



Elemental carbon in snow at Changbai Mountain, Northeastern China

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Elemental carbon in snow at Changbai Mountain, Northeastern China: concentrations, scavenging ratios and dry deposition velocities

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Received: 5 May 2013 – Accepted: 10 May 2013 – Published: 30 May 2013

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Light absorbing aerosol, in particular elemental carbon (EC), in snow and ice enhance absorption of solar radiation, reduce the albedo, and is an important climate driver. In this study, measurements of EC concentration in air and snow are performed concurrently at Changbai Station, Northeastern China, from 2009 to 2012. The mean EC concentration for surface snow is $987 \pm 1510 \text{ ng g}^{-1}$ with a range of 7 to 7636 ng g^{-1} . EC levels in surface snow around (about 50 km) Changbai Mountain are lower than those collected on the same day at Changbai station, and decrease with distance from Changbai station, indicating that EC load in snow around Changbai Mountain is influenced by local source emissions. Scavenging ratios of EC by snow are calculated through comparing the concentrations of EC in fresh snow with those in air. The upper-limit of mean scavenging ratio is 137.4 ± 99.7 with median 149.4, which is smaller than those reported from Arctic areas. The non-rimed snow process may be one of significant factors for interpreting the difference of scavenging ratio in this area with the Arctic areas. Finally, wet and dry depositional fluxes of EC have been estimated, and the upper-limit of EC wet deposition flux is $0.46 \pm 0.38 \mu\text{g cm}^{-2} \text{ month}^{-1}$ during the three consecutive snow season, and $1.32 \pm 0.95 \mu\text{g cm}^{-2} \text{ month}^{-1}$ for dry deposition flux from December to February during study period. During these three years, 77% of EC in snow is attributed to the dry deposition, indicating that dry deposition processes play a major role for EC load in snow in the area of Changbai, Northeastern China. Based on the dry deposition fluxes of EC and hourly black carbon (BC) concentrations in air, the estimated mean dry deposition velocity is $2.81 \times 10^{-3} \text{ ms}^{-1}$ with the mean median of $3.15 \times 10^{-3} \text{ ms}^{-1}$. These preliminary estimates for the scavenging ratio and dry deposition velocity of EC on snow surface will be beneficial for numerical models, and improve simulations of EC transport, fate and radiative forcing in order to ultimately make better climate prediction.

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1 Introduction

Elemental carbon (EC), used as proxy for Black Carbon (BC) is emitted from incomplete combustion of coal, fossil fuel and biomass burning (Novakov et al., 2003; Jacobson, 2004). EC causes a net positive climate forcing through both direct and indirect aerosol effects (Haywood and Shine, 1995; Koch and Del Genio, 2010), and through the EC-snow albedo effect (Warren and Wiscombe, 1980; Hansen and Nazarenko, 2004). Recent work has also identified EC as a much larger contributor to global climate change than previously anticipated (Flanner et al., 2007; Ramanathan and Carmichael, 2008). Several studies have suggested that EC may be the second or third most important climate driver after CO₂ on a global scale. In some regions it may be comparable to the greenhouse forcing (Bachmann, 2009; Shindell and Faluvegi, 2009).

Current model estimates of global present-day EC-snow forcing span from 0.03 to 0.13 W m⁻² (Flanner et al., 2007, 2009; Koch et al., 2009; Rypdal et al., 2009), and the maximum zonal averaged forcing has been predicted to reach almost 1.5 W m⁻² in the Arctic during the spring snow melt (Flanner et al., 2007). The results of controlling experiments for injecting artificial soot into snowmaking verified that BC reduced snow albedo at levels that have been predicted by radiative transfer models (Brandt et al., 2011; Hadley and Kirchstetter, 2012). Moreover, small changes in snow albedo can exert a large influence on climate by altering the timing of snow melt and triggering the snow/ice-albedo feedback (Hansen and Nazarenko, 2004; Jacobson, 2004; Flanner et al., 2007; Ming et al., 2012). Although Arctic-mean snow forcing is much greater than global-mean, radiative forcing exerted by BC outside the Arctic may have a significant influence on Arctic climate via dynamical changes (AMAP, 2011). Recently research indicated that EC is a key element in Himalayan glacier melting (Menon et al., 2010; Ming et al., 2012; Yasunari et al., 2012). Therefore, a highlight on EC climatic effects has received great attention also outside the Polar Regions.

