



Aerosol size distributions of elemental and organic carbon in urban and over-water atmospheres

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Abstract

Aerosol mass size distributions of elemental carbon (EC) and organic carbon (OC) were measured to determine the extent to which carbon speciations within ambient aerosols differ between urban and over-water atmospheres. Samples were collected during twenty-one 12-h periods in urban Chicago and over Lake Michigan during the July 1994 and January 1995 sampling intensives. Total particle bound concentrations (sum of all size fractions) of elemental and organic carbon ranged from 0.15 to 0.96 and from 0.94 to 3.04 g m⁻³, respectively. On average, organic material (OM = 1.4 × OC) accounts for 18% of the total aerosol mass collected, but ranges from 10% (over-water, winter) to 39% (urban, summer). With regard to individual size fractions, organic matter ranges from 3% (> 12 μm over-water, winter) to 49% (0.15–0.45 μm, urban, summer) of the particulate matter mass in the size class. Geometric mean aerodynamic equivalent diameters (GMDs) range from 0.72 to 2.4 μm for suspended particulate matter (Σ-PM), from 0.52 to 1.4 μm for EC, and from 0.60 to 1.9 μm for OC. Elemental and organic carbon GMDs are larger in the urban atmosphere than over the water during winter, while this trend is not observed in Σ-PM. Furthermore, geometric standard deviations are larger at the urban location in January for both EC and OC, indicating broader size distributions of both species under winter conditions due to greater quantities of EC and OC in large particles. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Particulate matter; EC; OC; Chicago; Lake Michigan

1. Introduction

Aerosol size is the major determining factor in atmospheric behavior of aerosol particles and thereby controls the residence time and removal mechanisms of aerosol bound contaminants from the troposphere (Junge, 1977; Wesely et al., 1977; Slinn et al., 1978; McMahan and Denison, 1979; Sehmel, 1980; Scott, 1981; Williams, 1982; Ligocki et al., 1985a,b; Bidleman, 1988; Poster et al., 1996a,b; Zufall and Davidson, 1997; Kaupp and McLachlan, 1999). The carbon content of aerosol particles

strongly influences the distribution of semi-volatile organic compounds between the gas and particle bound phases in the natural atmosphere (Finizio et al., 1997; Pankow, 1998). Many of the compound classes which are influenced by aerosol carbon content, including PAHs, PCBs, PCDDs and PCDFs, are potential carcinogens or mutagens and pose significant human health risks (e.g. Pitts, 1983; Schuetzle, 1983; IARC, 1980; LaVoie, 1979). Furthermore, particle size greatly influences inhalation exposure and human health effects of the aerosol bound contaminants (Brown et al., 1950; Lippman, 1970; Kerzetz-Saringer et al., 1971; Nautch and Wallace, 1974; Fairley, 1990; Schwartz and Marcus, 1990; Lane, 1991; Schwartz, 1990; Pope et al., 1992; Schwartz and Dockery, 1992a,b; Dockery et al., 1992,1993). However, few measurements have been made of elemental carbon (EC) and organic carbon (OC) size distributions in urban and over-water atmospheres. Therefore, quantification of EC

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and OC in various atmospheres is a significant step toward understanding the processes which control the atmospheric behavior of these and other trace organic pollutants.

In order to successfully collect size-segregated airborne particulate matter and to conduct chemical analysis of carbon species, the method utilized must minimize sampling artifacts that may alter contaminant concentrations and size distributions (Turpin et al., 1997). These artifacts can be either positive, such as sorption to collection medium, to the apparatus or to particles loaded on the collection medium, or they can be negative, such as, “blow-off” of volatile and semi-volatile compounds sorbed to aerosols. The extent of such artifacts can distort the size distributions measured for organic carbon, but are less likely to influence measurements of elemental carbon. The use of an impactor with ungreased impaction substrates minimizes such artifacts (Zhang and McMurry, 1987,1991; Poster et al., 1995; Turpin et al., 1997; Allen et al., 1999). This apparatus can collect sufficiently large samples during 12 h periods for analysis of EC and OC by the thermal-optical method (Birch and Cary, 1996).

