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On board measurement of black carbon aerosols over the Arctic Ocean in summer

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Abstract This paper presents aerosol black carbon (BC) concentrations measured at deck level on board the R/V XUE LONG icebreaker. The vessel cruised the Arctic Ocean carrying an in situ aethalometer during the summers of 2008 and 2010. The courses of the third Chinese National Arctic Research Expedition (3rd CHINARE-Arctic, August 2008) and fourth Chinese National Arctic Research Expedition (4th CHINARE-Arctic, from late July to August 2010) were bounded by $173^{\circ}W-143^{\circ}W$ and $178^{\circ}E-150^{\circ}W$, with northernmost points $85^{\circ}25'N$ and $88^{\circ}26'N$, respectively. Results show low surface BC concentrations over the ocean throughout the courses, with means (standard error) of $6.0~(\pm 4.7)~\rm ng\cdot m^{-3}$ for 3rd CHINARE-Arctic, and $8.4(\pm 7.1)~\rm ng\cdot m^{-3}$ for 4th CHINARE-Arctic. It is clear that these onboard BC concentrations are similar to reported data from coastal stations in the Arctic region. The latitude-average BC concentration varied from $3.0-26.2~\rm ng\cdot m^{-3}$ for 3rd CHINARE-Arctic, to $4.2-20.5~\rm ng\cdot m^{-3}$ for 4th CHINARE-Arctic. At latitudes higher than $72^{\circ}N$ for 3rd CHINARE-Arctic and $75^{\circ}N$ for 4th CHINARE-Arctic, BC concentrations were lower and had negligible latitudinal gradients. Analysis indicates that the presence of the Arctic front isolates the lower atmosphere of the high-latitude Arctic Ocean from low-latitude terrestrial transport. This maintains the very low BC concentrations and negligible concentration gradients at high latitudes of the Arctic front in 2010 was further north than in 2008, which caused different latitudinal variation of BC concentration in the two years.

Keywords Chinese National Arctic Research Expedition, black carbon, aerosol, observation

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0 Introduction

Although it is well known that atmospheric aerosol plays an important role in global and regional climate systems^[1-2], there is still greater uncertainty about aerosol impacts than about greenhouse gases. This uncertainty can be attributed to the complexity and tempospatial variability of aerosol composition, physical and chemical properties, as well as their various direct and indirect effects on global and regional climate systems^[3-4]. Aerosol observations over the oceans are lacking com-

pared with ample long-term observations from worldwide land-based networks, leading up to the wide use of proxy data from satellite sensing^[5-6]. Hence, direct in situ observations over the ocean are crucial both for the "truing" of satellite data and for the model constraint, to enhance our understanding on the behavior of aerosol in their radiative forcing over vast marine areas.

Recent studies indicate that, in the context of global warming, the warming of the Arctic is about two times faster than the global average^[7], resulting in rapid

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shrinking of Arctic sea ice extent^[8]. In addition to longlived greenhouse gases, short-lived atmospheric species, including black carbon (BC) aerosols, are considered as the key forcers contributing to the rapid changes in $Arctic^{[9-10]}$. Though BC only accounts for a small portion of aerosol mass, it is the most important optical absorbent constituent $^{[4,11-12]}$. Studies have also shown that BC has a positive radiative forcing effect. When mixed with sulfate and other aerosols, BC can greatly reduce the negative forcing effect of aerosols, which can be very significant in the Northern Hemisphere with higher surface albedo. Model study has pointed that during the Arctic spring, the total radiative forcing of BC aerosols in the lower troposphere can be as high as 1.6 $W \cdot m^{-2[13]}$. In summer, this can reach 1.0 $W \cdot m^{-2}$. In addition, BC on Arctic sea ice can reduce surface albedo, producing radiative forcing effects of 0.53 W·m⁻² and 0.21 W·m⁻² in spring and summer, respectively. Therefore, the contribution of BC to Arctic warming is nearly four times that to the average global warming rate, reaching $0.6^{\circ}\text{C}\cdot\text{a}^{-1}$ [14]. BC is the most important contributor, only next to carbon dioxide, to the warning of Arctic. Therefore, similar to the concern over "Arctic haze" during the 1970s and 1980s, BC is an important factor in the rapid changes of climate and environment in the $Arctic^{[15]}$.