The snow cover in China is mainly distributed in Northeast, Northwest and the Tibetan Plateau. Results of global climate modeling suggest that the Tibetan Plateau

ACPD

13, 14221–14248, 2013

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were obtained and each sampled for 12 h, and a MAC value of $8.68 \text{ m}^2 \text{ g}^{-1}$ ($R^2 = 0.96$, $n = 25$) was found for this PSAP-ITM. At Changbai station, the sample inlet of PSAP-ITM was installed 3 m above ground level. Aerosol particles were collected on a 3 mm diameter sampling spot on E70-2075 W filters (glass fibers with a cellulose baking, Pall Corporation, USA) within a 0.088 cm^2 circular surface area. The air flow was dynamic and adjusted depending on the BC concentration in the air, but typical value was 50 mL min^{-1} . Raw signal data were stored as minute averages and corrected for loading effects on the filter, and the influence from co-depositing scattering particles following Bond et al. (1999). As the aerosol light scattering coefficient has not been directly observed at our station, the initial scattering coefficient is assumed to be 10 % of the aerosol light extinction (scattering + absorption). With this assumption the data was corrected for the enhanced absorption effect by light scattering particles. Data was first made into 15 min averages from which hour averages were calculated.

Desert Research Institute (DRI) Model 2001A Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA) was used to analyze EC and organic carbon (OC) in the snow. A 0.5 cm^2 circular punch from the filter was analyzed for eight carbon fractions following the IMPROVEA (Interagency Monitoring of Protected Visual Environments) protocol (Chow et al., 2007). This procedure produces four OC fractions (OC1, OC2, OC3, and OC4, respectively at 140, 280, 480 and 580°C in pure He atmosphere) and three EC fractions (EC1, EC2 and EC3 respectively at 580, 740, and 840°C in O_2/He (2%/98%) atmosphere). Charring/pyrolysis of organic carbon (OP, carbon evolved when reflectance returns to its initial value) during pure He atmosphere is determined when the intensity of the reflected light by the surface filter attained its original intensity in the O_2/He atmosphere. In the IMPROVEA protocol, OC is defined as $\text{OC1} + \text{OC2} + \text{OC3} + \text{OC4} + \text{OP}$ and EC is defined as $\text{EC1} + \text{EC2} + \text{EC3} - \text{OP}$. The detection limit for a typical punch size of 0.5 cm^2 is 0.82 and $0.20 \mu\text{g C cm}^{-2}$ for OC and EC respectively.

The filtered water through PTFE filter was stored in a refrigerator and used for chemical analysis. Concentrations of anions and cations were determined using the ion Chromatograph (DX-120, AS14, 25- μ L loop).

2.3 Meteorological parameters

Meteorological data were provided by the local weather station at the Open Research Station of Changbai Mountain Forest Ecosystems, Chinese Academy of Sciences. The air temperature, precipitation, atmospheric pressure, relatively humidity and wind speed were recorded using a Vaisala Milos 520 weather station, with 1 h observation interval (Vaisala, 2001). Temperatures of air near snow surface (2 cm above the surface) and snow–soil interface were measured at 08:00, 14:00 and 20:00 LT every day during snow season. Snow depth was recorded manually using stick measurement. Evaporation rates were determined using E20 evaporation pan with a diameter of 20 cm. The conversion coefficients from the comparison results of Huang (2000) were used to revise the overestimate evaporation by the E20 (Fu et al., 2004; Sheng et al., 2007).

3 Results and discussion

3.1 EC concentrations in snow at Changbai station

Summary of statistical EC concentrations in snow at Changbai station is shown in Table 1. The EC mean concentration for surface snow is $987 \pm 1510 \text{ ng g}^{-1}$ ranging from 7 to 7636 ng g^{-1} , higher than values measured on Tibetan Plateau and Xinjiang area in Western China (Ming et al., 2008, 2009; Xu et al., 2006, 2009) and two orders of magnitude higher than those reported in Arctic areas (Forsström et al., 2009; Doherty et al., 2010). They are comparable to those recently investigated on a road transect in this area by Huang et al. (2011), and values reported in the French Alps in the early 1990s (Sergent et al., 1993; Fily et al., 1997). They may support the conclusion of a significant

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decrease of EC emissions in the Europe and North America, but a significant increase in Asia (Novakov et al., 2003; Ito and Penner., 2005).

In this study, fresh and aged snow samples are defined according to the sampling time after a major snow event. Snow is considered to be “fresh” if the sampling is performed less than 12 h after a snow event. After 12 h or more, the snow is termed as “aged”. The EC concentration in fresh snow is $215 \pm 169 \text{ ng g}^{-1}$, which is about one seventh of that in aged snow (Table 1). The highest EC concentration in snow from the Arctic survey was also found in aged snow (Doherty et al., 2010). It may be attributed to an increase of concentration by snow sublimation, melting or by dry depositional processes of aerosols.

