Science of the Total Environment 565 (2016) 519–528

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Long-term trends of chemical characteristics and sources of fine particle in Foshan City, Pearl River Delta: 2008–2014

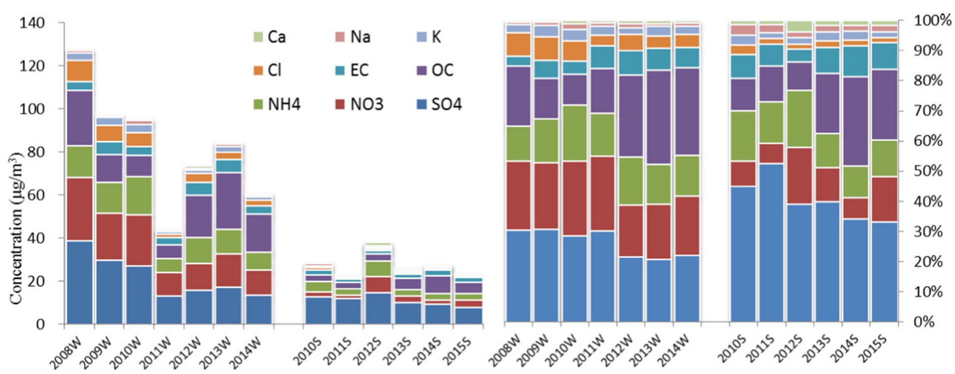
Jihua Tan [a,c,e], Jingchun Duan [b,c], Yongliang Ma [c,*], Kebin He [c], Yuan Cheng [c], Si-xin Deng [d], Yan-ling Huang [d], Shu-ping Si-Tu [d]

[a] Huairou Eco-Environmental Observatory, College of Resources and Environment, University of Chinese Academy of Sciences, Beijing 100049, China
[b] State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China
[c] School of Environment, Tsinghua University, Beijing 100084, China
[d] Foshan Environmental Protection Bureau, Foshan 528000, China
[e] Research Centre for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

HIGHLIGHTS

- Seven-years long characteristic of chemical and source of $PM_{2.5}$ were investigated
- $PM_{2.5}$ and most of chemical species decreased significantly from 2008 to 2014
- Sulfate and OC were the most important chemical species in 2008 and 2014
- Primary sources decreased significantly, but secondary sources increased.
- Continuous monitoring is an adequate strategy for air quality control policy

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 5 May 2015
Received in revised form 19 April 2016
Accepted 9 May 2016
Available online xxxx

Editor: D. Barcelo

Keywords:
Long-term
Source apportionment
$PM_{2.5}$
Chemical species
Pearl River delta

ABSTRACT

Foshan is a major international ceramic center and the most polluted city in the Pearl River Delta (PRD). Here we present the results of the first long-term $PM_{2.5}$ (particles <2.5 μm) sampling and chemical characterization study of the city. A total of 2774 samples were collected at six sites from 2008 to 2014, and analyzed for water soluble species, elements and carbonaceous species. The major constituents of $PM_{2.5}$ were sulfate, OC (Organic Carbon), nitrate, ammonium and EC (Elemental Carbon), which accounted for 50%–88% of $PM_{2.5}$. $PM_{2.5}$ and the most abundant chemical species decreased from 2008 to 2011, but rebounded in 2012–2013. After 2008, the chemical composition of $PM_{2.5}$ changed dramatically due to the implementation of pollution control measures. From 2008 to 2011, SO_4^{2-} and NO_3^- were the two largest components; subsequently, however, OC was the largest component. The respective contributions of SO_4^{2-}, NO_3^- and OC to the sum of water soluble species and carbonaceous species were 30.5%, 22.9% and 19.9% in 2008; and 20.2%, 16.5% and 30.2% in 2014. Distinct differences in nitrate and sulfate, and in mass ratio $[NO_3^-]/[SO_4^{2-}]$ imply that mobile sources tended to more important in Foshan during 2012–2014. The results indicate that pollution control measures implemented during 2008–2014 had a large effect on anthropogenic elements (Pb, As, Cd, Zn and Cu) and water soluble species, but little influence on crustal elements (V, Mn, Ti, Ba and Fe) and carbonaceous species. The PMF method was used for source apportionment of $PM_{2.5}$. Industry (including the ceramic industry and coal combustion), vehicles and dust were the three most

* Corresponding author at: State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China.
E-mail address: Liang@tsinghua.edu.cn (Y. Ma).

http://dx.doi.org/10.1016/j.scitotenv.2016.05.059

important sources and comprised 39.2%, 20.0% and 18.4% of $PM_{2.5}$ in 2008, respectively. However, secondary aerosols, vehicles and industry were the three most important sources and comprised 29.5%, 22.4% and 20.4% of $PM_{2.5}$ in 2014, respectively. During the seven year study interval, the contributions of primary sources (industry and dust) decreased significantly, but secondary sources increased dramatically. Industry, dust and vehicles contributed 36.6 μg m^{-3}, 13.9 μg m^{-3}, and 9.2 μg m^{-3} to the reduction of $PM_{2.5}$, respectively.

1. Introduction

In recent years there has been growing concern about the impact of air pollution in China, with the emphasis shifting from pollution on a local scale to that on a regional and larger scale. Fine particle is the primary pollutant of concern in the context of regional and complex air pollution in China (Chan and Yao, 2008), although rising NOx and high O_3 are also causes for concern. $PM_{2.5}$ has long been associated with adverse health impacts such as respiratory disease and increased mortality (Gens et al., 2014; Hagerman et al., 2014; Lin et al., 2014). Recent interest has focused on the chemical species, transformation and sources (Cao et al., 2003; Chan and Yao, 2008; He et al., 2001; Tan et al., 2009a, 2009b; Tang et al., 2006; Yang et al., 2011b) of $PM_{2.5}$ since it is both an air pollutant and also has climatic implications; moreover, the mechanisms of formation, evolution, as well as their quantification, are complex (Duan et al., 2007; Liu et al., 2008; Querol et al., 2008; Sun et al., 2006; Wang et al., 2002; Yang et al., 2011a).