Since 1999, China has made Arctic scientific expeditions (Chinese National Arctic Research Expedition, CHINARE-Arctic) [16–18]. On the routes of these expeditions, Chinese scholars have carried out observational research on a variety of atmospheric components, including aerosol. During the 3rd and 4th CHINARE-Arctic, the authors made onboard BC measurements with an aethalometer, to obtain first-hand information for enhancing understanding of the mechanisms of rapid change in Arctic climate and environment. These observations from the summers of 2008 and 2010 are reported here, and a back trajectory analysis is applied to discuss the air mass transport from surrounding lands to the Arctic Ocean.

1 Observation

1.1 The route

During the 3rd and 4th CHINARE-Arctic, the R/V $XUE\ LONG$ icebreaker entered and left the Arctic Ocean

through the Bering Strait. Its route is illustrated in Figure 1. In the 3rd CHINARE-Arctic, the vessel entered the ocean on 3 August 2008, and surveyed marine and sea ice in the Chukchi Sea and Canada Basin. From 21 to 29 August, the XUE LONG parked at 85°N to carry out a comprehensive sea ice survey. It exited the Arctic Ocean on 8 September. The extent of the 3rd CHINARE-Arctic voyage included the area north of 70°N, from 73°W to 143°W. The vessel sailed to its most northerly position of 85° 25′N during the 3rd CHINARE-Arctic. During the 4th CHINARE-Arctic, it entered the ocean on 22 July 2010, and parked at 87°N from 8 to 19 August, then exited on 30 August. The 4th CHINARE-Arctic voyage extent embraced the area north of 70°N, from 178°E to 150°W, with northernmost point 88°26′N.

1.2 Observation instrument

During the 3rd and 4th CHINARE-Arctic, an aethalometer (Model AE31, by Magee Scientific Co.) was used. The aethalometer uses an LED light source with seven wavelengths (central wavelengths 370, 470, 520, 590, 660, 880 and 950 nm). The light beam irradiates a sample collected on quartz filters, and light attenuation by the sample is measured. Given the assumption that optical absorption of non-black-carbon components within aerosols can be ignored relative to BC, BC concentration is calculated according to the attenuation increment, sample volume and BC mass absorption coefficient^[11]. To reduce the shading effect of accumulated aerosol samples on the quartz filters, the threshold attenuation value of filter renewal is set at 75%, and the light source stability upon each replacement of filter renewal is examined. To ensure measurement stability in low-BC air at sea, an external pump was used for a high sampling flow rate of 8–9 L·min⁻¹. The instruments were installed in the meteorological room, under the forecastle cab of the XUELONG. The sampling inlet used specific plastic tubes of 3/8 inch (1 inch=0.025 m) diameter, and extended to the left rear of the sample observation deck atop the cab. The air inlet was about 1.5 m higher than the deck fence. Five-minute averages were recorded with a data logger.

1.3 Data processing

There are no significant sources of BC over oceans, except for areas close to land or along busy shipping routes. After long-distance transport, air masses originating from land areas may attain a relatively even admixture state above oceans, so ocean BC concentrations should be low and of insipidness. However, emissions from the research ship may interfere with onboard measurements. When the vessel sails at low speeds, stops or turns, its emissions produce sharp but short-term disturbing on measurements of BC concentration, with peak values hundreds of times greater than undisturbed data. To distinguish and remove these concentration artifacts in the data, a "smooth baseline" was used as a reference, in combination with data on vessel speed, course, and relative wind speed. This baseline is obtained by applying a moving spline smoothing method to the five-minute BC dataset. Disturbed data with values 3-sigma (hourly standard deviation) higher than baseline in the 5-minute original record were excluded. Next, the hourly average value was calculated. When there were fewer than four five-minute data points within an hour, hourly average concentration was not calculated. The sailing durations north of 70°N during the two expeditions were 871 h and 943 h, respectively, from which 778 h and 690 h of valid data were obtained. This represents data integrity rates of 89.0% and 73.2%.