The mean and median EC concentrations in snow are much higher in 2011 than in 2009 and 2010 (Table 1), even though the hourly average BC concentration in air is found to be higher in 2009 ($1.85 \mu\text{g m}^{-3}$) and 2010 ($1.41 \mu\text{g m}^{-3}$) compared to 2011 ($1.33 \mu\text{g m}^{-3}$). It is also noteworthy that the EC concentrations in the third snow season are comparable with those in the first and second snow seasons except some significant high values during January and February 2012. Mean air temperature in January and February 2012 (-16.5°C) is lower than in 2010 (-14.1°C) and 2011 (-14.3°C), meaning that emissions are probably higher due to increase of coal and bio-fuel burning. Furthermore, few snowfall events occurred in January and February 2012 (see Fig. 2). These two reasons consequently results in higher values of EC in snow in January and February 2012. On the other hand, the variability of the total amount of precipitation in winter time is about a factor of four between 2009 (160 mm, snow water equivalent, similarly hereinafter) and 2010 (46 mm). Therefore, the lower EC concentration in snow in 2009 could be explained by the higher precipitation amount and dilution of EC into the snow.

In 2011, the EC concentration in surface snow significantly increased from 452 to 1066 ng g^{-1} from 3 January to 5 February, a period with no new snowfall. A slight decrease is observed on 13 February (968 ng g^{-1}) after 3 cm fresh snowfall, and reached the highest value (1512 ng g^{-1}) on 21 February (Fig. 2). Likewise, in the first twenty

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than that in the first survey. This may be attributed to lack of snowfall in the month before the investigation (see Fig. 2). This further indicates that dry deposition is playing a significant role to increase EC in the snow surface in Changbai Mountain area.

In both of these two surveys the EC concentrations around Changbai Mountain were significantly lower than the same-day concentration and monthly mean concentration from weekly snow sampling at Changbai Station. In contrast to the first survey, the second survey presented a clear horizontal gradient, where EC concentration was decreasing with distance from Changbai station. Moreover, the concentrations of BC in air from PSAP-ITM showed two distinct peaks in the diurnal variation, one in the morning and another in the evening, especially for the winter season, which is indicative of local emission from households and traffic (2013).

3.3 Scavenging ratio

The concept of scavenging ratio is based on the simplified assumption that the concentration of a component in precipitation is related to its concentration in the air (Engelmann, 1971). Thus the scavenging ratio (ω_s) of EC in snow can be calculated on a mass basis

$$\omega_s = \frac{C_{EC, \text{ fresh snow}} \cdot \rho_{\text{air}}}{C_{BC, \text{ air}}}$$

where $C_{EC, \text{ fresh snow}}$ is the concentration of EC in fresh snow, $C_{BC, \text{ air}}$ is BC concentration in air, and ρ_{air} is the density of air. The major uncertainty of the calculation of ω_s is the discrepancy between measurement techniques of BC in the air and EC in the snow. As the PSAP was calibrated using comparisons with thermo-optical method and as EC is taken as a proxy of BC in air, the BC/EC concentrations in air and snow can be directly compared. This method of calculating scavenging ratio, by comparing air and snow concentrations, has previously been used for Arctic regions (Noone and Clarke, 1988; Hegg et al., 2011).

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During the observation period, a total of 19 fresh samples were collected. The snow within the samples was probably a mixture of fresh and old snow as the protocol was to sample the top 5 cm of the snow surface. Thus, our derived result is an upper estimate of the scavenging ratio assuming old snow has higher concentrations of EC than fresh snow. According to the starting and ending time of snow events recoded by Vaisala Milos 520 weather station, the BC in air concentrations of the 1st and 3rd Quartile and median during this event were used to calculate the maximum, minimum and median of ω_s (Fig. 5). The upper-limit of mean ω_s is 137.4 ± 99.7 with mean median of 149.4 in Changbai area. It is still comparable with those in a rural site of Cheboygan County in Northern Michigan (Cadle and Dasch, 1988), but much smaller than those from Zeppelin and Ny Ålesund (Hegg et al., 2011). Three relatively heavy snow events occurred on 16 March (21.4 mm for snowfall), 21 March (22.2 mm for snowfall) and 3 December (18.4 mm for snowfall) 2010, which resulted in the dilution effect related to the depletion of the BC source since heavy snowfall. Therefore, the mean ω_s (13.8) of the three events may be taken as lower limit of scavenging ratio in Changbai Mountain area. On the other hand, much higher scavenging ratios were found on 6 April 2010 (247.8) and 5 April 2012 (381), with the snowfall of 4.3 and 0.7 mm, respectively. On these two days the BC concentrations in air was relative low with no distinct high peaks (Fig. 5). Meanwhile, both samples were collected from the last fresh snow event for each winter season with relatively warm temperatures, and where the precipitation process may have involved liquid as well as ice hydrometeors.

