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Evaluation of the thermal/optical reflectance method for discrimination between char- and soot-EC

Yongming Hana, Junji Caoa, Judith C. Chowa, b, John G. Watsona, b, Zhisheng Ana, hangdong Jina, c, Kochy Fungd, Suixin Liua

Abstract

Many optical, thermal and chemical methods exist for the measurement of elemental carbon (EC) but are unable or neglect to differentiate between the different forms of EC such as char- or soot-EC. The thermal/optical reflectance (TOR) method applies different temperatures for measuring EC and organic carbon (OC) contents through programmed, progressive heating in a controlled atmosphere, making available eight separate carbon fractions – four OC, one pyrolyzed organic carbon, and three EC. These fractions were defined by temperature protocol, oxidation atmosphere, and laser-light reflectance/transmittance. Stepwise thermal evolutional oxidation of the TOR method makes it possible to distinguish char- from soot-EC. In this study, different EC reference materials, including char and soot, were used for testing it. The thermograms of EC reference materials showed that activation energy is lower for char- than soot-EC. Low-temperature EC1 (550 °C in a 98% He/2% O2 atmosphere) is more abundant for char samples. Diesel and n-hexane soot samples exhibit similar EC2 (700 °C in a 98% He/2% O2 atmosphere) peaks, while carbon black samples peaks at both EC2 and EC3 (800 °C in a 98% He/2% O2 atmosphere). These results supported the use of the TOR method to discriminate between char- and soot-EC.

Keywords: Thermal/optical reflectance method; Elemental carbon; Char-EC; Soot-EC

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Different characteristics of char and soot in the atmosphere and their ratio as an indicator for source identification in Xi'an, China

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Abstract. Numerous definitions and analytical techniques for elemental (or black) carbon (EC) have been published in the scientific literature, but still no generally accepted interdisciplinary definition exists. EC is not a single chemical compound, but is mainly composed of two parts of carbon contents: combustion residues from pyrolysis and combustion emissions formed via gas-to-particle conversion. Accordingly EC is subdivided into two classes: char and soot. Char is defined as carbonaceous materials obtained by heating organic substances and formed directly from pyrolysis, or as an impure form of graphitic carbon obtained as a residue when carbonaceous material is partially burned or heated with limited access of air. Soot is defined as only those carbon particles that form at high temperature via gas-phase processes. Since the different classes of EC have different chemical and physical properties, their optical light-absorbing properties differ, so that it is essential to differentiate them in the environment. The thermal optical reflectance (TOR) method was used to differentiate between char-EC and soot-EC according to its stepwise thermal evolutional oxidation of different carbon fractions under different temperatures and atmosphere. Char-EC and soot-EC are operationally defined as EC1-OP and EC2+EC3 (EC1, EC2 and EC3 corresponding to carbon fractions evolved at 550, 700 and 800 °C in a 98% He/2% O2 atmosphere, respectively), respectively. One year of observations of the daily and seasonal variations of carbonaceous particles were conducted in Xi'an, China in 2004 to demonstrate the different characteristics of char and soot in the atmosphere. Total carbon (TC), organic carbon (OC), EC and char-EC showed similar seasonal trends, with high concentrations in winter and low concentrations in summer, while soot-EC revealed relatively small seasonal variations, with maximum concentration (1.85±0.72 μg m−3) in spring and minimum concentration (1.15±0.47 μg m−3) in summer. The strong correlation between EC and char-EC (R2 = 0.99) and poor correlation between EC and soot-EC (R2 = 0.31) indicate that previously reported total EC in the literature reflected the distribution characteristics of char only, while overlooking that of soot. However, soot exhibits stronger light-absorbing characteristics than char, and merits greater focus in climate research. The small seasonal variation of soot-EC indicates that soot may be the background fraction in total EC, and is likely to have an even longer lifetime in the atmosphere than previously estimated for total EC, which suggests that soot may has a greater contribution to global warming. While both char-EC/soot-

EC and primary OC/EC ratios vary with emission sources, only OC/EC ratio is affected by SOA. Thus char-EC/soot-EC may be a more effective indicator than OC/EC in source identification of carbonaceous aerosol. Comparison of seasonal variations of OC/EC and char-EC/soot-EC ratios in Xi'an confirms this point. However, wet scavenging by snow and rain was more effective for char than for soot and influenced the char-EC/soot-EC ratio, and this factor should be considered in source identification as well.

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Comparison and implications of PM2.5 carbon fractions in different environments

Chong-Shu Zhua, , Jun-Ji Caoa, d, Chuen-Jinn Tsaib, Zhen-Xing Shenc, Yong-Ming Hana, Sui-Xin Liua, Zhu-Zi Zhaoa

Highlights

The eight carbon fractions, char and soot at rural, urban, tunnel and remote sites were compared.

OC/EC and char/soot among four sites were elucidated as effective source indicator.

The results might give implications for models in estimating their climate effects.

