Science of the Total Environment 430 (2012) 217–222

Contents lists available at SciVerse ScienceDirect

Science of the Total Environment

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

Soot particles at an elevated site in eastern China during the passage of a strong cyclone

Hongya Niu [a,b,1], Longyi Shao [a], Daizhou Zhang [b,*]

[a] State Key Laboratory of Coal Resources and Safe Mining, School of Geoscience and Surveying Engineering, China University of Mining and Technology, Beijing 100083, China
[b] Faculty of Environmental and Symbiotic Sciences, Prefectural University of Kumamoto, Kumamoto 862-8502, Japan

ARTICLE INFO

Article history:
Received 13 January 2012
Received in revised form 18 April 2012
Accepted 19 April 2012
Available online 29 May 2012

Keywords:
Inclusion
Coated soot
Individual particle analysis
Morphology
Size
Mountain site

ABSTRACT

Atmospheric particles larger than 0.2 μm were collected at the top of Mt. Tai (36.25°N, 117.10°E, 1534 m a.s.l.) in eastern China in May 2008 during the passage of a strong cyclone. The particles were analyzed with electron microscopes and characterized by morphology, equivalent diameter and elemental composition. Soot particles with coating (coated soot particles) and those without apparent coating (naked soot particles) were predominant in the diameter range smaller than 0.6 μm in all samples. The number–size distribution of the relative abundance of naked soot particles in the prefrontal air was similar to that in the postfrontal air and their size modes were around 0.2–0.3 μm. However, the distribution of inclusions of coated soot particles showed a mode in the range of 0.1–0.3 μm. The coating degree of coated soot particles, which was defined by the ratio of the diameter of inclusion to the diameter of particle body, showed a mode around 0.5 with the range of 0.3–0.6. These results indicate that the status of soot particles in the prefrontal and postfrontal air was similar although air pollution levels were dramatically different. In addition, the relative abundance of accumulation mode particles increased with the decrease of soot particles after the front passage.

1. Introduction

Soot, originating from biomass burning and biofuel and fossil fuel combustion, is one of the most substantial anthropogenic aerosols. After being emitted into the air, soot particles adsorb gaseous species and water vapor (Adachi and Buseck, 2008; Zuberi et al., 2005) and enhance the production of secondary species such as sulfate and nitrate via catalyzing heterogeneous reactions on their surface (Cofer et al., 1984; Wang et al., 2010). These processes modify the size, shape, mixture state and suspension time of the particles and their ability to absorb and scatter radiative energy in the air (Jacobson, 2001; Ramanathan and Carmichael, 2008; Shi et al., 2008; Zhang et al., 2008) and can also indirectly affect hydrological cycles (Jacobson, 2006; Menon et al., 2002). In addition to public concern regarding air pollution and health risk (Wilker et al., 2010), the influence of soot particles on climate change is a still critical challenge for reducing the uncertainties in global climate models (Adachi et al., 2010; IPCC, 2007; Jacobson, 2002).

A recent estimate showed that the emission of black carbon from China in 2006 doubled since 2000 (Ramanathan and Carmichael, 2008). The emission of anthropogenic trace gasses and aerosols was very large in eastern and northeastern parts of China (Gao et al., 2005). Dispersing in the middle-latitude westerly in the northern hemisphere, soot particles can be transported to distant downwind areas, sometimes several hundred and thousand kilometers away from their source areas (Koch and Hansen, 2005).

The temporal and spatial distributions of aerosol particles and their variations in the air are closely related to meteorological conditions. High pressure systems usually favor the accumulation of air pollutants because of the stagnant movement of air and weak wind (Hu and Guo, 2009; van Pinxteren et al., 2009). Passage of cold fronts is usually accompanied with rapid pollution state variation (An et al., 2009), and soot particles aged quickly in the postfrontal air in Beijing (Niu et al., 2011). However, observation of soot particles on the ground is always significantly influenced by local emissions, and it is usually difficult to obtain their characteristics on a regional scale.

