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Carbonaceous aerosols in the air masses transported from Indochina to Taiwan: Long-term observation at Mt. Lulin

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HIGHLIGHTS

- Long-term observation of carbonaceous aerosols in East Asia.
- Transported Indochina biomass burning plume is observed at Mt. Lulin.
- Aerosol carbonaceous content is enhanced in biomass burning plume.
- The impact of Indochina biomass burning is more serious than other places.
- The West Pacific background OC and EC are 1.33 μg m^{-3} and 0.35 μg m^{-3}.

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ABSTRACT

Eight carbonaceous fractions from aerosols were resolved using the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol (Chow et al., 1993). The aerosols were collected at the Mountain Lulin Atmospheric Background Station (Mt. Lulin, 2862 m a.s.l.) in Central Taiwan from April 2003 to April 2012. The monthly and yearly levels of organic carbon (OC) and elemental carbon (EC) varied consistently with $PM_{2.5}$ mass concentrations during biomass burning (BB) period. The highest monthly carbonaceous content was observed in March and the highest yearly carbonaceous concentration was observed in 2007. This finding is consistent with the BB activity in Indochina and indicates that carbonaceous content is a major component of BB aerosols. Lee et al. (2011) classified four trajectory groups from the air masses transported to Mt. Lulin during the aerosol collection period. For the air masses transported from the BB area (the *BB* group) in Indochina, the carbonaceous content was greater than the water-soluble ions in $PM_{2.5}$, and the OC/EC ratio (4.8 ± 1.5) was high. With EC as the indicator of primary emission sources, the air masses of the *BB* group were found to contain more primary than secondary OC. The *Anthropogenic* group (from the local and free troposphere below the 700-hPa pressure level over the Asian continent) probably contained more secondary than primary OC or the sources of OC and EC could be quite diverse. The average char-EC/soot-EC (low-temperature EC/high-temperature EC) ratios were 3.9 ± 3.5, 0.4 ± 0.4, 0.9 ± 0.8, and 0.3 ± 0.4 for the trajectory groups *BB*, *SNBB* (from BB source areas during the non-BB period), *Anthropogenic*, and *FT* (from the oceanic area and the free troposphere above the 700-hPa pressure level over the Asian continent), respectively. The presence of a high char-EC/soot-EC ratio confirmed the correct classification of the *BB* group, whereas the low ratios from the other groups indicated the strong influence of vehicle exhaust. It is noted that higher OC and EC levels were obtained at Mt. Lulin as compared with those obtained at other high-elevation sites. This difference suggested that the Indochina BB plume exhibited a more serious climatic impact on the background air

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in East Asia than in other places in Asia and Europe. On the basis of the carbonaceous levels of the *SNBB* and *FT* groups, the background OC and EC levels of approximately 3000 m in the West Pacific are around 1.33 μg m^{-3} and 0.35 μg m^{-3}, respectively.

1. Introduction

Black carbon (BC) or elemental carbon (EC) can directly absorb solar radiation and alter the climate (Menon et al., 2002). Carbonaceous aerosols can act as cloud condensation nuclei, which can decrease the size of cloud droplets, increase cloudiness, extend their residence time in the atmosphere, and indirectly influence the solar radiation budget of the Earth (Charlson et al., 1992). Thus, the impact of carbonaceous aerosols on regional climatology has drawn significant interest in recent years (Hansen et al., 1997; Jacobson, 2004; Lau and Kim, 2006; Li et al., 2010; Panicker et al., 2010; Ramana et al., 2010; Cappa et al., 2012; Chung et al., 2012). Studies on the effect of carbonaceous aerosols on the radiation budget gradually focused on the Southeast Asian region (Hsu et al., 2003; Wang et al., 2007; Lee and Kim, 2010). Hsu et al. (2003) analyzed satellite data in March 2000 and found that smoke aerosol from biomass burning (BB) emitted from Indochina was present over large areas of cloud in northern Vietnam to southern China, which reduces substantial outgoing shortwave radiation flux. Wang et al. (2007) adopted numerical models and predicted that an anticyclone over the Bay of Bengal would dominate the transport of pollutants in Southeast and South Asia. Results of the numerical simulation by Lee and Kim (2010) suggested that the BC emitted from BB plume could extend from the surface up to the free troposphere, absorb solar radiation, and heat the mid-troposphere, further perturbing the atmospheric circulation and reducing the precipitation over Indochina. Given the close relationship between carbonaceous aerosols and solar radiation, long-term measurement of carbonaceous aerosols can significantly help evaluate the impact of carbonaceous aerosols on atmospheric climate change.

