

Interactive comment on “Measurement of black carbon at Syowa station, Antarctica: seasonal variation, transport processes and pathways” by K. Hara et al.

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Reply to Comments from reviewer 2

We would like to thank reviewer 2 for helpful comments on the manuscript. Author's responses (preceded by **R**) to comments by the reviewer 2 (preceded by Ref. 2) are as follows;

(1)**Ref. 2:** a thorough descriptions of analytical aspects associated with the raw BC data (i.e. clear statement and reasoning regarding used calibration and estimates of the lower detection limit (depending on time resolution)).

R: We added description about quality of BC data and comparison between BC con-

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centration from PSAP and aethalometer and mass concentration of carbonaceous particles from single particle analysis, as follows.

Data quality: Figure 5 indicates short-term variation of raw BC data with 15 minute resolution and hourly mean BC data. Although several negative values were found in the raw data, negative value was not observed in the hourly mean data. Some hourly-BC data showed negative values (14 % in PSAP and 3 % in aethalometer). All negative values of the hourly-mean data were removed before the data processing and discussion. Mean standard deviation in the period of Figure 5 was approximately 1.32 in the hourly mean BC data and 0.94 in daily mean data. Therefore, detection limit of aethalometer was estimated to about 1.3 ng m^{-3} in hourly mean data and 0.94 ng m^{-3} in daily mean data in the present study. Likewise, detection limit of PSAP was estimated to ca. 10.2 ng m^{-3} .

Comparison of mass concentration: In order to calibrate BC data, we attempted to compare BC data with single particle analysis. Figure 7 indicates typical example of aerosol particles during BC peak on 18 June 2004. This particle has compact aggregate structure. In single particle analysis by SEM-EDX, no elements with atomic number larger than 9 were detected from the particle, so that this type of the particle should be identified as external mixing states of carbonaceous particle. The external mixture of carbonaceous particles was not observed from samples collected in background BC concentration (a few ng m^{-3}). The closest source of BC is diesel power generator located at main area of Syowa as shown in Figure1. High temperature processes in diesel engine, however, can release chain-like agglomerates of spherical particle (15-30 nm in diameter) (Maricq, 2007). The chain-like agglomerate particles (soot) were observed in Arctic haze due to significant anthropogenic impact (Hara et al., 2003), whereas the compact aggregate particles such as particle in Figure 7 were not observed in the Arctic haze. The compact aggregate particles were observed in the haze due to biomass burning in Southern Africa (Li et al., 2003). The carbonaceous particles with compact aggregate structure might be not released from diesel power

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station. Also, presence of mineral particles (as listed in Table 1), which may be transported from mid-latitudes to Antarctic coast, suggests long-range transport of carbonaceous particles (BC). Table 1 showed relative abundance (number fraction) of each aerosol particle on 18 June 2004. Sea-salt and mineral particles were identified by detection of sea-salts (Na and Cl) and crustal elements (Si and Al). Since single particle analysis was operated using SEM-EDX in the present study, internal mixture between carbonaceous particle and other particles cannot be identified. Using relative abundance and the number concentration (75706 L^{-1} in $D_p > 0.3 \text{ }\mu\text{m}$ and 201 L^{-1} in $D_p > 2.0 \text{ }\mu\text{m}$) from OPC, the number concentration of carbonaceous particles were estimated to 16 L^{-1} in coarse particles ($D_p > 2.0 \text{ }\mu\text{m}$) and 757 L^{-1} in fine particles ($0.3 < D_p < 2.0 \text{ }\mu\text{m}$). Assuming density of $0.7 - 1.0 \text{ g cm}^{-3}$ for carbonaceous particles and spherical shape with diameter of $2 \text{ }\mu\text{m}$ in coarse particles and $0.2 \text{ }\mu\text{m}$ in fine particles, mass concentration of carbonaceous particles was estimated to $31.6 - 45.2 \text{ ng m}^{-3}$. This estimated mass concentration was corresponded well to BC concentration obtained from PSAP (42.7 ng m^{-3}) during aerosol sampling on 18 June 2004. Although some investigations pointed out that scattering by solid aerosol particles on filter can lead to overestimation of absorption coefficient and BC concentration especially in higher aerosol concentration (Bond et al., 1999; Arnott et al., 2005), this coincidence strongly suggests that sea-salt and mineral particles caused insignificant or less overestimation of BC concentration (or absorbing coefficient) in BC measurements by PSAP and aethalometer. Therefore, BC concentration from PSAP and aethalometer may depend on the concentration of absorbing particles such as BC in the present study. Since there seem to be matter to be discussed for data correction in the case of low BC and aerosol concentration (Bond et al., 1999; Weingartner et al., 2003; Arnott et al., 2005), non-corrected BC concentration was used in the present study.

