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Exploring the Variation between EC and BC in a Variety of Locations

Gbenga Oladoyin Salako[1], Philip K. Hopke[1*], David D. Cohen[2], Bilkis A. Begum[3], Swapan K. Biswas[3], Gauri Girish Pandit[4], Yong-Sam Chung[5], Shamsiah Abd Rahman[6], Mohd Suhaimi Hamzah[6], Perry Davy[7], Andreas Markwitz[7], Dagva Shagjjamba[8], Sereeter Lodoysamba[8], Wanna Wimolwattanapun[9], Supamatthree Bunprapob[9]

[1] *Center for Air Resources Engineering and Science, Department of Chemical and Biomolecular Engineering, Clarkson University, Potsdam, NY 13699-5708, USA*
[2] *Australian Nuclear Science and Technology Organisation (ANSTO), Physics Division, Private Mail Bag 1, Menai 2234, NSW, Australia*
[3] *Bangladesh Atomic Energy Commission (BAEC), Atomic Energy Centre, Dhaka (AECD), P.O. Box 164, Dhaka, Bangladesh*
[4] *Bhabha Atomic Research Centre, Trombay, Mumbai 400085, India*
[5] *Hanaro Center, Korea Atomic Energy Research Institute (KAERI), 150 Dukjin-dong, Yusung-ku, P.O. Box 105, Daejon 305-600, Korea*
[6] *Waste and Environmental Technology Division, Malaysian Nuclear Agency, Bangi 43000 Kajang, Selangor, Malaysia*
[7] *Institute of Geological and Nuclear Sciences (GNS), 30 Gracefield Road, P.O. Box 31-312, Lower Hutt, New Zealand*
[8] *National University of Mongolia, Ikh Surguuliin, P.O. Box 46/789, Ulaanbaatar 210646, Mongolia*
[9] *Thailand Institute of Nuclear Technology (TINT), 16 Vibhavadi Rangsit Road, Bangkok 10900, Thailand*

ABSTRACT

Despite intensive research over the past three decades, a generally accepted standard method to measure black carbon (BC) or elemental carbon (EC) still does not exist. Data on BC and EC concentrations are method specific and can differ widely. This work was motivated by the lack of any prior study that established the variability between these two measures of carbonaceous particulate matter. Measurements of BC and EC were performed at different locations across Asia and the South Pacific in both urban and suburban locations. Filter samples were collected during the winter of 2007 to the winter of 2010 and analyzed for both BC and EC. EC was measured using the Interagency Monitoring of Protected Visual Environments (IMPROVE_A) protocol. Black carbon was measured by the EELS reflectometer (Diffusion Systems, Ltd). Bangladesh had the highest correlation coefficient of 0.93. Bangkok, Thailand on the other hand had the lowest correlation coefficient of 0.34. A review of previously reported source apportionment of BC concentrations in these locations showed that New Zealand had the highest percentage (82%) of BC from biomass while Mongolia had the lowest percentage of 3.1%. The fraction of BC emissions from diesel vehicles was found predominant in Mumbai, India with values as high as 80%. Mongolia had the lowest emission of BC from diesel vehicle (5.4%) with coal- and biomass-combustion being the dominant sources.

Keywords: Elemental carbon; Black carbon; Light absorbing aerosols; Particulate matter.

INTRODUCTION

Black carbon (BC) and elemental carbon (EC) in airborne particulate matter originate from the incomplete combustion of carbonaceous fuel. They are used to describe aspects of ambient particulate matter. However, they are not interchangeable because they are each defined by the way they are measured. These two particulate matter constituents are important components of the atmospheric aerosol because of their light absorbing characteristics and their possible health effects. Light absorption results in a positive (warming) forcing that exceeds that of methane (the third most important greenhouse gas) on a global scale (Jacobson, 2001; Ramanathan and Carmichael, 2008). These light absorbing particles can produce a large indirect positive forcing by enhancing cloud evaporation in the tropics (Ackerman *et al*., 2000).

Characterizing light absorbing aerosols is conceptually

* Corresponding author. Tel.: +1315-268 3861;
Fax: +1-315-268-4410
E-mail address: hopkepk@clarkson.edu

ambiguous (Watson *et al*., 2004, 2005). However, black carbon (BC) and elemental carbon (EC) are not measures of the same properties of particulate matter, even though, these two carbonaceous species are often well correlated (Jeong *et al.*, 2004). Thus, careful definition of these quantities is required to allow proper discussion of their sources and how they behave in the atmosphere.

