Supplementary information

Seasonal features and origins of carbonaceous aerosols at Syowa Station, 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), the 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 procedures. 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 passing 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

$$M_{BC} = \frac{b_{abs}}{\sigma_{abs}} = \frac{b_{ATN}}{\sigma_{ATN}\left(\left(\frac{1}{f}-1\right)\frac{lnATN-\ln(10\%)}{ln(50\%)-ln(10\%)}+1\right)}, \tag{3}$$

where b_{abs}, σ_{abs}, and σ_{ATN} respectively denote the optical absorption coefficient, mass specific optical absorption cross-section, and mass specific attenuation cross-section. Correction parameter f is estimated using the following relation.

$$f = a(1-\omega_0)+1 \tag{4}$$

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 14625 nm m^2 g^{-1} λ^{-1}.

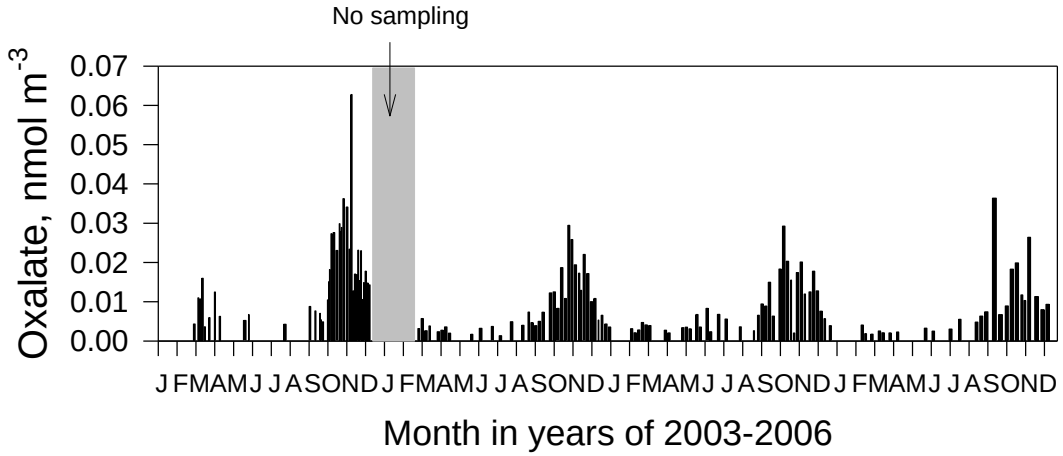

Figure S1: Seasonal variation of oxalate concentration in aerosols at Syowa Station, Antarctica.

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 because 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).

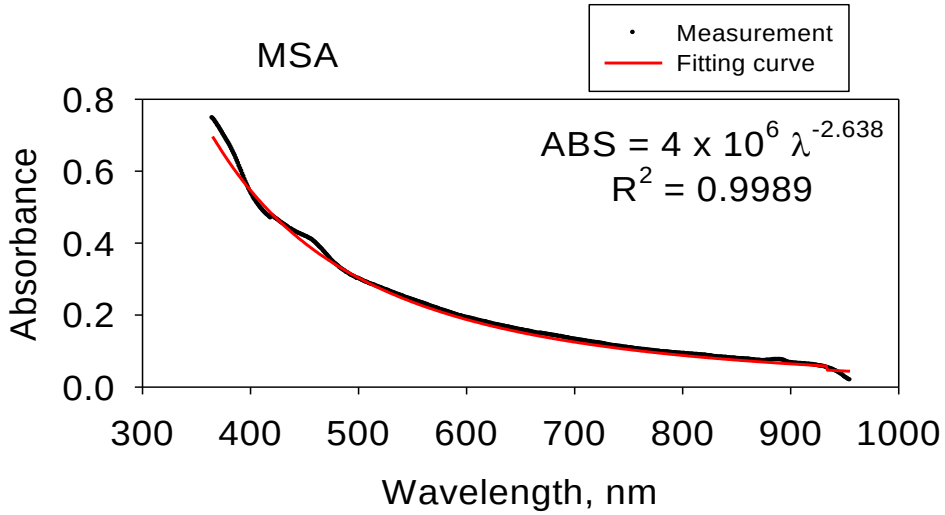

Figure S2: Absorbance of aqueous solution (ca. 3 M) of CH_3SO_3H.