Abstract

The concentrations of PM2.5 carbon fractions in rural, urban, tunnel and remote environments were measured using the IMPROVE thermal optical reflectance (TOR) method. The highest OC1 and EC1 concentrations were found for tunnel samples, while the highest OC2, OC3, and OC4 concentrations were observed for urban winter samples, respectively. The lowest levels of most carbon fractions were found for remote samples. The percentage contributions of carbon fractions to total carbon (TC) were characterized by one peak (at rural and remote sites) and two peaks (at urban and tunnel sites) with different carbon fractions, respectively. The abundance of char in tunnel and urban environments was observed, which might partly be due to traffic-related tire-wear. Various percentages of optically scattering OC and absorbing EC fractions to TC were found in the four different environments. In addition, the contribution of heating carbon fractions (char and soot) indicated various warming effects per unit mass of TC. The ratios of OC/EC and char/soot at the sites were shown to be source indicators. The investigation of carbon fractions at different sites may provide some information for improving model parameters in estimating their radiative effects.

Keywords: PM2.5; Elemental carbon; Organic carbon; OC/EC; Char/soot

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Spatio-temporal variability in atmospheric abundances of EC, OC and WSOC over Northern India

Kirpa Ram, M.M. Sarin,

Abstract

The atmospheric abundances of elemental carbon (EC), organic carbon (OC) and water-soluble organic carbon (WSOC) have been measured in aerosol samples collected during wintertime (December–March) from selected sites (urban, rural and high-altitude) in northern India. A characteristic feature of their abundance pattern, at urban sites, is reflected in the OC/EC ratios (range: 2.4–14.5, Av=7.8±2.4, n=77) indicating dominant contribution from biomass burning sources (wood-fuel and agriculture waste). This is in sharp contrast to the OC/EC ratios at a rural site (range: 2.1–4.0, Av=3.1±0.6, n=7) influenced by emissions from coal-fired industries. The long-term measurements made from a high-altitude site (∼2000 m amsl) reveal significantly lower abundances of EC and OC; suggesting that boundary layer dynamics (during wintertime) play an important role in efficient trapping of pollutants within the Indo-Gangetic Plain (northern India). The WSOC/OC ratios are fairly uniform (∼0.35) in aerosols over urban sites but relatively enhanced contribution of WSOC and higher ratios (∼0.5) at a high-altitude site emphasizes the significance of secondary organic aerosols. The comprehensive data set on EC, OC and WSOC/OC ratios from northern India is crucial to improve model parameterization of carbonaceous aerosols for atmospheric scattering and absorption of solar radiation on a regional scale.

Keywords: Aerosol; Organic carbon; Elemental carbon; Water-soluble organic carbon

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OC/EC ratio observations in Europe: Re-thinking the approach for apportionment between primary and secondary organic carbon

Casimiro Pioa, , Mário Cerqueiraa, Roy M. Harrisonb, Teresa Nunesa, Fátima Mirantea, Célia Alvesa, César Oliveiraa, Ana Sanchez de la Campaa, c, Begoña Artíñanod, Manuel Matose

Abstract

This study explores a large set of OC and EC measurements in PM10 and PM2.5 aerosol samples, undertaken with a long term constant analytical methodology, to evaluate the capability of the OC/EC minimum ratio to represent the ratio between the OC and EC aerosol components resulting from fossil fuel combustion (OCff/ECff). The data set covers a wide geographical area in Europe, but with a particular focus upon Portugal, Spain and the United Kingdom, and

includes a great variety of sites: urban (background, kerbside and tunnel), industrial, rural and remote. The highest minimum ratios were found in samples from remote and rural sites. Urban background sites have shown spatially and temporally consistent minimum ratios, of around 1.0 for PM10 and 0.7 for PM2.5.The consistency of results has suggested that the method could be used as a tool to derive the ratio between OC and EC from fossil fuel combustion and consequently to differentiate OC from primary and secondary sources. To explore this capability, OC and EC measurements were performed in a busy roadway tunnel in central Lisbon. The OC/EC ratio, which reflected the composition of vehicle combustion emissions, was in the range of 0.3–0.4. Ratios of OC/EC in roadside increment air (roadside minus urban background) in Birmingham, UK also lie within the range 0.3–0.4. Additional measurements were performed under heavy traffic conditions at two double curbside sites located in the centre of Lisbon and Madrid. The OC/EC minimum ratios observed at both sites were found to be between those of the tunnel and those of urban background air, suggesting that minimum values commonly obtained for this parameter in open urban atmospheres over-predict the direct emissions of OCff from road transport. Possible reasons for this discrepancy are explored.

Highlights

► OC/EC ratio is commonly used in the source apportionment of carbonaceous aerosol.
► European urban aerosol has a similar minimum OC/EC ratio.
► Ambient aerosol seems to be always contaminated with secondary OC.
► Tunnel OC/EC ratios are probably a better indicator of primary fossil fuel OC.

Keywords: Organic carbon; Elemental carbon; OC/EC ratio; Secondary organic carbon; Size distribution