Black carbon (BC) is an optical measurement that is commonly used to denote the extent of light-absorption by the sample. It is not a direct measure of the carbon concentration. In most environments, BC is produced by the incomplete combustion of fossil and bio-fuels that dominates particulate light absorption (Horvath, 1993). Elemental carbon (EC) represents thermally refractory carbon with a graphitic structure measured by oxidation and detection of the evolved gas. Emissions from diesel heavy trucks are the major source of EC in urban areas of the United States (Gray and Cass, 1998).

These two carbon fractions are typically defined operationally by their measurement protocols. EC is determined by thermal optical protocols such as National Institute of Occupational Safety and Health (NIOSH) Method 5040 with correction by the thermal–optical transmission (TOT) (Birch and Carey, 1996; Babich *et al*., 2000; Chow *et al*., 2001; Watson and Chow, 2002; Lim *et al*., 2003; Baxla *et al*., 2009; Behera *et al*., 2010; Stone *et al*., 2010; Gangwar *et al*., 2011; Kaul *et al*., 2011) and the Interagency Monitoring of Protected Visual Environments/Thermal Optical Reflectance (IMPROVE/TOR) (Chow *et al*., 1993, 2001; Schauer, 2003; Schauer *et al*., 2003; Chow *et al*., 2004, 2007; Pavuluri *et al*., 2011).

Optical attenuation methods such as the aethalometer (Hansen *et al*., 1984; Lou *et al*., 2005; Park *et al*., 2010; Lili *et al*., 2011) or the EELS Reflectometer (Diffusion Systems, Ltd) (Edwards *et al*., 1983; Biswas *et al*., 2003; Begum *et al.*, 2007, 2011; Cohen *et al.*, 2011; Wimolwattanapun *et al.*, 2011; Begum *et al.*, 2012) have also been commonly used to determine BC. The EELS Reflectometer provides a measurement of "British Smoke" that dates back to the famous 1952 London fog episode (Edwards *et al*., 1983). Optical methods are easy to operate and inexpensive although they only measure light absorption and estimate BC using an assumed mass absoption coefficient.

There have been a limited number of comparisons of BC/EC relationships particularly for specific size fractions of the ambient aerosol. Early comparisons like that provided by Edwards *et al.* (1983) examined total suspended particulate (TSP) samples where the wide range of particle size and composition made direct mass comparisons difficult. Cyrys *et al*. (2003) showed good site specific correlations between BC and EC using data from Germany, the Netherlands and Sweden. Jeong *et al*. (2004) also found strong correlations between EC and BC in Rochester, NY and Philadelphia, PA, but again with different slopes in the two locations. The slope in Philadelphia changed during the intrusion of a large wildfire aerosol into the area. Nordmann *et al*. (2009) compare black and elemental carbon measurements in Europe again supporting the variability of the EC-BC relationships.

The EELS reflectometer has been widely used to provide a measure of BC typically using a single fixed mass absorption coefficient. The purpose of this work is to explore the variability in the relationship between black carbon and elemental carbon in a variety of location across Asia and the South Pacific and to provide information regarding the potential errors in the estimation of BC mass with a fixed mass absorption coefficient.

METHODS

Samples were collected at various times in different locations as documented in Table 1. The filters were sampled over the averaging times given in the table. Parallel sampling with identical Gent samplers (Hopke *et al.*, 1997) was performed, except in Australia and Bangladesh. In Australia and Bangladesh, ASP samplers and $PM_{2.5}$ MiniVol (AirMetrics) samplers, respectively, were used. The fine filter in one sampler was a standard 0.45 μm nuclepore. A prebaked quartz fiber filter was used as the fine filter in the second sampler. The specific analytical techniques employed in this study are described below.

Thermal Carbon Analysis

Elemental carbon on the quartz filter was measured by the Interagency Monitoring of Protected Visual Environments (IMPROVE_A) protocol with a DRI 2100A system. Details of this method have been presented by Chow *et al*. (2007).

Black Carbon Analysis

Black carbon was measured using EELS reflectometers (Diffusion Systems, Ltd) in each country. Edwards *et al*. (1983) describes the basis for this measurement and Biswas *et al*. (2003) described its application to nuclepore filter

Table 1. Logistics of the field operations for samples collection at multiple sites.