These disruptions were added to the text in the revised manuscript.

(2) **Ref. 2:** a clear outline of the deployed data selection procedure, which reliability needs to be substantiated, at least, by demonstrating the sensitivity of the final results

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by the choice of alternative selection criteria. In view of the present (loosely stated) method, it is not clear why only the variability of the continuous CN readings is used and why high wind speed episodes are not skipped from the data set? It is known that the latter (as going along with snow drift) may easily bring fragmented ice crystals to the aerosol filter, adding to artefacts by anthropogenic BC or biogenic, light absorbing material.

R: As shown in Figure 3 (typical example of local contamination) in the manuscript, BC concentration increased obviously due to local contamination under the condition of wind blowing from dirty air sector (main area of Syowa) and weak wind. For filtering of local contaminate (or contaminable) data, BC data was filtered using wind data (wind speed and direction) and CN concentration, because CN concentration is very sensitive to local contamination. The contaminated BC data can be screened by the criteria. Variation of CN concentration, however, has larger standard deviation under the storm conditions with wind blowing from clean air sector. The larger standard deviation should be caused by transport of sea-salt particles from open sea and sea-ice and fragments from snow surface. Some investigations (e.g., Bond et al., 1999; Weingartner et al., 2003) showed that solid particles such as sea-salt particles cause overestimation in BC measurement using PSAP and aethalometer. The contribution of the overestimation in BC measurements still seems to be matters to discuss (e.g., Bond et al., 1999; Weingartner et al., 2003, Arnott et al., 2005). When the overestimation by solid particles was insignificant, filtering of BC data in the storm conditions lead to underestimation of BC concentration. From the reasons, we remained BC data in strong wind ($>15 \text{ m sec}^{-1}$) to avoid the underestimation of BC concentration. Indeed, solid particles lead to insignificant influence (overestimation) on BC measurements in the present study (details are described below).

Insignificant influence of solid particles on BC measurement: In order to verify the effect of solid particles on the filter on BC measurement in the present study, we attempt to compare between aerosol number concentrations and BC concentration

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(output values) from PSAP and aethalometer, as shown in Figure 6. Relationship was estimated as follows;

$$[BC] = 0.0005 \times [D_p > 0.3 \mu\text{m}] + 3.63 \text{ (R}^2=0.299\text{)}, \text{ and}$$

$$[BC] = 0.0117 \times [D_p > 2.0 \mu\text{m}] + 5.39 \text{ (R}^2=0.025\text{)},$$

where $[BC]$, $[D_p > 0.3 \mu\text{m}]$, and $[D_p > 2.0 \mu\text{m}]$ mean BC concentration, the number concentration of aerosol particles with size of $D_p > 0.3 \mu\text{m}$ and $> 2.0 \mu\text{m}$, respectively. Poor correlation in both size ranges suggests that deposition of aerosol particles on the filter did not result in the overestimation. Also, comparison between BC concentration and Na^+ concentration with 2-3 day resolution (not shown in this study) indicates poor correlation ($[BC] = 0.0238 \times [\text{Na}^+] + 5.63$, $R^2 = 0.0149$). Therefore, BC concentration (or output signal) may depend mostly on concentration of light absorbing particles such as BC, though solid particles can lead to overestimate slightly BC concentration.

