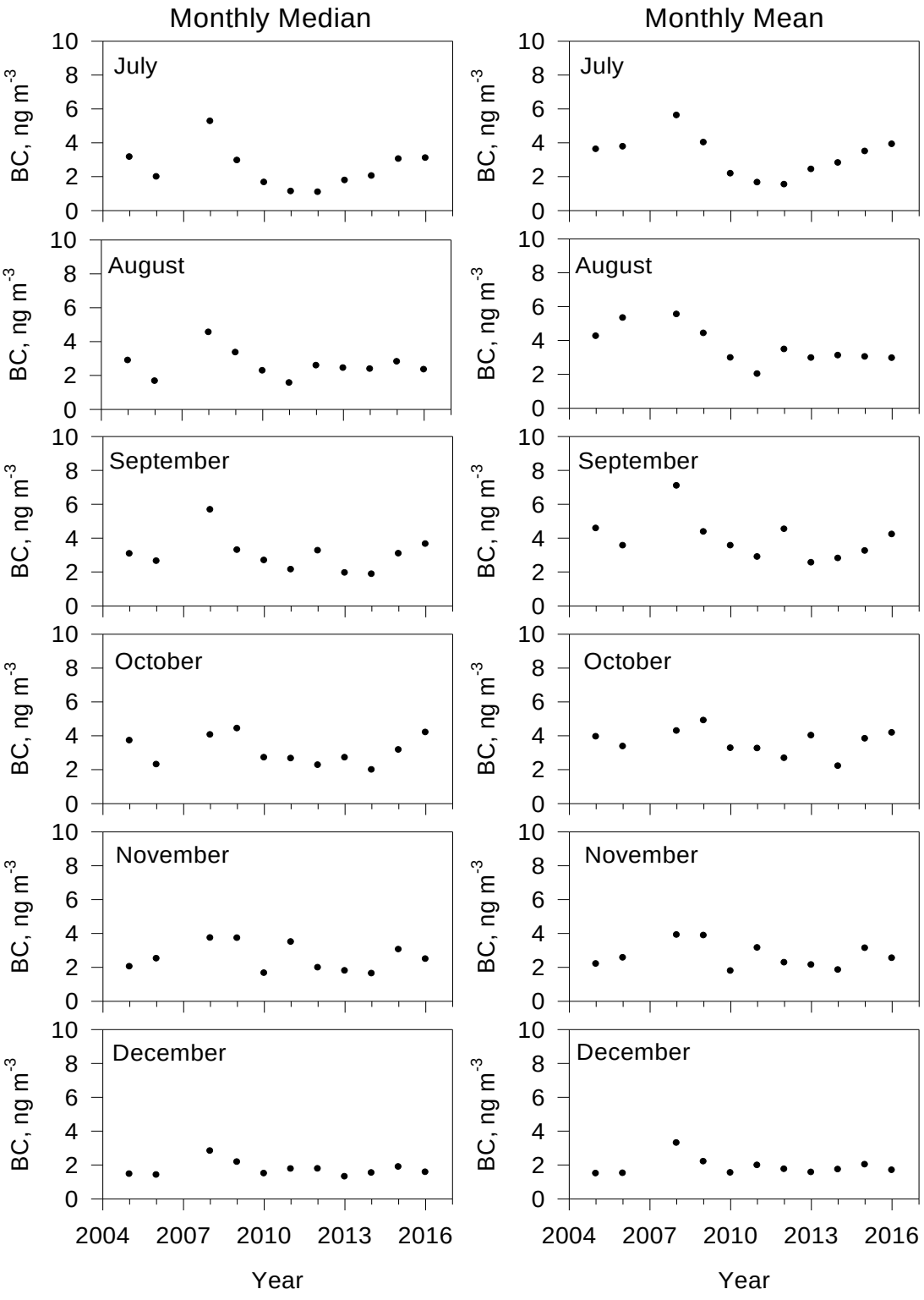

Figure S4: (continued)