Location	Latitude	Longitude	Start date	End date	Sample Collection Time
Australia-Lucas Heights	34.05 S	150.98 E	10/31/07	01/30/08	72 hrs & 96 hrs alternately
Bangladesh-Dhaka	23.73 N	90.40 E	01/04/10	01/13/10	24 hrs
India-Mumbai	19.04 N	72.92 E	02/18/08	04/03/08	24 hrs
Korea-Daejon	36.35 N	127.40 E	12/13/07	01/15/08	24 hrs
Malaysia-Kuala Lumpur	N/A	N/A	02/04/08	05/27/08	24 hrs
Mongolia-Ulanbaatar	N/A	N/A	11/06/07	11/22/07	24 hrs
New Zealand-Wainuiomata	40.95 N	175.65 E	07/07/07	09/30/07	24 hrs
Thailand-Bangkok	13.75 E	100.49 E	12/23/07	02/04/08	18 hrs
Thailand-Pathumthani	14.02 E	100.52 E	02/27/08	04/17/08	18 hrs

samples. BC on the filters was estimated using the following equations.

$$BC\,(\mu g/m^3) = \frac{BC\,(\mu g/cm^2) \times \text{Filter Area } (12.56\ cm^2)}{\text{Volume of the sample } (m^3)} \quad (1)$$

$$BC\,(\mu g/cm^2) = \left[1000 \cdot \log\frac{I_0}{I} + 2.39\right] / 45.8 \quad (2)$$

where

I_0 - Unloaded filter reflectance

I - Loaded filter reflectance

To obtain the numerical values in the above equation, a mass absorption coefficient, ε value of 9.7 m^2/g was used. This value was established by collecting samples of candle soot that were weighed as well as their optical density measured for white light reflectance with the EELS reflectometer (Begum *et al*., 2004).

The BC and EC detection limits were 0.02 $\mu g/m^3$ and 0.2 $\mu g/m^3$, respectively, based on the approach described by the U.S. Environmental Protection Agency (40CFR136, Appendix B). Results of our replicate sample analyses showed a typical relative percentage difference of 5%. Additionally, all of the measured concentrations were large compared to the MDL values across all locations with exception of Australia and New Zealand.

Source Apportionments

To better understand the differences in mass absorption coefficients across the region, previously performed and reported source identification and apportionments of BC across all of the locations have been reviewed. Each participating group has independently analyzed $PM_{2.5}$ compositional data from samples collected at their monitoring sites and presented their results. Table 3 summarizes the source apportionments for BC and provides references to the literature in which the results have been reported.

RESULTS AND DISCUSSION

The results from the analyses of both EC and BC samples from a variety of locations are shown in Fig. 1. In most locations, the sample collection time was 24 hours with the exception of Thailand (18 hours) and Australia (72 hours, alternating with 96 hours). The total number of samples collected varied from site to site. The lowest number of samples was 10 from Dhaka, Bangladesh and the suburban site in Thailand while the highest number of sample collection was 34 from New Zealand. The lowest value for both elemental and black carbon concentration was 0.05 $\mu g/m^3$ and 0.01 $\mu g/m^3$ respectively from Australia. The highest values for EC and BC concentrations were 62.23 $\mu g/m^3$ and 53.22 $\mu g/m^3$. The reason for this significant difference could be attributed to differences in the population densities effectiveness of air quality management systems. The sites in Australia and New Zealand are rural with low population densities whereas Dhaka is a megacity in a developing country with many substantial sources of carbonaceous materials (Begum *et al*., 2012).

The overall average concentrations of EC and BC were calculated as 6.21 $\mu g/m^3$ and 7.29 $\mu g/m^3$, respectively, while their standard deviation concentrations were calculated as 2.01 $\mu g/m^3$ and 1.95 $\mu g/m^3$. The results of the average concentrations show that BC was estimated to exceed EC by 17.39%. On the other hand, the variability of the EC results as indicated by the standard deviation of the concentrations exceeds BC by 3%. The variation in EC and BC concentration indicates the possibility of an error in the assumed mass absorption coefficient in specific locations. Minimum, maximum and average concentrations of elemental and black carbon as well as their ratios are given in the Table 2. The average BC/EC ratios were found to range from 0.47 to 3.56. Overall, BC was found to be more abundant at six sites in five different countries while EC is higher at the remaining three sites in three different countries.

To explore the variation in black and elemental carbon concentrations, regression analysis was performed for these methods. It can be seen from Fig. 1 that two of the regression coefficients were low with considerable scatter in the data. The lowest correlations were found for the two Bangkok, Thailand sites. The highest correlations were found for the samples collected in Bangladesh and in New Zealand.