2 Results and discussion

2.1 Variation of hourly average concentration

Figure 1 shows hourly average BC concentrations over the Arctic Ocean, obtained during the two expeditions. The western routes were departing the Arctic, and the eastern ones were arriving. Given that invalid data eliminated by the data quality check (Section 1.3) are essentially those collected during turning, stopping and low-speed sailing of the research vessel, Figure 1 can still show the integral spatial continuity of hourly average BC concentrations. BC concentrations were within the range from 0 to 40 ng·m⁻³, with gentle variation. Concentrations near low-latitude land areas were relatively high, and those over high-latitude ocean interior areas

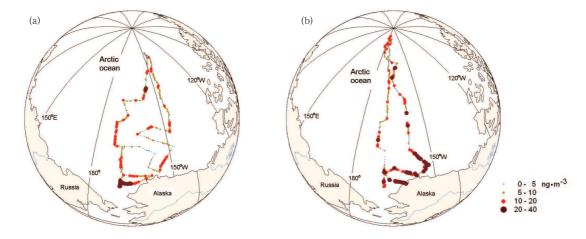


Figure 1 Hourly average concentrations of black carbon aerosols along the route. (a) The 3rd CHINARE-Arctic; (b) the 4th CHINARE-Arctic.

were low.

Statistics in Table 1 demonstrate that more than 70% (84.8% for 3rd CHINARE-Arctic and 75% for 4th CHINARE-Arctic) of BC concentration were below $10~\rm ng\cdot m^{-3}$. Concentrations during the 3rd CHINARE-Arctic were generally lower than the 4th CHINARE-Arctic.

2.2 Latitudinal variation of black carbon concentrations

Latitudinal BC concentrations from the two expeditions

are shown in Figure 2. The concentrations from the 3rd CHINARE-Arctic were in range of $3.0{\text -}26.2~{\rm ng\cdot m^{-3}}$, and those from the 4th CHINARE-Arctic were in range of $4.2{\text -}20.5~{\rm ng\cdot m^{-3}}$. Although the routes of the XUE LONG were confined to a limited latitude range for both expeditions, sharp drops in BC concentration occurred near $72{^\circ}{\rm N}$ in 3rd CHINARE-Arctic and $75{^\circ}{\rm N}$ in 4th CHINARE-Arctic. In higher-latitude seas, the latitudinal average concentrations were low and varied little, i.e., there were negligible latitudinal gradients. Zonal average BC concentrations for both expeditions to the

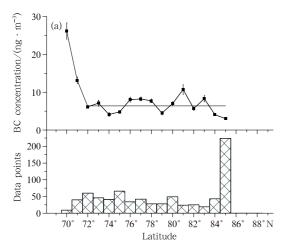
high-latitude Arctic (north of 72°N during 3rd CHINARE-Arctic, north of 75°N during 4th CHINARE-Arctic) were similar, with values of 6.4 $\rm ng\cdot m^{-3}$ and 6.9 $\rm ng\cdot m^{-3}$, respectively. This indicates that higher latitudes are more isolated from airmass transport from land areas than lower latitudes.

A prior study has demonstrated that the Arctic front plays a significant role in blocking the transport of low-latitude pollutants to the lower atmosphere over the Arctic Ocean^[19]. In winter, this front may extend to areas near 40°N, and recede north to 65°N–70°N in summer. This seasonal displacement of the front can vary from year to year, and it may vary longitudinally. When isolated by the Arctic front, the lower atmosphere in the Arctic tends toward a well-mixed and relatively homogeneous state. Modeling with the Lagrangian method calculates transport between the Arctic lower atmosphere

and low-latitude areas^[20]. This has indicated that the life span of the Arctic airmass(i.e., continuous residence time in the area north of 70°N) is shorter in winter, averaging one week, increasing in summer to about two weeks. From a geographic standpoint, this life span is longest in the sectors 90°W-180°W (winter) and 90°W-180°W-135°E (summer). The routes during the 3rd and 4th CHINARE-Arctic were within the sector 140°W–180°W, which is situated in the ocean area with the longer-lived Arctic atmosphere, as cited in the literature^[20]. When the route of vessel reached high latitudes of 70°N or greater, where the impact of low-latitude transport is much smaller, there were very low BC concentrations and negligible concentration gradients, and the atmosphere was more evenly mixed. Thus, the Arctic front is important to maintaining BC aerosols at low concentrations in the Arctic Ocean during summer.