Mt. Tai is located at the inter-medium area between eastern and northeastern China and its peak is considered ideal to investigate the physical and chemical properties of particles transported in meso- and large-scale extent (Gao et al., 2005; Yin et al., 2006). A number of observations targeting aerosol particles and gaseous species in eastern China have been conducted from the same location. A comprehensive observation campaign (MTX2006) was held to study the characteristics of ozone and nitrogen dioxide (Irie et al., 2008; Kanaya et al., 2009; Li et al., 2007, 2008), the concentration, source and variation of organic compounds (Inomata et al., 2010; Suthawaree et al., 2010), and the mass concentration of black carbon with optical and thermal

* Corresponding author at: Faculty of Environmental and Symbiotic Sciences, Prefectural University of Kumamoto, Tsukide 3-1-100, Kumamoto 862-8502, Japan. Tel.: +81 96 3216712; fax: +81 96 3846765.
E-mail address: dzzhang@pu-kumamoto.ac.jp (D. Zhang).
[1] Now at: State Key Joint Laboratory of Environmental Simulation and Pollution Control, College of Environmental Sciences and Engineering, Peking University, Beijing 100871, China.

0048-9697/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
doi:10.1016/j.scitotenv.2012.04.050

instruments (Kanaya et al., 2008). Numerical model simulations were conducted to identify the emissions of black carbon aerosols from different sources (Yamaji et al., 2010). These studies provided useful information on the integrated properties of soot particles, such as the origin of soot particles, the variations of concentration and optical depth, and the enhancement of secondary species production. However, knowledge on shape, mixed status and composition of soot particles, critical to the evaluation of the response of climate change and air pollution to the particles (Li et al., 2011a), is very limited. And aging processes of soot particles during their dispersion are not yet well understood.

In the beginning of May 2008, we collected aerosol particles at the top of Mt. Tai, when a strong cyclone passed. The particles were later analyzed with electron microscopes. Morphology, size, and elemental composition of individual particles were statistically analyzed. Soot particles were found to be the majority except accumulation mode particles which were considered to be mainly formed via the coagulation of secondary particles in all samples. Here, we report the observation to show the status of soot particles at different stages of the cyclone passage at the elevated site.

2. Methods

2.1. Sample collection and meteorological record

Particle collection was carried out at the top of Mt. Tai (36.25°N, 117.10°E, 1534 m a.s.l.) in five periods between May 2 and 5, 2008. A two-stage cascade impactor was used to collect particles onto copper meshes which were coated with carbon-sprayed Formvar film. The jet diameters of the first and second stages were 0.7 and 0.4 mm, respectively. The flow rate for particle collection was 1.5 l min^{-1}. Assuming that the particle density is 1.0 g cm^{-3}, the aerodynamic diameter at which particles can be collected at 50% efficiency is 0.6 μm for the first stage and 0.2 μm for the second stage. Therefore, particles in the range of 0.2–0.6 μm would be efficiently collected on meshes at the second stage and particles larger than 0.6 μm on meshes at the first stage. During each sampling period, 3 or 4 sets of first- and second-stage mesh samples were collected within 1 h. After particle collection, each mesh was kept in a plastic capsule. All the capsules were sealed in plastic bags with packaged desiccant and stored in a refrigerator for cooling.

It must be noted that on the top area of Mt. Tai there are several temples, hotels, and restaurants from which soot particles are constantly emitted due to praying activities, cooking and heating. The instruments we used were all portable and this enabled us to collect particles always at an upwind position of those buildings to avoid possible influence from them. This ensured that the collected particles were those transported to the mountain top from distant areas.

Temperature, relative humidity (RH), sea surface pressure, wind speed and wind direction measured by the Taishan meteorological observatory (WMO Station No. 54826) were used in the study. The sampling periods and relevant meteorological records are listed in Table 1. For the convenience of the following description, the samples are termed as MTA, MTB, MTC, MTD, and MTE according to the sampling order.