Scott (1981) found, studying sulfate aerosols, that snow events had evidence of riming originated from clouds with high liquid water content, and they determined values of scavenging ratios 10 to 50 times higher compared to when the snow had no evidence of riming. Snowfall with mostly non-rimed ice crystals removed sulfate from the atmosphere much less efficiently than precipitation with rimed ice crystals. Rayno and Hayer (1983) also indicated that nitrate is preferentially scavenged by snow flake compared to sulfate. Therefore the ratio of nitrate to sulfate can be used an indicator for rimed or non-rimed snow. For rimed snow, the ratio of nitrate to sulfate is usually less than 0.5

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(Mitchell and Lamb, 1989; Duncan, 1992; Endoh et al., 2003). Using the nitrate to sulfate ratio measured with the water stored from filtering, it was possible to suggest from which process the EC was scavenged. At Changbai Station, about 70 % of the snow samples had ratio of nitrate to sulfate in the range of 1.0 to 3.0 (Fig. 6), with mean ratio of nitrate to sulfate of 1.9 (ranging from 0.8 to 4.2). This is higher than e.g. in Abisko (Hegg et al., 2011) and much larger than those from Svalbard (Endoh et al., 2003). Therefore, the non-rimed snow process in Changbai station may be the significant factor for interpreting the difference between EC scavenging ratio in Changbai Station and in the Arctic.

3.4 EC deposition fluxes and velocities

Wet and dry depositional fluxes of EC have been estimated (Fig. 7). Wet deposition fluxes were obtained from the monthly volume weighted mean concentration of EC in fresh snow and the monthly amount of precipitation. As described above, the fresh samples were probably a mixture of fresh and old snow due to the sampling procedures. Again, our calculations may serve as an upper limit. The mean wet deposition was $0.46 \pm 0.38 \mu\text{g cm}^{-2} \text{ month}^{-1}$ in winter season from 2009 to 2012, which is comparable to those earlier reported in Europe (Ogren et al., 1984; Armalis, 1999).

The dry deposition is generally more difficult to measure directly, and it depends on many factors, including meteorological conditions, characteristics of the pollutants being deposited (e.g. different gaseous chemical and particle size) and characteristics of the surface on which deposition occurs (Jurado et al., 2008). Here, the difference in EC concentration between two measurements without snow fall during the time period was used. Additionally, the daily mean, near surface, temperature and the snow-soil interface temperature, both had to be below zero (see Fig. 8). This constraint reduced the possible effect from melting events enhancing the surface concentration. The enhancement effect from evaporation was roughly estimated on the basis of evaporation rate (Fig. 3) and EC concentration in surface snow. The dry deposition fluxes of EC in December, January, and February are shown in Fig. 7. The mean EC dry deposition fluxes

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was $1.32 \pm 0.95 \mu\text{g cm}^{-2} \text{ month}^{-1}$ from December to February in 2009–2012, which was more than three times that from wet deposition ($0.39 \pm 0.32 \mu\text{g cm}^{-2} \text{ month}^{-1}$) during the same time period. For the period December–February, the percentages of wet to dry deposition flux are 52 %, 40 % and 10 % for 2009, 2010 and 2011, respectively. Overall, an average of about 77 % EC in snow is attributed to the dry deposition, indicating that dry deposition process is the major contribution of EC in snow in Changbai, Northeastern China.

The dry deposition velocity of EC in Fig. 7 is calculated by dividing the dry deposition flux on snow surface with its ambient concentration, expressed as

$$V_d = F_d / \bar{C} \quad (1)$$

where, V_d (ms^{-1}) is the dry deposition velocity of EC, F_d ($\mu\text{g cm}^{-2} \text{ month}^{-1}$) is the EC dry deposition flux and \bar{C} ($\mu\text{g m}^{-3}$) is the mean BC concentration in air. Using the quartile BC concentrations in air, presented in Sect. 3.3 for calculation of the scavenging ratio, the mean dry deposition velocity is $2.81 \times 10^{-3} \text{ ms}^{-1}$ and the median $3.15 \times 10^{-3} \text{ ms}^{-1}$ with a range from 1.34×10^{-3} to $7.26 \times 10^{-3} \text{ ms}^{-1}$. Over the snow/ice surface, the dry deposition velocities for fine particles were within the range of $1.0 \times 10^{-5} \text{ ms}^{-1}$ to $1.0 \times 10^{-2} \text{ ms}^{-1}$ (Dovland and Liassen, 1976; Bergin et al., 1995; Yasunari et al., 2012). The knowledge about dry deposition velocity of EC is especially poor compared to other pollutants. Therefore, this preliminary estimate may be beneficial for numerical models and may serve to improve simulations of EC transport, fate and radiative forcing in order to ultimately make better climate predictions.