In recent years, many studies have investigated the level, temporal and spatial variation, transformation and sources of chemical species of $PM_{2.5}$ (Cao et al., 2003, 2004; Duan and Tan, 2013; Duan et al., 2007; Keene et al., 2014; Querol et al., 2008; Tan et al., 2009a, 2009b). Zhang and Friedlander (2000) compared the inorganic elements databases for $PM_{2.5}$ during 1980–1993 in China. Chan and Yao (2008) reviewed the chemical composition of $PM_{2.5}$ across China with a focus on Metropolitan Regions. Chemical species and the reconstructed speciation of $PM_{2.5}$ were compared in representative Chinese megacities in order to characterize $PM_{2.5}$ (Yang et al., 2011b). However, many of the studies were based on small samples and observations were of short duration, making source apportionment and reliable trend analysis difficult. Systematic and long term observations of fine particle and their composition are critical for understanding their sources, transport and transformation processes, which in turn are essential for the formulation of effective air quality management strategies.

Foshan is located in the hinterland of the Pearl River Delta (PRD) in central Guangdong Province. It is the third largest city in the PRD, a major manufacturing center and one of the most important built-up areas in China. The key industries in Foshan include ceramics, nonmetal ore processing, and manufacturing including household electrical appliances and furniture. Most of these industries are highly polluting; for example, in 2008 the ceramic plants in Foshan account for around 30% of total world ceramics production and they consumed about 40% of the coal used in Foshan (Shen and Wei, 2012). These industries emit large amounts of pollutants into the atmosphere, and they have led to a rapid deterioration in air quality in and around the region. According to the Pearl River Delta Regional Air Quality Monitoring Network, SO_2, NO_x and PM_{10} (particulate matter <10 μm in diameter) in Foshan city are significantly higher than in other areas of the PRD.

In order to improve the air quality and thus public health, since 2007 the government has implemented a wide range of measures to reduce emissions from vehicles, power plants, and industrial and commercial processes. However, the primary emphasis of control measures has varied greatly from year to year. Here we present the results of an investigation of $PM_{2.5}$ in Foshan from 2008 to 2014, which was supported by the Chinese government. The interval of study overlaps with the implementation of major air pollution control measures in Foshan city. $PM_{2.5}$ samples were collected simultaneously at downtown, urban and rural sites with the aims of: 1) investigating their characteristics, trends and sources; 2) evaluating the impact of air quality policy on the chemical composition of $PM_{2.5}$; and 3) producing emission control policy suggestions for $PM_{2.5}$ in Foshan.

2. Environmental materials and methods

2.1. Field observations

Foshan is one of the most populated areas in the Pearl River Delta region in south China and one of the largest manufacturing bases in the world (Tan et al., 2014b). The terrain of Foshan is generally flat, and is surrounded by mountains from three directions; thus stagnant air conditions are frequent over the area and polluted air is not readily dispersed. Six sampling sites were chosen for study (Table 1) and the sampling network descriptions are as follows.

The Downtown site (CC) was situated at about 35 m above ground level in the Foshan Environmental Monitoring Centre, ChanCheng district; it is surrounded by four main roads, residential buildings and business offices. Four urban sites, SS, NH, SD and GM, were located in the Environmental Monitoring Station of SanShui district, NanHai district, ShunDe district and GaoMing district, respectively. These sites are representative of the residential and commercial environment of the urban area in the vicinity of the CC site. A rural site was selected at GengHe town in GaoMing district (GH), about 45 km from the downtown area of Foshan city. PM_{10}, SO_2 and NO_2 were continuously monitored automatically by the Environment Protection Bureau of Foshan. Meteorological data were downloaded from Wunderground (www.wunderground.com).

The 24-h $PM_{2.5}$ samples were collected on quartz filters (PALL 2500QAT-UP) using median-volume samplers (100 L/min). The sampling was performed from November 2008 to December 2014 at the six sites simultaneously (Fig. 1), yielding a total of 2774 $PM_{2.5}$ samples (Table 1). All quartz filters were annealed at 450 °C for 4 h to remove trace organics before use. Before and after collection, the filters were wrapped in baked aluminum foil. The mass of $PM_{2.5}$ was determined by weighing the filters before and after exposure. Prior to weighing, the filters were stored in conditions of constant temperature and humidity (25 °C and 50%) for 24 h. After weighing, the samples were wrapped in aluminum foil and stored at −30 °C until analysis.

Table 1
The number of $PM_{2.5}$ samples collected in Foshan city from 2008 to 2014.

Sampling site	ChanCheng	NanHai	SanShui	ShunDe	GaoMing	GengHe
	CC	NH	SS	SD	GM	GH
2008 winter	29	31	32	28	29	
2009 winter	32	30	31	32	29	28
2010 summer	45	42	39	40	41	46
2010 winter	51	45	46	44	39	45
2011 summer	40	42	38	45	37	44
2011 winter	53	44	43	45	41	43
2012 summer	38	36	42	43	38	36
2012 winter	50	46	45	47	43	45
2013 summer	45	44	43	46	43	46
2013 winter	49	46	44	46	40	46
2014 summer	16	17	16	15	16	15
2014 winter	45	43	45	41	42	47
Sum	493	466	464	472	438	441
Total			2774			