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The identification of source regions of black carbon at a receptor site off the eastern coast of China

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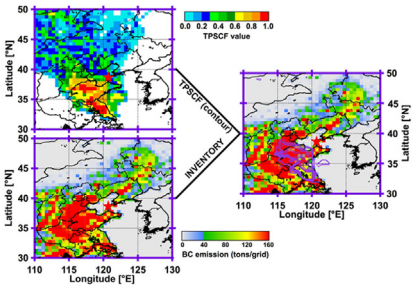
HIGHLIGHTS

- Source regions of black carbon (BC) in a regional coast site of China were identified.
- Source regions were identified by total potential source contribution function (TPSCF).
- TPSCF values were consistent with BC emission rates in the emission inventory.
- The residential source had a significant effect on BC during the heating period.

GRAPHICAL ABSTRACT



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ABSTRACT

The black carbon (BC) mass concentration and the particle chemical compositions were continually measured at Changdao Island, which is a regional receptor site off the eastern coast of China. This island is in the transport passage of the continental outflow to the Pacific Ocean when the East Asia monsoon prevails in the winter and spring. The campaign period was for March and April 2011, which corresponded to heating and non-heating periods in northern China. The effect of BC emission source regions on BC measured at Changdao Island between the heating and non-heating periods was determined by integrating the total potential source contribution function (TPSCF) model with the new monthly emission inventory in 2010 and the fire counts retrieved from MODIS during the campaign. BC concentrations were determined to be highest for similar times of day for both the heating and non-heating periods: 4.27 μg m^{-3} at 8:00 AM and 3.06 μg m^{-3} at 9:00 AM, respectively. The probable source regions for BC were primarily located in Shandong and Jiangsu provinces (and in other neighboring provinces) for both periods. However, the source regions for the non-heating period extended more to the north and southwest than those of the heating period. TPSCF values were correlated with the emission rates from residential, industry, transportation, and power plants sources in the anthropogenic emission inventory. This correlation provides an indirect and qualitative process to verify the emission inventory. In the heating period, the predominant source was the residential source in the emission inventory, and this

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source had a significant effect on the BC concentration. The differing peak concentrations between the two periods may be observed because of the increased residential heating during the heating period, which suggested that the measures employed by the government and environmental managers to reduce the emissions of pollutants should be stricter in the identified source regions during the heating period.

1. Introduction

Black carbon (BC), brown carbon (BrC), and dust are the three types of atmospheric aerosols that can absorb solar radiation and have direct radiative effects (Bond et al., 2013). Among these aerosols, BC has unique properties and plays an important role in climate systems. BC is produced by the incomplete combustion of fossil fuel, wood, and other carbon fuels. Residential sector, industry, transportation, and power plants are major anthropogenic sources of BC (Zhang et al., 2009). Once BC is emitted to the atmosphere, the aerosol eventually ages and internally mixes with other components and forms cloud condensation nuclei (CCN) to indirectly effect radiative forcing (Bond et al., 2013).

Among these anthropogenic emission sources of BC, the predominant source in a developed continent such as North America and Europe is transportation, whereas the residential source dominates in developing continents such as Africa and most of Asia (Bond et al., 2004; Streets et al., 2003; Venkataraman et al., 2005; Wang et al., 2012). Asia is the leading contributor of BC in the world (Zhang et al., 2009). The emissions of BC and other pollutants from this region could significantly influence the air quality in downwind regions, especially when the East Asia northwesterly monsoon prevails. Numerous ground, airborne, and satellite-based observations and modeling studies focused on the influence of the Asian continental outflow (e.g. Huebert et al., 2003; Peltier et al., 2008; Wang et al., 2009a). Though the eastern coastal zone of China is in the passage for the Asian continental outflow, field observations based on islands or cruises in this research hotpot are currently lacking.

Changdao Island was one of the receptor sites in the CAPTAIN (Campaign of Air PolluTion At INshore Areas of Eastern China) campaign, which focused on studying the influence of East Asia continental outflow on the air quality in downwind regions. Only a few studies at this site used off-line sampling and analysis (Adams and Liu, 2009; Feng et al., 2007, 2012), and a highly resolved dataset of the aerosol composition is scarce. Our previous publications about the campaign at Changdao Island have focused on the formation and evolution of secondary organic aerosols (SOA) (Hu et al., 2013; Yuan et al., 2013). In this study, we focus on the primary emissions by using BC as a representative aerosol and try to address the following questions: (1) which source regions potentially contribute to the BC concentration measured at Changdao Island; and (2) which sources from these regions have a significant effect on the BC concentration at this receptor site. To address these questions, the receptor model, the newly developed emission inventory and satellite retrieved fire counts were utilized. As demonstrated by many inventories (Liu et al., 2009a; Wang et al., 2012; Zhang et al., 2009), the predominant BC source in China is residential. Our observations were conducted during both heating and non-heating periods in northern China, enabling us to study the different effects of the residential source on the BC concentration between two periods at Changdao Island.

2. Measurements and methodology

2.1. Measurements and sampling site

BC mass concentrations were continually measured by a mutliwavelength Aethalometer (AE-31, Magee Scientific, USA) at Changdao Island (120.74° E, 37.92° N) from 20 March to 24 April, 2011. An Aerodyne Aerosol Mass Spectrometer (AMS) was also deployed to measure the mass concentration of the PM_1 chemical composition. A detailed description of this instrument and the exclusion criteria of data influenced by local coal combustion and biomass burning events can be seen in the previous study and its supplemental material (Hu et al., 2013). Changdao Island is located near the demarcation line between the Bohai and Yellow Seas off the eastern coast of China (Fig. 1). The upwind regions from the island are the provinces of Shandong, Hebei, and Liaoning and the municipalities of Beijing and Tianjin, which are regions with significant economic growth and substantial air pollutant emissions. As illustrated in Fig. 1, these regions emit a large amount of BC.

2.2. Total potential source contribution function (TPSCF)

Ashbaugh et al. (1985) developed a statistical method called residence time analysis in 1985. Based on this method, Malm et al. (1986) established the potential source contribution function (PSCF) model in 1986. Cheng et al. (1993) modified this method by integrating air trajectories from different endpoint heights and called it total PSCF (TPSCF). PSCF combines the aerosol chemistry and air mass transport information to identify possible source regions or dominant transport pathways for a receptor site. This method has been widely used to study the source regions of aerosols on regional scales or the long-range transport of air pollutants to a receptor site (Hopke et al., 1995; Kedia et al., 2012; Wu et al., 2009; Zhu et al., 2011).

For TPSCF analysis, air-mass back trajectories from the previous 72 h are determined by using the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998) at four different endpoint heights (50 m, 100 m, 500 m, and 1000 m) and a time interval of 1 h for each day. After that, the geographic regions covered by the air trajectories are divided into an array of 0.5° × 0.5° grid cells, and for every grid cell at latitude i and longitude j, the number of the trajectory segment endpoints terminating within the cell is counted. Because the air masses originate from different heights, calculating the total potential source contribution function (TPSCF) is more reasonable than the PSCF. The TPSCF at the ijth grid cell can be calculated by the following (Hopke et al., 1995; Kedia et al., 2012):

$$\mathrm{TPSCF}_{ij} = \frac{\sum m_{ij}^{k}}{\sum n_{ij}^{k}}$$

where the number of the trajectory segment endpoints over the ijth grid cell for height k is counted as n_{ij}^{k}, and the number of these endpoints corresponding to the pollutant concentration higher