4 Conclusions

Elemental carbon concentrations in surface snow in Changbai station are $987 \pm 1510 \text{ ng g}^{-1}$ with a range of 7 to 7636 ng g^{-1} during the snow seasons in the years 2009–2012, which is comparable to those found in Europe in the early 1990s. Concen-

trations in surface snow around Changbai Mountain indicated that the load of EC in snow at the station was influenced by local source emission.

The upper-limit of mean scavenging ratio of EC was 137.4 ± 99.7 with median of 149.4, which is much smaller than those reported from Arctic sites. The non-rimed snow process at Changbai station is a significant factor for interpreting the difference in EC scavenging ratio between Changbai Station and Arctic sites.

Atmospheric deposition fluxes and dry deposition velocity of EC have been estimated. The upper-limit of EC wet deposition fluxes is $0.46 \pm 0.38 \mu\text{g cm}^{-2} \text{ month}^{-1}$ in winter seasons, while the dry deposition flux is estimated at $1.32 \pm 0.95 \mu\text{g cm}^{-2} \text{ month}^{-1}$ from December to February during the studied period. The mean dry deposition velocity is then calculated to $2.81 \times 10^{-3} \text{ m s}^{-1}$ with a median value of $3.15 \times 10^{-3} \text{ m s}^{-1}$. Over the three years studied, 77% of the EC in the surface snow was calculated to be attributed to dry deposition, indicating that dry deposition processes are a major process of deposition of EC in snow in Changbai area.

Acknowledgements. This work is a part of the project “Long range transport of black carbon and the effect on snow albedo in north-east China and in the Arctic (LOTUS)” financed by the Research Council of Norway. We appreciate our Co-workers Shijie Han and HaoXu in Changbai Station for help with collecting the snow samples and filtering procedure. Thanks also to the Swedish research council FORMAS for support via the project Black and White.

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Table 1. Summary of statistic EC concentrations (ngg^{-1}) in snow at Changbai station.

Sampling period	Number <i>N</i>	Snowfall (mm^*)	Snow depth cm	EC Min	EC 25 %	EC Median	EC 75 %	EC Max	EC Mean	EC Std.
Nov 2009–Apr 2010	23	160	19.3 ± 8.8	7	171	389	674	3165	527	645
Nov 2010–Mar 2011	16	46	14.6 ± 4.6	61	238	569	960	1512	586	428
Nov 2011–Apr 2012	18	107	9.4 ± 2.8	101	305	919	3020	7636	1930	2331
2009–2012	57	104	17.2 ± 7.7	7	207	530	942	7636	987	1510
Fresh snow	19			7	76	171	237	790	215	169
Aged snow	38			81	530	840	1066	7636	1418	1728

* Millimeter (mm) in snow water equivalent.

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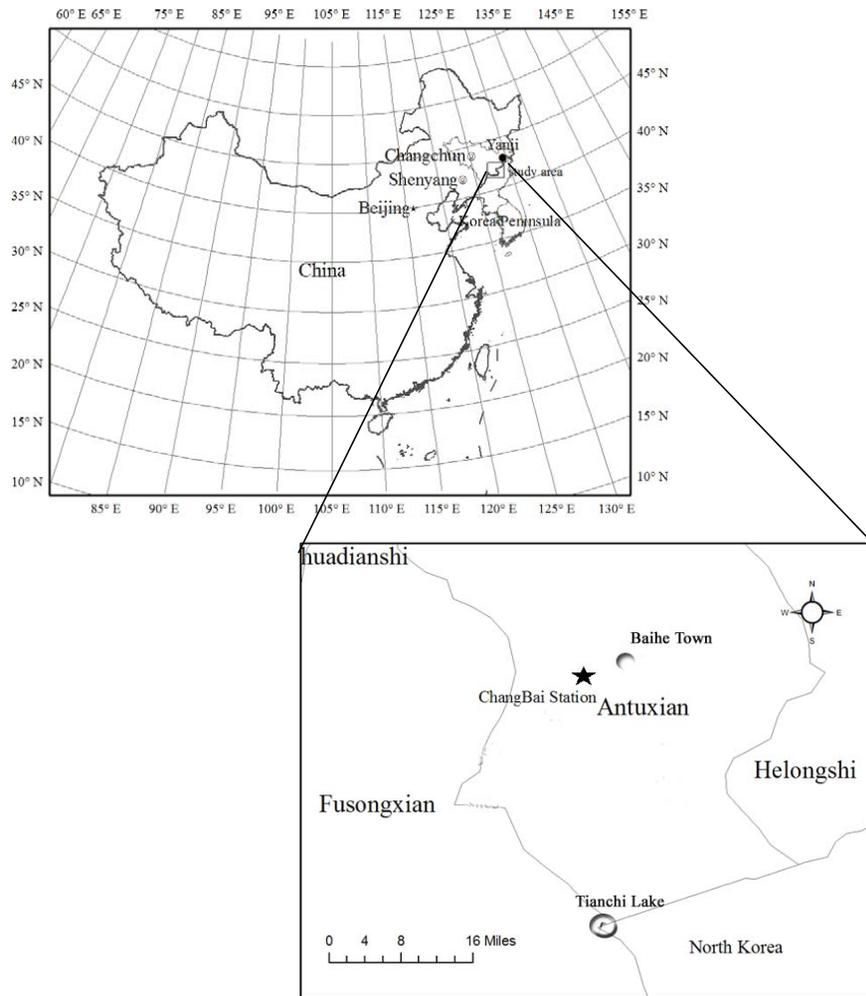