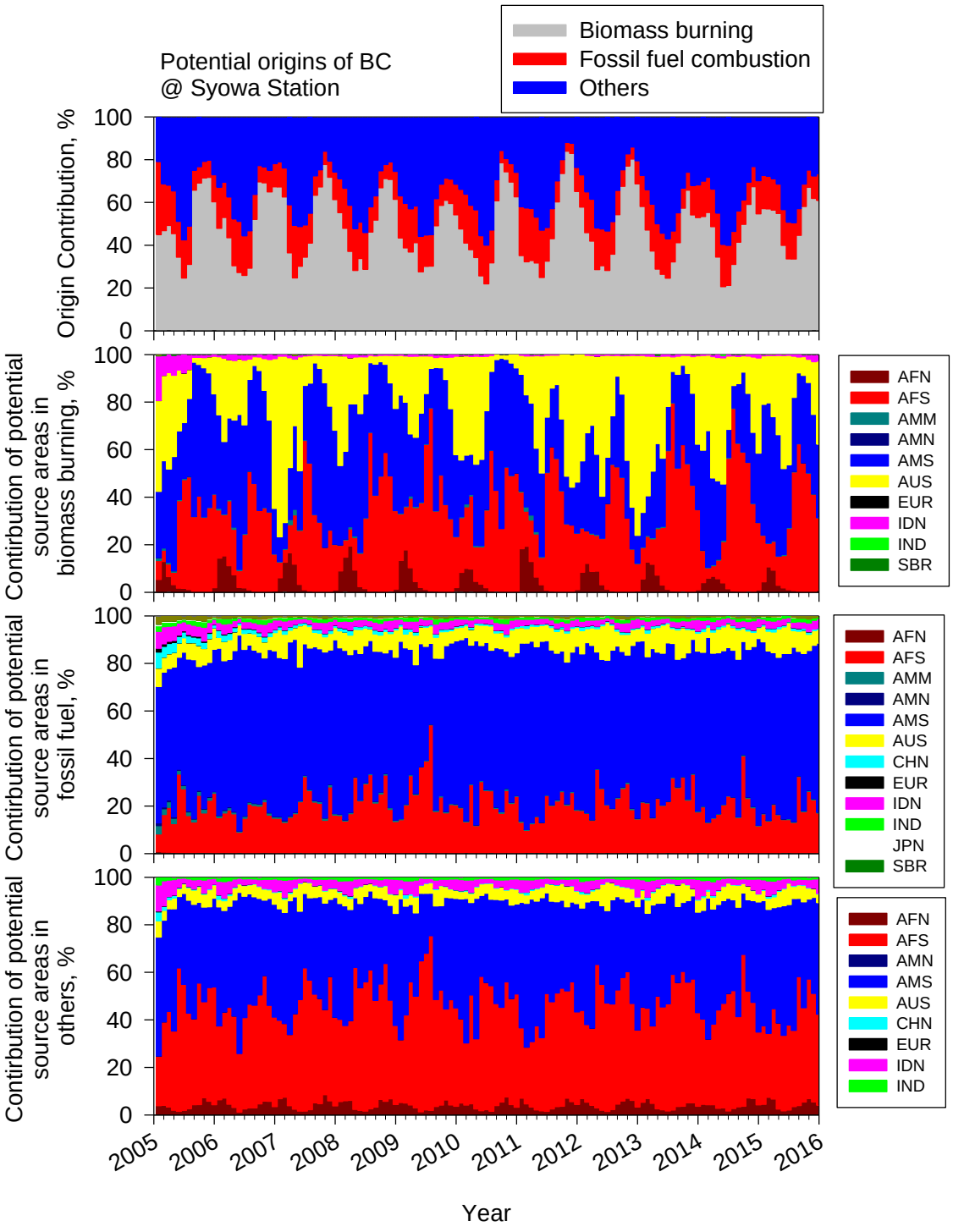

Figure S5: Seasonal features of contributions of BC origins and their PSA. In the figure, AFN, AFS, AMM, AMN, AMS, AUS, EUR, CHN, IDN, IND, JPN, and SBR respectively denote northern Africa, southern Africa, Central America, North America, South America, Australia, Europe, China, Indonesia, India, Japan, and Siberia (as shown in Figure 2).

