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High resolution of black carbon and organic carbon emissions in the Pearl River Delta region, China

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HIGHLIGHTS

- BC and OC emissions are mainly from mobile sources and residential combustion.
- Primary OC/BC ratios show strong associations with economic development levels.
- Gridded BC and OC emissions are developed and ready for air quality model use.
- Higher uncertainties in OC and BC emission estimates than in other primary pollutants.

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ABSTRACT

A high-resolution regional black carbon (BC) and organic carbon (OC) emission inventory for the year 2009 was developed for the Pearl River Delta (PRD) region, China, based on the collected activity data and the latest emission factors. $PM_{2.5}$, BC and OC emissions were estimated to be 303 kt, 39 kt and 31 kt, respectively. Industrial processes were major contributing sources to $PM_{2.5}$ emissions. BC emissions were mainly from mobile sources, accounting for 65.0%, while 34.1% of OC emissions were from residential combustion. The primary OC/BC ratios for individual cities in the PRD region were dependent on the levels of economic development due to differences in source characteristics, with high ratios in the less developed cities and low ratios in the central and southern developed areas. The preliminary temporal profiles were established, showing the highest OC emissions in winter and relatively constant BC emissions throughout the year. The emissions were spatially allocated into grid cells with a resolution of 3 km × 3 km. Large amounts of BC emissions were distributed over the central–southern PRD city clusters, while OC emissions exhibited a relatively even spatial distribution due to the significant biomass burning emissions from the outlying area of the PRD region. Uncertainties in carbonaceous aerosol emissions were usually higher than in other primary pollutants like SO_2, NO_x, and PM_{10}. One of the key uncertainty sources was the emission factor, due to the absence of direct measurements of BC and OC emission rates.

1. Introduction

Carbonaceous aerosol emissions have recently attracted great attentions because of the unfavorable effects on human health (Saikawa et al., 2009), visibility (Seinfeld, 2008) and climate (Ramanathan and Carmichael, 2008). Primary carbonaceous particles are mainly emitted from incomplete combustion of fossil fuels and biomass, and can be divided into black carbon (BC) and organic carbon (OC) fractions (Venkataraman et al., 2005). BC is the light absorbing composition of the particles. OC scatters light and consists of different organic compounds (Chuang and Seinfeld, 2002; Roden and Bond, 2006).

In recent years, numerous efforts have been made to estimate the BC and OC emissions at different geographical scales, such as in global inventories (e.g. Bond et al., 2004, 2007) and at national scales in Europe and the United States (Kupiainen and Klimont, 2007; U.S. EPA, 2007). BC and OC emissions in China were significant, accounting for about 30% and 24% of global anthropogenic emissions, respectively (Bond et al., 2004). However, BC and OC emission inventories specifically targeting on China were not available until recent years. Streets et al. (2001) developed the first province-specific BC emission inventory and projections in China for the years 1995 and 2020, based on non-local emission factors and estimated activity data. Cao et al. (2006) upgraded carbonaceous aerosol emission inventories with seasonal variations. Several Asian emission inventories were holistically

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developed that covered China (Zhang et al., 2009; O'Hara et al., 2007; Klimont et al., 2009). Meanwhile, efforts were also made to develop local BC and OC emission factors, based on in-situ measurements of carbonaceous fraction (Ho et al., 2003) or field measurement of small coal stove combustion, biomass burning and vehicle sources (Chen et al., 2009; Cheng et al., 2010; Li et al., 2009).

As one of the fastest developing regions in China, the PRD region has been experiencing frequent pollution episodes with high concentrations of ozone and fine particulate matters (Zheng et al., 2010a). Previous studies discovered that carbonaceous aerosol accounted for about 20%–50% of total fine particulate mass in the PRD region (Niu et al., 2006; Huang et al., 2011), indicating the importance of carbonaceous aerosols in this region. OC and BC emissions are important information for investigating the formation and transport of atmospheric aerosols using air quality models (Yu et al., 2004). Although currently there are several nation-wide inventories that addressed carbonaceous aerosol emissions in Guangdong (e.g. Cao et al., 2006; Zhang et al., 2009), the low spatial and temporal resolutions limited their usage in the regional air quality modeling. Moreover, with the economic development and energy structure adjustment, previous inventories cannot properly or accurately represent the recent regional emission characteristics. Hence, spatially allocated and temporally resolved BC and OC emission inventories in the PRD region are urgently needed.

The objectives of this study are: (1) to estimate the PRD regional BC and OC emissions for the year 2009; (2) to analyze source contribution characteristics of BC and OC emissions; (3) to develop a model-ready high resolution of BC and OC emission inventories; and (4) to assess the associated uncertainties in emission estimates. With the emphasis on primary BC and OC, the fine particle emissions were also included in our inventories for a better understanding of fine carbonaceous emission characteristics.