Fig. 1. Location of Changbai station in Northeastern China.

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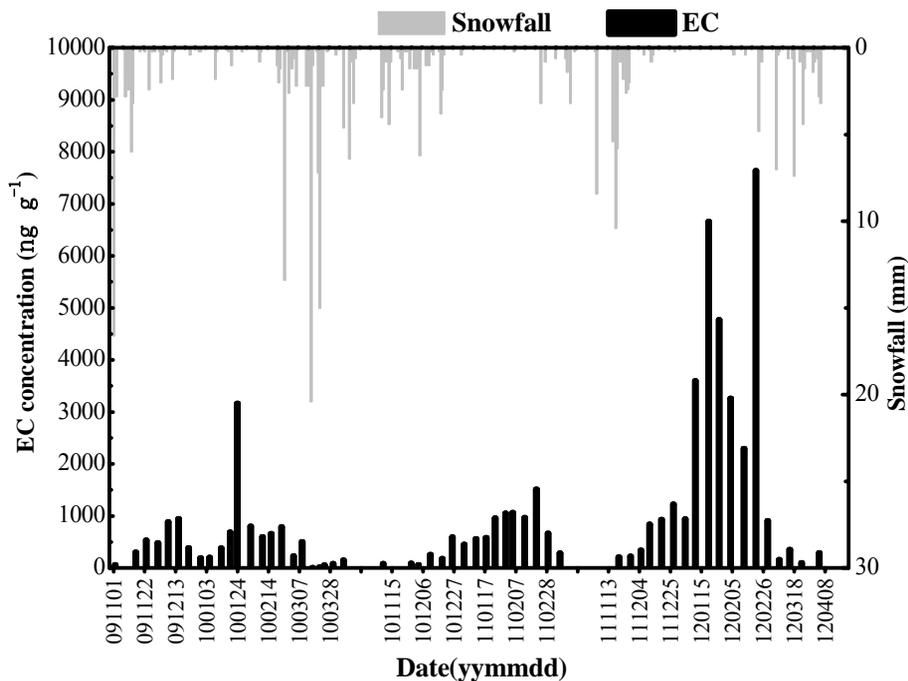


Fig. 2. EC concentrations in surface snow (ng g^{-1} , weekly sampling) and daily snowfall in mm water equivalent for winter season 2009 to 2011.

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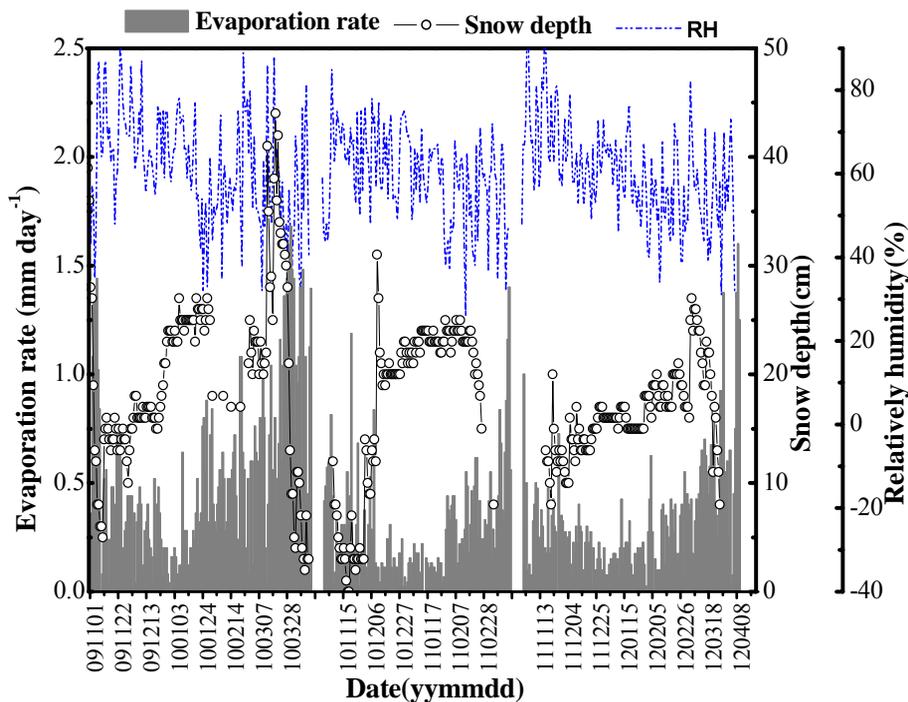


Fig. 3. Daily means of evaporation rate, snow depth and relatively humidity in winter season 2009 to 2011.