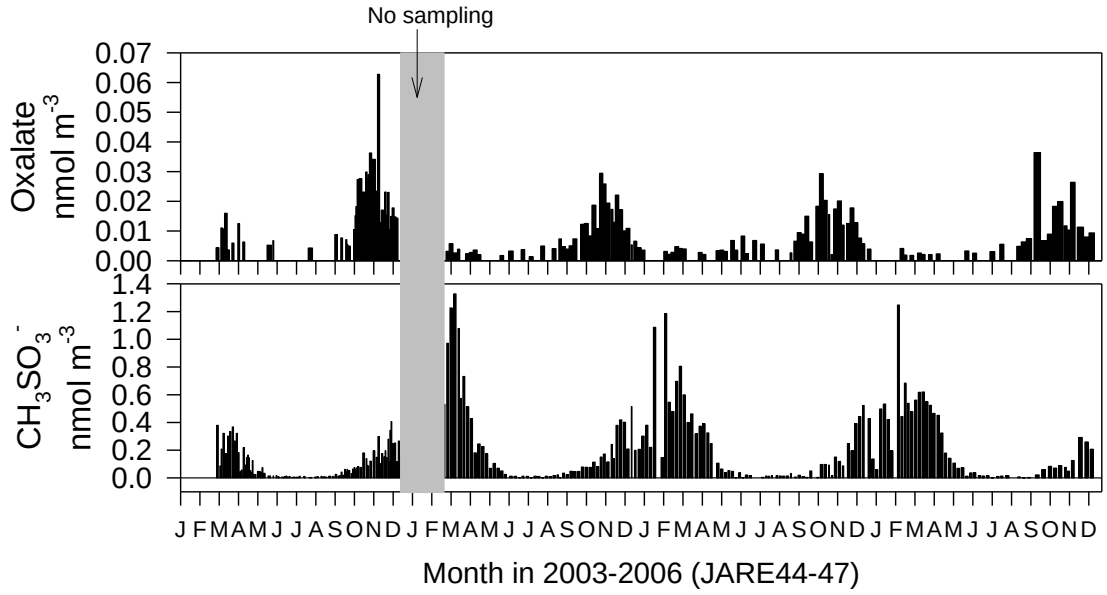

Figure S6: Seasonal variation of concentrations of oxalate and $CH_3SO_3^-$ in aerosols at Syowa Station, Antarctica.

Non-size-segregated aerosol sampling was done at Syowa Station, Antarctica. Aerosol sampling in 2003 and 2004–2006 was conducted, respectively, at an atmospheric observatory and clean air observatory. The clean air observatory was built in January 2004. Aerosol sampling was controlled using a wind selector to avoid local contamination. Sampling and analytical procedures were applied in accordance with Hara et al. (2004, 2010).

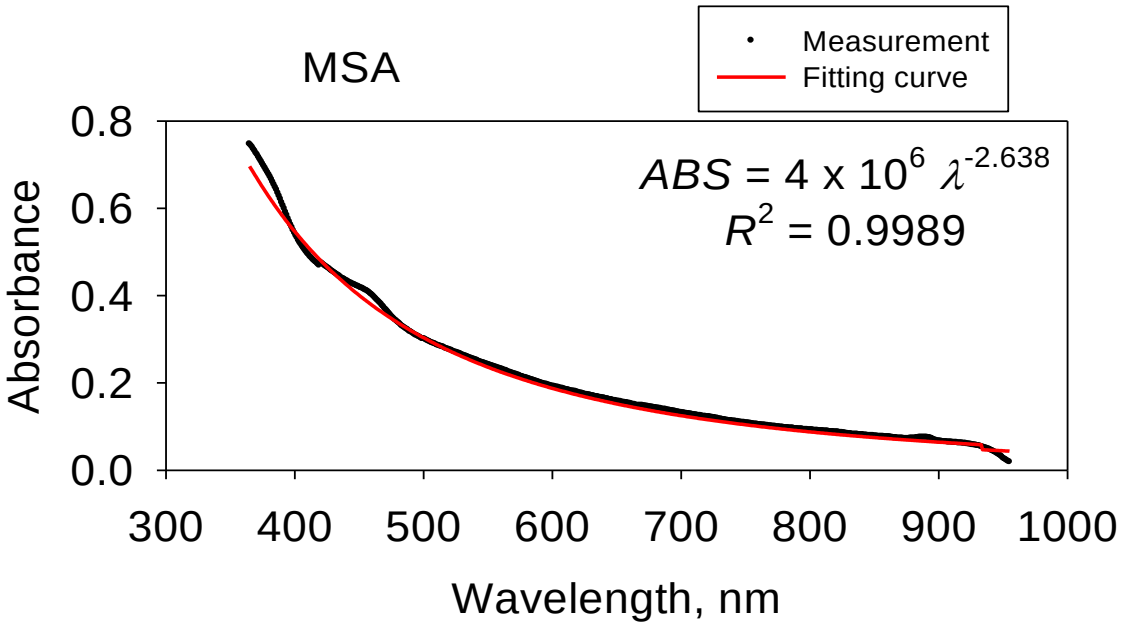

Figure S7: Absorbance of aqueous solution (ca. 3 M) of CH_3SO_3H.

Absorbance (optical absorption) of CH_3SO_3H aqueous solution was found using a spectrophotometer with 5 nm bandwidth and light wavelength accuracy of ±2 nm (Genesis 30; Thermo Scientific). The measurable wavelength range is 325–1000 nm. Before determination, CH_3SO_3H was diluted to ca. 3 M using ultrapure water.

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