Table 1 Statistics of hourly average concentrations of BC aerosols over the Arctic Ocean

	Average $(\pm 1\sigma)/(\text{ng}\cdot\text{m}^{-3})$	$Median/(ng \cdot m^{-3})$	Concentration bins/(ng·m ⁻³)			
			0–5	5-10	10-20	20-40
3rd CHINARE-Arctic	$6.0 {\pm} 4.7$	4.7	53.0%	31.9%	13.1%	1.9%
4th CHINARE-Arctic	$8.4{\pm}7.1$	6.6	37.1%	37.9%	17.2%	7.7%



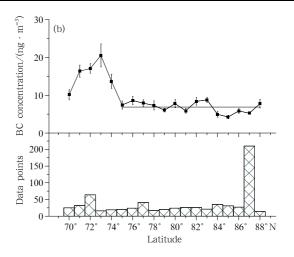


Figure 2 Black carbon concentrations over seas at different latitudes. (a) The 3rd CHINARE-Arctic; (b) the 4th CHINARE-Arctic. In the top panels, solid squares (■) denotes latitudinal average value, vertical solid line the standard error, and horizontal line the zonal average value at high latitudes.

2.3 Comparison with observational results of surrounding coastal stations

Since the 1980s, BC has continuously attracted concerns from scientific community as its role in the Arctic climate and environment^[21–22]. There have been observation

campaigns of atmospheric BC concentration in the Arctic using aircraft, and BC measurements in Greenland ice. In addition, several monitoring stations in coastal areas surrounding the Arctic Ocean started long-term BC monitoring $^{[23-25]}$. Chinese scientists also measured BC at Chinese Yellow River Station in the Arctic, during

the summers of 2005 and 2008^[23-24]. These observations revealed that there is a clear seasonal variation of BC concentration in the Arctic atmosphere, with highest concentrations in the Arctic haze period (late winter and early spring) and lowest in summer. The observations of Greenland ice show that in the early part of the last century, BC concentrations peaked, followed by a decreasing tendency. Observations at three long-term Arctic coastal stations (Alert in northeastern Canada near

Greenland, Barrow in Alaska and Zeppelin in northern Norway) also show that in the last one to two decades, BC concentration declined at varying rates. However, these observations are limited to coastal areas or islands. No observations focused on the vast Arctic seas.

In Table 2, observational results from several Arctic coastal stations are listed for comparison to this work. It is seen that BC concentrations over Arctic seas are similar to those observed at coastal stations.

Table 2 Black carbon aerosol concentrations at Arctic land sites and Arctic Ocean

Site	Observation period	BC concentration/ $(ng \cdot m^{-3})$	Ref.
Alert, Canada 82.45°N, 62.52°W, 210 m	2004–2005, August	Average: $14.3(\pm 14.0)$, Median: 9.2	Note 2
$\begin{array}{c} \rm Barrow^{Note~1},~USA \\ 71.32^{\circ}N,~156.61^{\circ}W,~210~m \end{array}$	2006–2007, August	Average: $3.4(\pm 5.7)$, Median: 2.1	Note 2
Zeppelin, Norway 78.90°N, 11.88°E, 474 m	1998–2007, June–September	Average: 0–10	18
Chinese Yellow River Station 78.9° N, 11.9° E, 10 m	2005–2008, Summer	15	14
Arctic Ocean 70.0° N-85.42° N, 143° W-173° W	2008, August	19.7 (Average, south from 71°N) 6.4 (Average, north from 72°N) 8.0 (Average, north from 70°N)	This work
Arctic Ocean 70.0°N–88.43°N, 150°W–178°E	2010, July–August	15.6 (Average, south from 74°N) 6.9 (Average, north from 75°N) 9.2 (Average, north from 70°N)	This work

Note 1: Barrow Station used the Particle Soot Absorption Photometer for observation, which is different from the other stations and this work. Directly measured data are aerosol absorption coefficient (Labs). Labs are converted to equivalent black carbon concentration. (EBC), via the empirical conversion formula EBC ($ng \cdot m^{-3}$) = Labs ($ng \cdot m^{-1}$)×1 000/19($ng \cdot g^{-1}$). This formula is from the National Oceanic and Atmospheric Administration/Global Monitoring Division, USA.

Note 2: World Meteorological Organization World Data Centre for Aerosols, http://wdca.jrc.it/.