Most of the slopes of the lines in Fig. 1 are reasonably close to 1 supporting the choice of the specific mass absorption coefficient as being a reasonable estimate. However, the two sites in Thailand have much lower slopes suggesting the need to reassess the absorption coefficient value. However, given the low correlation, it may mean that there is a sufficient variation in the nature of the source emissions that

Table2. Minimum, maximum, average concentrations of IMPROVE EC and BC and their ratios.

Location	EC ($\mu g/m^3$)				BC ($\mu g/m^3$)				BC/EC ($\mu g/m^3$)			
	Min.	Max.	Mean	Std. dev.	Min.	Max.	Mean.	Std. dev.	Min.	Max.	Mean	Std. dev.
Australia	0.05	0.85	0.33	0.206	0.01	0.63	0.28	0.18	0.2	0.74	0.47	0.38
Bangladesh	25.45	62.23	39.9	11.45	28	53.22	36.57	8.58	0.86	1.1	0.98	0.17
India	0.44	2.01	1.29	0.441	1.22	5.12	3.58	1.28	2.55	2.77	2.66	0.16
Korea	0.64	2.36	1.18	0.508	0.57	2.94	1.98	0.82	0.89	1.25	1.07	0.25
Malaysia	0.52	3.24	1.78	0.994	1.08	5.5	3.85	1.22	1.7	2.08	1.89	0.27
Mongolia	1.64	4.83	3.47	0.943	1.89	4.15	3.29	0.56	0.86	1.15	1.01	0.21
New Zealand	0.07	5.89	1.62	1.501	0.29	5.41	2.05	1.29	0.92	4.14	2.53	2.28
Thailand (Bangkok)	1.4	6.42	4	1.457	7.02	13.58	9.4	2.38	2.12	5.01	3.56	2.04
Thailand (Pathumthani)	1.6	3.35	2.35	0.562	2.43	6.88	4.65	1.22	1.52	2.06	1.79	0.38

Table 3. Source contributions to mass concentrations of black carbon.

Location	Diesel Vehicles (%)	Biomass Burning (%)	Other (%)
Australia-Lucas Heights[a]	52	24	24
Bangladesh-Dhaka[b]	54	19	27
India-Mumbai[c]	80	20	-
Korea-Daejon[d]	28	48	24
Malaysia-Kuala Lumpur[e]	60	35	5
Mongolia-Ulanbaatar[f]	5.4	3.1	91.5
New Zealand-Wainuiomata[g]	17	82	1
Thailand-Bangkok[h]	33	20	47
Thailand-Pathumthani[h]	43	32	25

a. Cohen *et al*., 2011; b. Begum *et al*., 2011; c. Kothai *et al*., 2011; d. Chung *et al*., 2006; e. Davy *et al*., 2011; f. Rahman *et al*., 2011; g. Davy, private communication, 2011; h. Wimolwattanapun *et al*., 2011.