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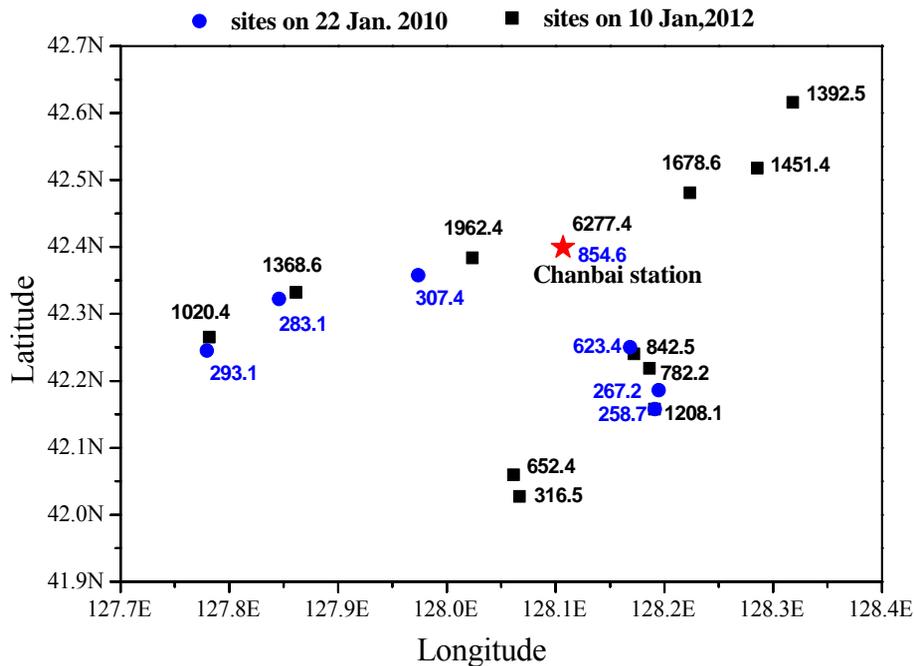


Fig. 4. EC concentrations (ng g^{-1}) around Changbai Mountain from the surveys in January 2010 and 2012.

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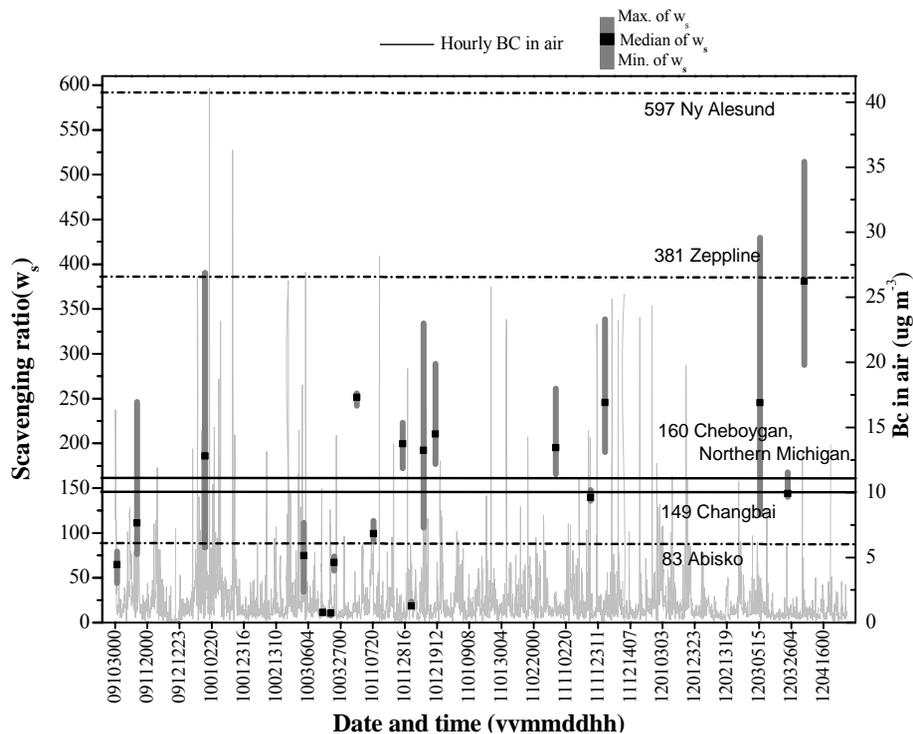


Fig. 5. Hourly mean concentrations of BC ($\mu\text{g m}^{-3}$) in air and EC scavenging ratio from 2009 to 2012. (Data from Zeppelin and Ny-Alesund are from Hegg et al., 2011, Abisko from Noone and Clarke, 1988, and Cheboygan from Cadle and Dasch, 1988.)