2.4 Impact from terrestrial transport

Black carbon aerosols originate from the combustion of carbonaceous material. Over the ocean, there are no large-scale sources of BC emissions save those from individual navigation. Thus, terrestrial transport is the dominant driving force on BC concentration magnitudes and variations over the ocean, especially in the Arctic, which is surrounded by the massive North American and Eurasian continents. To evaluate the impact of terrestrial transport on BC concentrations in the Arctic Ocean, the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) 4.9 model (http://www.arl.noaa.gov/HYSPLIT.php) was used to calculate 96-hour airmass back trajectories at 00, 03, 06, 09, 12, 15, 18 and 21 GMT along the route, using a

National Centers for Environmental Prediction dataset. The initial calculation height was 50 m. Following the discussion in Section 2.2, the trajectories of southern routes and northern routes (both routes from 72°N for 3rd CHINARE-Arctic, from 75°N for 4th CHINARE-Arctic) are shown by blue and cyan colors in Figure 3.

During the 3rd CHINARE-Arctic, the airmass back trajectories for the XUE LONG tended to lie in high-latitude waters, with a few exceptions flowing from/through coastal areas of Alaska and Russia for the route south of 72°N. During the 4th CHINARE-Arctic, many southern route trajectories (south of 75°N) flowed from/through coastal areas of Alaska and Russia, some extended deep into Alaska. During the vessel's return route of the 4th CHINARE-Arctic from 78°N to 70°N, airmass trajectories extended further south, into the

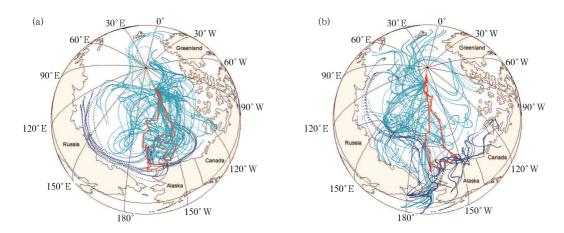


Figure 3 4-day airmass back trajectories of the two expeditions. Left panel shows airmass back trajectories for the 3rd CHINARE-Arctic, right panel for the 4th CHINARE-Arctic. Red dots show route of the XUE LONG. Cyan dots show trajectories for northern routes. Blue dots show trajectories for southern routes (both routes from 72°N for the 3rd CHINARE-Arctic, from 75°N for the 4th CHINARE-Arctic).

Bering Sea area. By comparing the back trajectories for the XUE LONG during the 3rd CHINARE-Arctic and 4th CHINARE-Arctic, it may be concluded that the Arctic front in 2010 was further north than in 2008, which caused different latitudinal variations of BC concentration in the two years.

3 Conclusions

During the 3rd CHINARE-Arctic (August 2008) and 4th CHINARE-Arctic (July–August 2010), black carbon (BC) aerosol concentrations in the Arctic Ocean between 140° W and 180° W were continuously observed. The observations that were furthest north were at $85^{\circ}25'$ N and $88^{\circ}26'$ N. The observation and analysis demonstrate that:

- (1) Concentrations of BC aerosols in the Arctic Ocean are relatively low. Average concentrations north of 70°N during the two expeditions were 6.0 (± 4.7) and 8.4 (± 7.1) ng·m⁻³. Compared to limited data from coastal stations around the Arctic, the concentrations reported here are similar to those from the coastal stations in summer, reported in the literature.
- (2) Latitudinal average BC concentrations in the 3rd CHINARE-Arctic from 70°N to 85°N were between 3.0–26.2 $\rm ng\cdot m^{-3}$; corresponding concentrations in the 4th CHINARE-Arctic from 70°N to 88°N were between 4.2–20.5 $\rm ng\cdot m^{-3}$. BC concentration dropped sharply with latitude around 72°N in 2008, and 75°N in 2010. In the high-latitude seas north of these latitudes, zonal average BC concentrations during both expeditions were

very similar. Analysis indicates that the presence of the Arctic front isolates the lower atmosphere over the high-latitude Arctic Ocean from the impacts of low-latitude terrestrial transport. This maintains very low BC concentrations and a negligible concentration gradient over the high-latitude ocean during summer.

(3) Calculated airmass back trajectories during the two expeditions showed the Arctic front in 2010 to be further north than in 2008, causing different latitudinal variation of BC concentration in those two years.

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