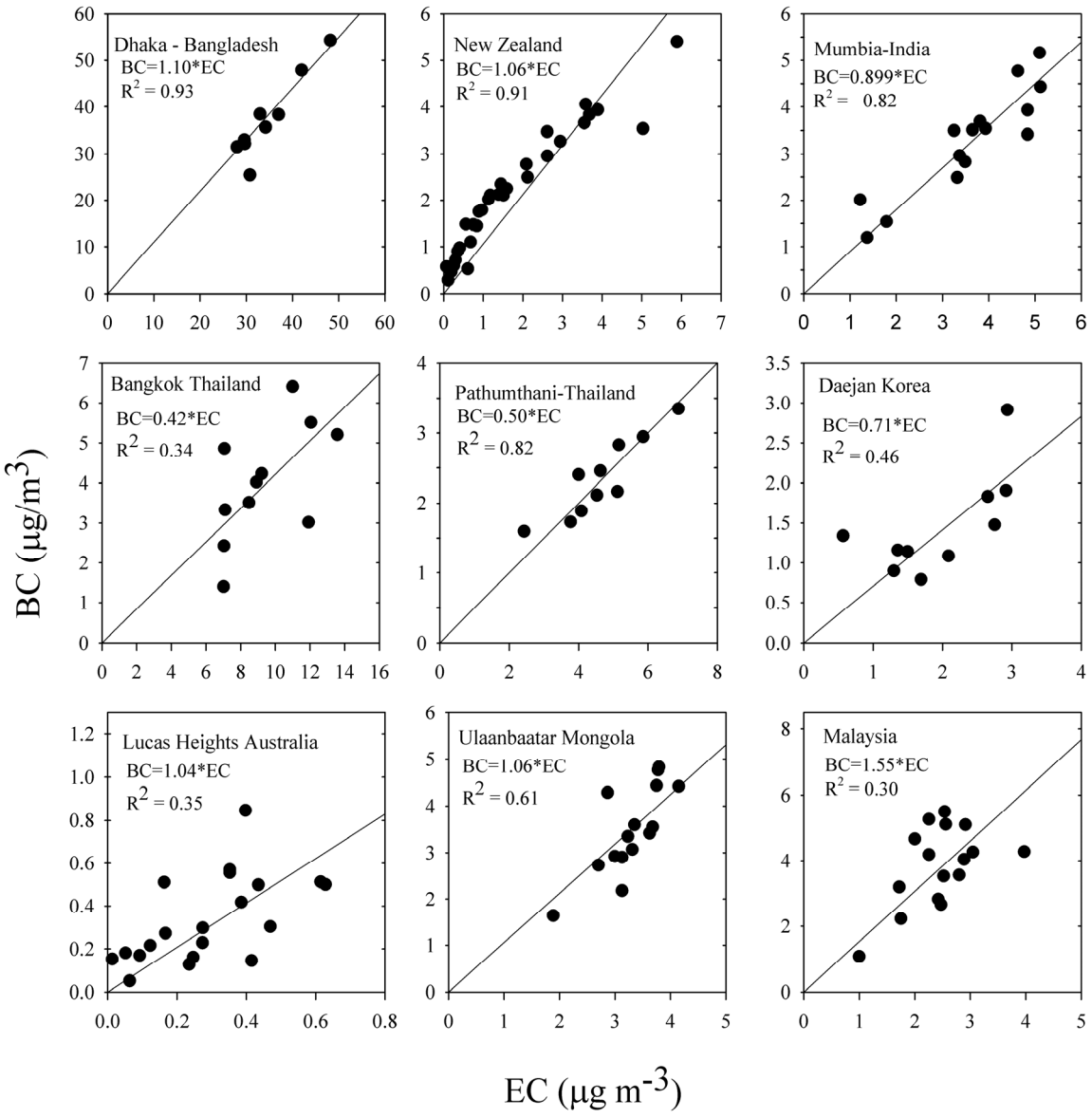

Fig. 1. Relationships between BC and EC concentrations across Asia and Pacific regions.

there are at least two elemental carbon sources with varying light absorptivity as suggested by Jeong *et al*. (2004). In Malaysia, the slope is much greater than 1 but again with a very low correlation coefficient suggesting the possibility of multiple sources contributing to the BC concentrations.

Particulate matter concentrations and compositions differ

from location to location (Hopke *et al*., 2008). The BC source apportionments at all of the locations revealed that New Zealand had highest percentage of black carbon from biomass with values as high as 82% while Mongolia had the lowest percentage of 3.1%. Emissions from diesel vehicles to black carbon concentration were found to be dominant in Mumbai, India with values up to 80% (Kothai *et al*., 2011). Alternatively, Mongolia has the lowest emission of BC from diesel vehicle as 5.4%. The majority of their BC emissions are from home heating and cooking through the combustion of coal and biomass (Davy *et al*., 2011). In general, there is a mixture of biomass burning and traffic as the primary sources of the soot.

The variations in correlation of BC and EC concentrations can be attributed to a number of factors such as differences in particle size distribution, mixing state, and chemical composition leading to variations in light absorption efficiency. This variability results from difference in source contributions such biomass combustion, vehicular emissions, etc. Jeong *et al*. (2004) showed that wood smoke particles had lower light absorptivity than traffic generated particles. They also found that the degree of atmospheric processing changed the absorptivity where EC collected within 2 m of a roadway was twice as light absorbing as that collected a 1 km from a large interstate highway. Thus, although the actual light absorption can be measured with very good precision and accuracy, the variability in the absorptivity per unit mass results in the variability in the EC/BC relationships shown in Fig. 1.

SUMMARY

Collection and analysis of airborne $PM_{2.5}$ samples made in this study indicate that EC and BC concentrations can vary substantially due to wide mixture of sources and degree of atmospheric processing from one location to another location across different countries .This study was performed during the winter of 2007 to the winter of 2010. Bangladesh had the highest correlation coefficient of 0.93, followed closely by New Zealand with 0.91. The two sites in Thailand and the site in Malaysia had low correlation coefficients suggesting a broader range of possible BC sources. Most sites appeared to obtain good results using a fixed mass absorption coefficient of 9.7 m^2/g. Results obtained from the sites with the low correlation coefficients between EC and BC showed a substantial variability in the mass absorption coefficient, likely the result of multiple sources of black carbon with quite different absorptivities.

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