2. Data and methods

2.1. Study domain

The PRD region, located in the central coast of Guangdong Province, is one of the most populated city clusters in China. It covers nine cities, including Guangzhou (GZ), Shenzhen (SZ), Dongguan (DG), Zhuhai (ZH), Jiangmen (JM), Foshan (FS), Zhongshan (ZS), and parts of Huizhou (HZ) and Zhaoqing (ZQ). The study domain is from 111.135°E to 115.669°E, and 21.530°N to 25.580°N, and spatially allocated into 16,720 grid cells with a resolution of 3 km×3 km.

2.2. Methods for estimating carbonaceous aerosol emissions

The emission sources were classified into six groups, including stationary combustion, industrial process, mobile, fugitive dust, open biomass burning, and waste incineration sources. Detailed sub-categories and the related data sources were listed in Table 1.

The emissions of BC and OC from a particular source were estimated based on corresponding emission factors and activity data by the following equation:

$$E_i = \sum_p A_p \times EF_{pi} \quad (1)$$

where i and p represent the pollutant type and specific sector or subsector, respectively; E is the annual emission of a pollutant; A is the activity level and EF denotes the emission factor. A bottom-up approach was used for emission sources where detailed activity data were available, such as power plants and industrial combustion, etc. For categories with only city- or region-wide statistical data available, such as residential combustion, mobile sources and fugitive dust, the top-down approach was adopted instead.

Table 1
Categories and subcategories for BC and OC emission sources.

Sector	Subsector	Sector	Subsector
Stationary combustion	Power plants[a]	Industrial processes	Brick production[a]
	Industrial combustion[a]		Glass production[a]
	Residential combustion[b,c]	Mobile sources	On-road mobile sources[b,d,e]
Industrial processes	Coke production[d]		Off-road mobile sources[b, d, f]
	Sinter plants[d]	Fugitive dust	Construction dust[b]
	Pig iron production[d]		Paved road dust[d,g]
	Steel production[d]	Open biomass burning	Field burning of crop residues[b,h]
	Cement production[a]		Forest fire[i]
	Lime production[a]	Waste incineration[j]	

[a] Data collected from Guangdong Provincial Pollutant Statistical Reports (GPPSR) 2009.
[b] Data collected from SBG(2010).
[c] Data collected from NBS (2010).
[d] Data collected from Statistics Bureau of Guangzhou/Shenzhen/Zhuhai/Foshan/Jiangmen/Dongguan/Zhongshan/Huizhou/Zhaoqing, 2010.
[e] Data collected from Che (2010).
[f] Data collected from EBYCTC (2010).
[g] Data collected from Research team of air quality assurance for Guangzhou Asian Games (2009).
[h] Data collected from He et al. (2011).
[i] Data collected from EBASYG (2010).
[j] MHURD (2010).

Activity data usually refers to fuel consumption for stationary combustion, product output for industrial processes, or vehicle kilometers traveled (VKT) for on-road mobile sources and others. Most activity data in this study can be directly obtained at the city level. For categories with only provincial data available, reasonable surrogates were chosen to determine the city-based data, like rural population, crop sown area etc.

The emission factors of carbonaceous aerosol were obtained directly from field measurements of residential combustion and open biomass burning (Table 2). For other sectors, emission factors were approximated by applying the mass fractions of BC and OC to $PM_{2.5}$, which can be illustrated by Eq. (2).

$$EF_{p,BC} = EF_{p,PM_{2.5}} \times f_{p,BC} \quad (2)$$

where EF_{BC} represents the emission factor of BC (or OC); $EF_{PM_{2.5}}$ is the emission factor of $PM_{2.5}$; and f_{BC} is the BC (or OC) fractional abundance in $PM_{2.5}$. Tables 3 and 4 presented the $PM_{2.5}$ emission factors and shares of BC and OC used in this study. The majority of BC/OC share values were obtained from Zhang (2005) and Lei et al. (2011), both of which made a comprehensive summary of BC/OC fractions on the basis of Kupiainen and Klimont (2004, 2007) and Bond et al. (2004).

The detailed methods for compiling activity data and determining emission factors were introduced below by category.

2.2.1. Stationary combustion

Stationary combustion sources involved three sectors (power plants, industry and residential) and eight types of fuel (coal, heavy oil, diesel, kerosene, natural gas, coke, liquefied petroleum gas (LPG), woody fuel and crop residues). Fuel consumption in power plants and industrial combustion was obtained from the Guangdong Provincial Pollutant Statistical Report (GPPSR) 2009, in which detailed activity data, including location (latitude and longitude), fuel type, fuel consumption, product output, ash content, removal efficiency and others, are available for each plant. For residential combustion, the fuel data were obtained from China Energy Statistical Yearbook 2010 (NBS, 2010) and Guangdong Statistical Yearbook 2010 (SBG, 2010).

In the PRD region, approximately 99% of coal boilers in the power plants and 60% of coal boilers for industry were equipped with Air