Table 1
Sampling periods and meteorological conditions.

Sample ID	Sampling period (BST[a])	T (°C)	RH (%)	Wind speed (m s^{-1})
MTA	16:10–16:55 May 2	16.7	80	14.0
MTB	16:20–17:10 May 3	14.0	100	7.5
MTC	06:00–06:50 May 4	4.5	48	20.0
MTD	16:55–17:40 May 4	12.3	24	3.5
MTE	05:30–06:05 May 5	8.3	34	4.5

[a] BST — Beijing standard time, 8 h prior to GMT.

In addition to collecting particle samples, size-segregated number concentrations of particles in the air were measured with an optical particle counter (OPC: Rion KR-12A, Rion Corporation, Japan) during the sampling periods. The available diameter ranges were 0.3–0.5, 0.5–0.7, 0.7–1.0, 1.0–2.0, 2.0–5.0, and >5.0 μm.

2.2. Particle analysis

A transmission electron microscope (Hitachi, H-8100) was used to view and photograph particles on meshes. From each mesh, about 20 pictures, with an effort to document as many particles as possible, were randomly taken. The particles were first investigated by their morphology and characterized as soot particles, accumulation mode particles, and other particles.

An equivalent diameter was used to represent particle size. The equivalent diameter of a particle was defined as a diameter of a circle that had the same projection area as the measured particle on the mesh film, and the equivalent diameters of those in chain or aggregate shapes were measured in the same way. Particles in aqueous phase must have expanded after being captured due to splash. With an assumption that such particles on the mesh after impaction were hemi-spherical with their volume equal to that before being captured, the equivalent diameter of a liquid particle on the film was about 1.3 times of its real geometric diameter in the air. Therefore, the equivalent diameter of a 0.6 μm droplet or coated particle was approximately 0.8 μm.

An environmental scanning electron microscope (ESEM; Philips XL30) equipped with an energy dispersive X-ray (EDX) spectrometer was used to detect the elemental composition of individual particles. The ESEM-EDX can detect elements of atomic numbers larger than 5 except nitrogen in individual particles. Tens of particles in each category were analyzed to confirm whether particles in the same category had similar elemental composition. The operation accelerating voltage was 20 kV, and the integrated time was 50 s for each particle.

3. Results

3.1. Meteorological condition and particle number concentration

The cyclone passed Mt. Tai from May 3 to 4, 2008 (Fig. 1). Its cold front passed the top of the mountain from 21:00 on May 3 to 3:00 on May 4, 2008. The sampling periods in Table 1 indicate that the samples of MTA and MTB were collected in the prefrontal air, MTD and MTE were collected in the postfrontal air, and MTC was collected right after the passage of the front.

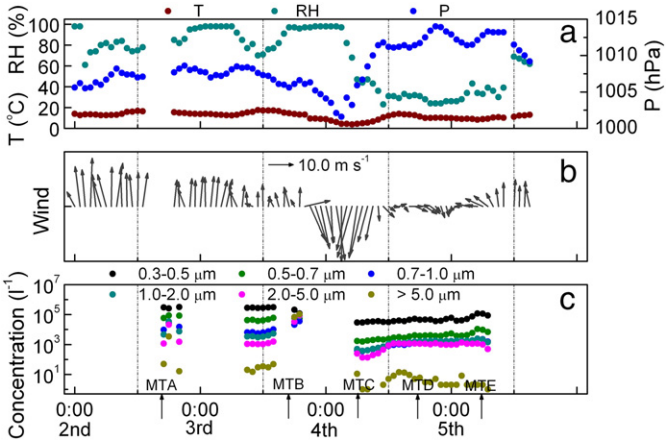

Fig. 1. Time series of surface weather conditions. (a): Temperature (T), relative humidity (RH) and sea surface pressure (P). (b): Wind speed and direction. (c): Particle number concentrations in different size ranges. The start time of MTA, MTB, MTC, MTD and MTE is marked by arrow.