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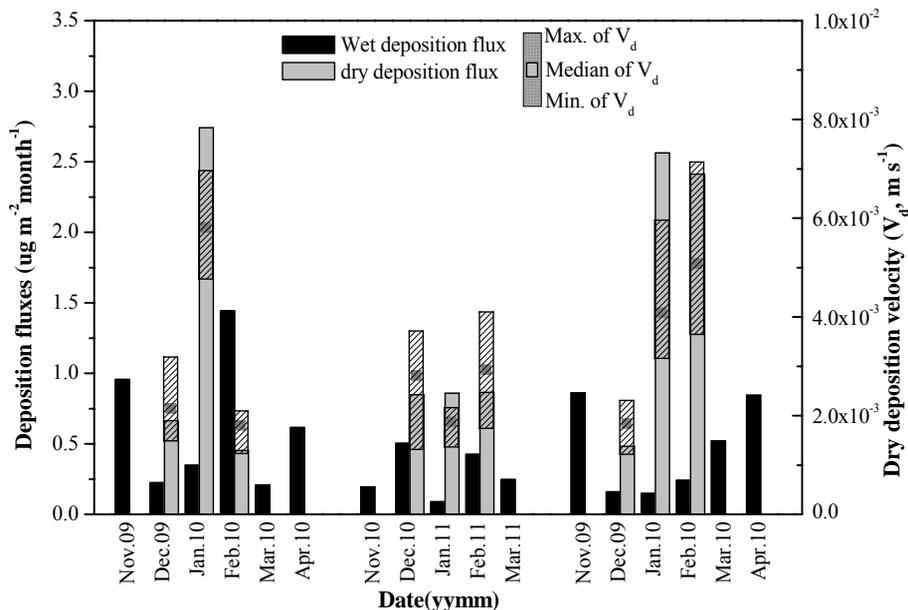


Fig. 7. Dry and wet deposition fluxes, and dry deposition velocity (V_d) of EC on snow surface in winter seasons from 2009 to 2011.

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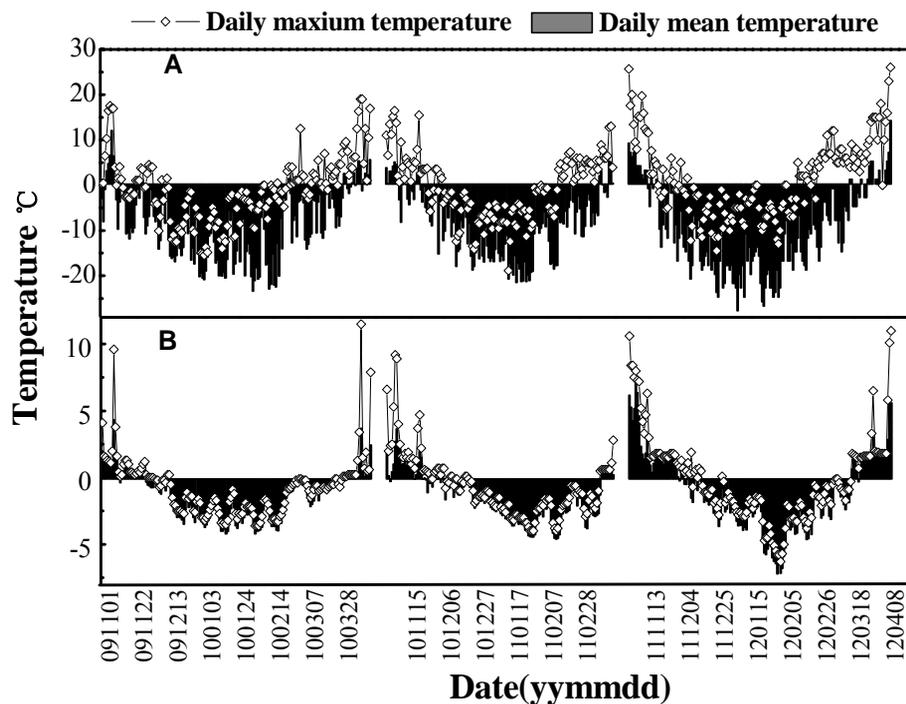


Fig. 8. Daily mean and maximum temperatures of air temperature near snow surface **(A)** and snow-soil interface **(B)** from 2009 to 2012.

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