Aerosol contains thousands of organic components, but only a small percentage of organic aerosols can be resolved by analytical methods (Saxena and Hildemann, 1996; Turpin et al., 2000; Maria et al., 2002, 2003). To infer the source contributions of carbonaceous aerosols, tracers are often used to represent specific sources; e.g., levoglucosan can be used to distinctly identify the BB contribution. The ratio of organic carbon (OC) to EC was used to differentiate various emission sources, such as gasoline vehicles, diesel vehicles, coal combustion, and BB (Watson et al., 2001; Chow et al., 2004; Liu et al., 2006). However, with the development of the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal/optical reflectance (TOR) protocol (Chow et al., 1993), more carbon fractions that evolved from different temperatures can be used to enhance the identification of major emission sources (Chow et al., 2004; Kim et al., 2004; Cao et al., 2006). The ratio of char-EC to soot-EC (low-temperature EC to high-temperature EC) has been considered an effective tracer for source identification (Han et al., 2009; Zhu et al., 2010) because emission sources have different combustion temperatures.

In the last decade, carbonaceous aerosols collected at high-elevation sites have mostly been located in Europe (Hitzenberger et al., 1999; Lavanchy et al., 1999; Krivácsy et al., 2001; Gelencsér et al., 2007; Legrand and Puxbaum, 2007) and Asia (Carrico et al., 2003; Cao et al., 2009). The West Pacific lacks observation sites for background carbonaceous aerosols. The effect of West Pacific aerosol is not limited to the regional scale (Wang et al., 2007) but discernible as well in the air quality in North America by long-range transport (Park et al., 2003; Jaffe et al., 2005; Peltier et al., 2008). The Mountain Lulin Atmospheric Background Station (Mt. Lulin, 2862 m a.s.l.) is a unique high-elevation site in the West Pacific, which monitors transported anthropogenic aerosols from East Asia and BB aerosols from Indochina (Sheu et al., 2010; Lee et al., 2011). The present study reports on the properties of long-term carbonaceous aerosols, which can provide valuable information regarding the properties of regional-scale aerosol in East Asia.

2. Methods

2.1. Sampling site and sample collections

The geographic location of the Mt. Lulin site (120° 52′ 25″ E, 23° 28′ 07″ N), which is situated at the summit of Mt. Lulin in Central Taiwan, is indicated in Fig. 1. The Mt. Lulin site was established to investigate the impact of regional and long-range transported air pollutants in the free troposphere in East Asia. Mt. Lulin is located in Yushan National Park, where construction of residential housing has been banned. The observatory provides an unobstructed view of the surrounding area. Each sample was collected for 24 h with filters installed in R&P 3500 ChemComb Speciation Sampling Cartridges from April 2003 to April 2012. Aerosol mass was collected with Teflon® filters (R2PJ047 Teflo™, PALL Life Sciences, Inc., Ann Arbor, MI, USA) at a flow rate of 16.7 L min^{-1} and weighed using a Mettler MX5 Microbalance (Mettler Toledo Co. Inc., Greifensee, Switzerland) with ±1 μg sensitivity in a room controlled in the range of 20–23 °C and 30–40 %RH. Carbonaceous aerosols were collected using quartz fiber filters (TISSUQUARTZ 2500QAT-UP, PALL Life Sciences, Inc., Ann Arbor, MI, USA). The collected filters were refrigerated and transported to the National Central University laboratory for subsequent carbon analysis.

2.2. Aerosol carbonaceous content

Aerosol carbonaceous content was analyzed using a DRI Model 2001A OC/EC Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA) that employs a TOR correction scheme for pyrolized OC (OP), following the IMPROVE protocol to monitor OC and EC fractions (Chow et al., 1993; Watson et al., 2005). The operating principle of this protocol is based on the preferential oxidation of OC and EC at different temperatures. The split of OC and EC is according to the finding that OC can be volatilized from the 0.526 cm^2 sample punch in a pure helium (He) atmosphere at low temperatures, whereas EC is not evolved until the introduction of minor oxygen (O_2) in the He environment at high temperatures. The analytical protocol was thus programmed to gradually increase the oven temperature step by step from room temperature to 840 °C. Depending on the elution time of the corresponding temperature, OC is defined as the carbon evolved from the filter punch in a pure He atmosphere ending at 140 °C (OC1), 280 °C (OC2), 480 °C (OC3), and 580 °C (OC4 plus OP); and EC is the carbon evolved in a 98% He/2% O_2 atmosphere ending at 580 °C subtracted by OP (EC1-OP), 740 °C (EC2), and 840 °C (EC3). OP is defined as the carbon evolved between the moment when 2% O_2 is introduced into the 98% He atmosphere at 580 °C and the moment when the surface reflectance of a laser on the filter punch returns to its