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 measureable wavelength range is 325–1000 nm. Before determination, CH_3SO_3H was diluted to ca. 3 M using ultrapure water.

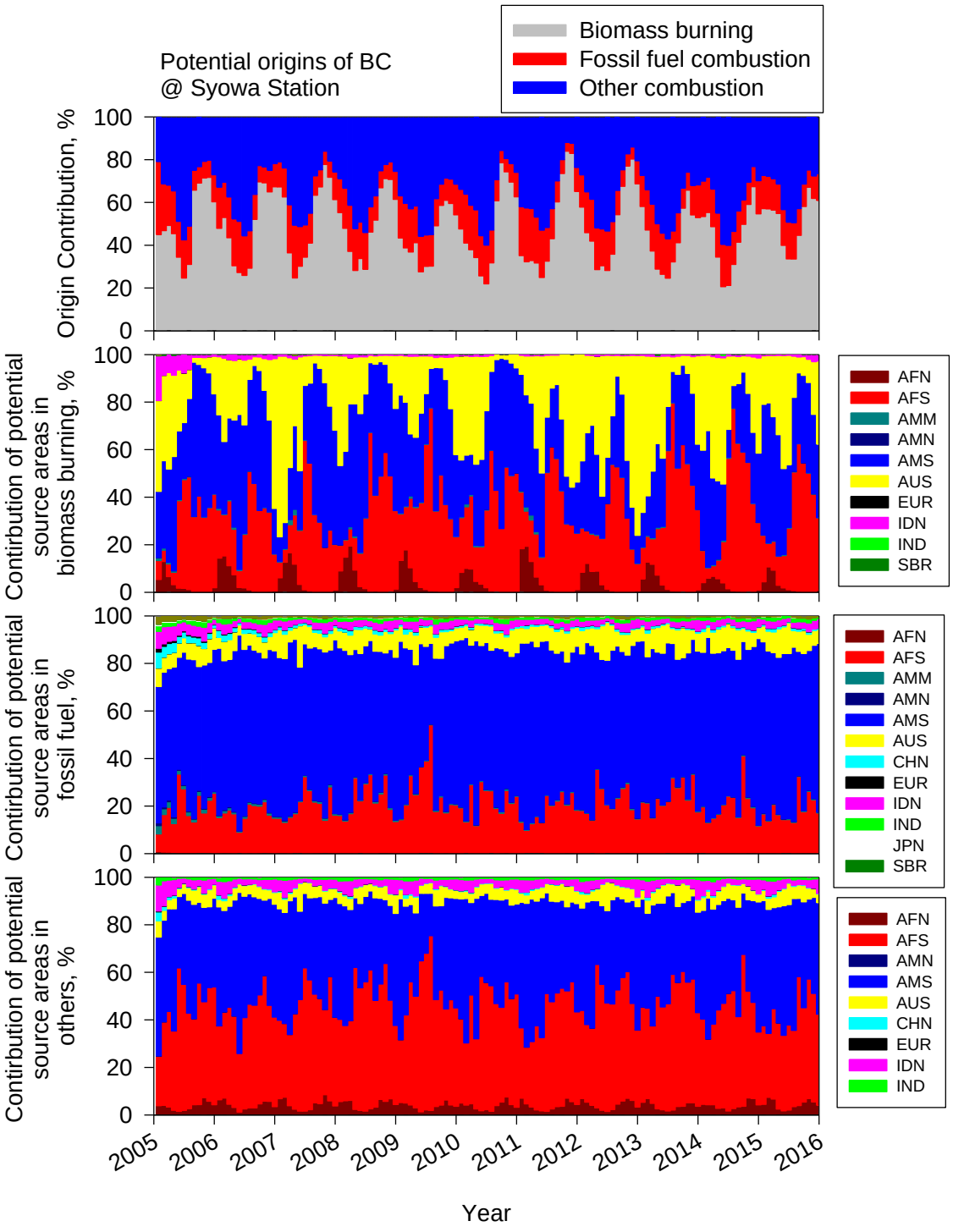

Figure S3: Seasonal features of BC origins and their PSA. AFN, AFS, AMM, AMN, AMS, AUS, EUR, CHN, IDN, IND, JPN, and SBR respectively denote northern Africa, southern Africa, Central America, North America, South America, Australia, Europe, China, Indonesia, India, Japan, and Siberia (as shown in Figure 2).

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