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Characterization of carbonaceous aerosols in urban air

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Abstract

Concentrations of black carbon, [BC], were determined with an Aethalometer every 5 min at Lahore, a city of about 10 million in Pakistan, from 22 November 2005 to 31 January 2006. [BC] were very high, ranging from about 5 to 110 $\mu g\,m^{-3}$, with a mean of 21.7 $\mu g\,m^{-3}$. A distinct diurnal variation was observed: concentrations were lowest from about 10 a.m. to 4 p.m. local time (LT), and highest around 5–9 p.m. No clear relationship was observed between surface wind directions and [BC], although some of the highest concentrations were observed when the airflow was from southwest to northwest. The daily variations in concentrations were strongly affected by the diurnal variations in the mixing height; BC concentrations were low during the day when the mixing heights were high, ~1000 m, and very high at night when the mixing heights were low <250 m. Periods of light to dense fog occurred from 22 December through 4 January. [BC] were generally lower than average when fog occurred during the night and early morning, but they were not necessarily lower during daytime fogs. We also collected aerosols on quartz filters every 3, 6, or 12 h and determined the concentrations of elemental, [EC], and organic carbon, [OC], using the thermal–optical method. The [BC] were highly correlated with EC ($r^2 = 0.71$), but on average 25% higher than [EC]. The [EC] and [OC] concentrations were moderately correlated ($r^2 = 0.65$). The [OC]/[EC] ratios varied from 2.8 to 12, with a mean of 5.6. Although a large component of the carbonaceous aerosols in Lahore originated from fossil fuel combustion, a significant fraction was derived from biomass burning.

Keywords: Black carbon; Elemental carbon; Organic carbon; Carbonaceous aerosols; $PM_{2.5}$ mass; Urban aerosols; Fog; Asia

1. Introduction

Incomplete combustion of fossil fuels or bio-fuels produces soot in the form of elemental or black carbon, and numerous organic carbon (OC) species. Although the terms are often used interchangeably, there are subtle differences between BC and EC. BC is an operational definition for that fraction of the carbonaceous material that absorbs visible light. In principle, all EC is BC, but not necessarily all BC (as measured optically) is EC, since some organic species at very high concentrations can contribute to optical absorption (e.g., Andreae and Gelencsér, 2006). The bulk of BC aerosols (~90%) reside in the $PM_{2.5}$ fraction (Viidanoja et al., 2002). BC constitutes ~5−15% of $PM_{2.5}$ in urban air. Excessive $PM_{2.5}$ concentrations have been shown to negatively

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affect the respiratory and cardiac health of humans (e.g., Dockery et al., 1993; Gwynn et al., 2000; Pope et al., 2002) and also reduce visibility. BC aerosols absorb solar radiation and are the second largest contributor to global warming, after greenhouse gases (Jacobson, 2002). On a global basis BC contributes $\sim 0.5\,W\,m^{-2}$ to radiative forcing (Sato et al., 2003). BC may also have regional climatic impacts. For example, Menon et al. (2002) have suggested that the high concentrations of soot over India and China are responsible for a trend toward increased flooding in the south and drought in the north. It is estimated that the reduced atmospheric transparency caused by high soot concentrations over India and China decreases agricultural productivity by 10–20% (Chameides et al., 1999). Soot deposited on plant leaves also reduces plant productivity (Bergin et al., 2001).

Since the mean residence time of BC aerosols in the atmosphere is about 6 days, they can be transported up to a few thousand km from their sources. So the observed concentrations at a given location are likely the sum of local emissions plus a component transported from distant sources. Hence, for radiative forcing calculations, it is important to determine [BC] that represents specific regions. Thus far, radiative forcing calculations have been based on model estimates of [BC] from energy consumption, and emission factors for various combustion technologies used (Bond et al., 2004). Direct measurements of [BC] from globally diverse regions are needed to validate these models.

Sources of BC are bio-fuel combustion, agricultural residuals burning, forest fires, and fossil fuel combustion including diesel use in automobiles and industry. Atmospheric [BC] vary widely from a background level of $0.0011\,\mu g\,m^{-3}$ at a South Polar site (Hansen et al., 1988) to $\sim 10-20\,\mu g\,m^{-3}$ in Paris, France (Ruellan and Cachier, 2001), to $3-14\,\mu g\,m^{-3}$ in Singapore (Balasubramanian et al., 2003), to as high as $50\,\mu g\,m^{-3}$ in Kanpur, India (Tripathi et al., 2005). The environmental regulations in the countries in the western hemisphere have greatly decreased the concentrations of many environmental pollutants including BC. For example, the concentrations of many trace elements and SO_4 in the Northeastern US have decreased two-folds or more from 1979 to 2002 (Husain et al., 2004). On the other hand owing to rapid industrialization, concentrations of many chemical species in South Asia are rising and are expected to continue to increase rapidly. Therefore the impact of aerosols on global climate and on human health would also increase with time. We have earlier reported exceedingly high concentrations of SO_4 and a few trace elements in Lahore, Pakistan, during the winter fog that can cover a substantial portion of the Indian subcontinent (Hameed et al., 2000; Rattigan et al., 2002). Accordingly, we undertook an extensive investigation to: (1) determine $PM_{2.5}$ mass present in the atmosphere; (2) chemically characterize $PM_{2.5}$ aerosols in Lahore during the winter of 2005–2006; and (3) resolve the sources of various chemical species present. Furthermore, the thermal optical method is increasingly used to determine the atmospheric EC and OC. Since such EC data will provide an important source for estimating radiative forcing, we conducted a comparison of [BC] and [EC] by simultaneously determining [BC] by an Aethalometer and [EC] by the thermal optical method. In this paper we present: (1) continuous measurements of [BC] with 5-min time resolution using an Aethalometer, (2) [EC] and [OC] measurements on quartz filters using the thermal–optical method with a time resolution of 3, 6, or 12 h, (3) measurements of $PM_{2.5}$ mass, (4) a comparison of the results from Aethalometer and the thermal–optical method, and (5) an evaluation of the impact of biomass burning on carbonaceous aerosols in Lahore. The major ion and trace element composition and a resolution of sources of $PM_{2.5}$ and chemical species in Lahore will be published elsewhere.

2. Methods

2.1. Sampling

The sampling was initiated on 22 November 2005 and ended on 31 January 2006. The sampling site was centrally located in the city on the rooftop of a 3-storys building on the campus of Punjab University (31.571 °north, 74.313 °east). The site is south and slightly east of the old city proper and 8.9 km west and slightly south of Lahore International Airport. Walton Airport is 3.2 km east of the site. We measured real-time [BC] with a Magee Scientific Model AE21 Aethalometer. The Aethalometer was in a room on the second floor, with the inlets suspended outside a window. The inlet consisted of a BGI sharp-cut cyclone and the 6 mm ID tubing supplied by Magee Scientific. The flow was maintained at 4 liter per minute (lpm) corresponding to a 3.2 μm cut-point through the