These disruptions were added to the text in the revised manuscript.

Influence of BC in snow surface: When significant amount of BC was present in surface snow, wind blowing can lead to re-emission of BC from snow surface. According to soot measurement in surface snow at South Pole (Warren and Clarke, 1990), most of soot was deposited in the prevailing downwind direction. Consequently, BC in snow surface may be lower or background level in prevailing upwind direction (clean air sector) even at Syowa. Here, we attempt to estimate BC concentration in snow surface in order to understand the contribution of the re-emission. Under the strong winds, sea-salt particles increased approximately by $7 \mu\text{mg m}^{-3}$ of sea-salt particles (100 n mol m^{-3} of Na^+) at Syowa (Hara et al., 2004). Assuming that sea-salt and BC particles in the strong winds was derived only from snow surface by wind blowing, BC concentration in surface snow was estimated to more than 700 ng g^{-1} , to reach 1 ng m^{-3} of atmospheric BC concentration, using ratio of BC and Na^+ in aerosols and Na^+ concentration in surface snow around Syowa (Osada et al., 2001). The estimated value should be unrealistic level in surface snow of "clean air sector", because the highest

concentration of soot in downwind direction at South Pole was 3 ng g^{-1} (Warren and Clarke, 1990). Thus, re-emission of BC from snow surface by wind blowing cannot make significant contribution to ambient BC concentration.

These disruptions were added to the text in the revised manuscript.

(3) **Ref. 2:** a presentation of the deployed data reduction, collapsing the remaining raw data into the final records at various time resolution. Here, the probability distribution of the BC data greatly matters, which is expected to be heavily skewed towards higher concentrations making the arithmetic means (apparently reported here?) controlled by few outstanding values.

R: Monthly mean BC data was needed to compare the seasonal variation of BC between at Syowa and other sites, because some data was published by monthly mean data. In order to see the variation of BC concentration in each month, we change to box plot including mean, median, 5 %, 25 %, 75 %, and 90 % values. Median values also show similar variation to monthly mean, though median value was lower than the monthly mean.

(4) **Ref. 2:** an adequate reporting and illustrating of the final continental BC data set backed up by an overall quality assessment (addressing among others the temporal distribution of contamination or negative signal readings).

R: During the daytime, weak winds can lead to local BC distribution around Syowa on scale of a few kilometers, as suggested by Hagler et al. (2008). Higher BC concentration, however, was obtained after stronger wind blowing from prevailing wind direction without local BC sources. According to investigation about local anthropogenic impact at Summit, Greenland by Hagler et al. (2008), atmospheric BC concentration can drop rapidly within first few kilometers of distance from BC source. In their estimation (Hagler et al., 2008), distance from the source is approximately 10, 14, and 27 km to decrease to 1 ng m^{-3} of BC concentration in the case of wind speed of 7.9, 4.5 and 1.2 m s^{-1} , respectively. Although BC can be dispersed from main area of Syowa in

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weak winds, weak winds lead to local contamination with very local scale. Hence, wind blowing for 1 - 3 hours should be enough to transport air mass from "clean air sector" without local impact even in wind speed of $2\text{--}5\text{ m s}^{-1}$. Thus, it may take 1 - 2 hours to obtain BC concentration without local anthropogenic impact under the conditions with wind blowing from clean air sector. According to Sato and Hirasawa (2007), the diurnal variation of the surface wind speed at Syowa is caused by the diurnal variation of katabatic wind. Hence, the higher BC concentration in the nighttime may be caused by transport of continental air by katabatic wind. Negative values of hourly mean BC concentration was observed during the measurement by aethalometer. The percentage of the negative values was only 3

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 9883, 2008.

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