In this paper we present aerosol mass size distributions of elemental and organic carbon in the urban and over-water atmospheres. Size-segregated airborne particulate samples were collected in Chicago, IL and over southern Lake Michigan during July 1994 and January 1995. This field sampling was conducted as part of the Atmospheric Exchange Over Lakes and OceanS (AEOLOS) project (Caffrey et al., 1996; Offenberg and Baker, 1997; Simcik et al., 1997,1998; Franz et al., 1998; Zhang et al., 1999; Caffrey et al., 1998; Zufall et al., 1998; Paode et al., 1998; Offenberg and Baker, 1999). The overall objective was to quantify gradients in atmospheric concentrations over Lake Michigan adjacent to Chicago, IL. The size-segregated EC and OC concentrations measured in these samples show significant differences in relative contribution of carbonaceous species to total particulate matter mass according to particle size.

2. Methods

2.1. Sampling

The details of sampling have been described previously and will be briefly summarized here. Un-paired air samples were collected over 12 h periods using a single sampling apparatus at one mobile and two stationary sampling platforms covering a trajectory from Chicago over southern Lake Michigan to southwestern Michigan (Simcik et al., 1997; Offenberg and Baker, 1999). During July 1994 and January 1995, ambient aerosol samples were collected at these stations to assess the impact of the urban/industrial complex on neighboring Lake Michigan. Numerous physical and chemical parameters,

including organic and elemental contaminant concentrations, were measured during these intensives (Offenberg and Baker, 1997,1999; Simcik et al., 1998; Franz et al., 1998; Zhang et al., 1999; Caffrey et al., 1998; Zufall et al., 1998; Paode et al., 1998; Offenberg and Baker, 1999).

Airborne particles were collected using a Berner-type impactor as described elsewhere (Hauke LPI 150/0, 15 ln3; Hauke GmbH, Gmunden, Austria; Berner and Lurzer, 1980; Wang and John, 1988; Hillamo and Kauppinen, 1991; Kaupp et al., 1994; Poster et al., 1995; Schnelle et al., 1995; Kaupp and McLachlan, 1999; Offenberg and Baker, 1999). Aerodynamic equivalent diameter size cuts for this impactor are 0.15, 0.45, 1.4, 4.1 and 12.2 μm . Uncoated aluminum foil was used for impaction substrates, thereby eliminating analytical complexities associated with the use of grease. Although some fraction of particles are not collected on uncoated substrates, earlier work demonstrated that because these particles are also not collected on subsequent stages that this ‘particle bounce’ does not bias the measurement of particle size distributions with the Berner impactor (Berner and Lurzer, 1980).

2.2. Analysis

Prior to sampling, impaction substrates were weighed to 0.0001 mg (Mettler UMT-2), and were re-weighed upon returning to the laboratory to determine the particulate matter (PM) concentration for each size fraction of every sample. Foil substrates were equilibrated for 24 h in a dessicator containing silica gel prior to all weighings. Approximately half of each foil (determined gravimetrically) was analyzed for elemental and organic carbon by the thermal-optical method (Birch and Cary, 1996). This method employs a temperature and atmosphere controlled oven to produce an operational EC/OC speciation. Carbon evolved during the temperature/atmosphere program is oxidized to CO_2 followed by reduction to CH_4 and quantified by flame ionization detection. Instrument calibration is performed via injection of a known mass of methane into the apparatus. Collection of samples on foil impaction substrates precludes the use of optical pyrolysis correction to the EC/OC speciation. Thus, carbon speciation was determined manually from the plots of flame ionization detector response vs. time. This analysis was performed by Sunset Laboratory, Inc. (Forest Grove, OR), and resulted in a total carbon concentration with an operational EC/OC speciation. Chemical analysis of the remaining half of the aluminum foil disks for polycyclic aromatic hydrocarbon concentrations is described elsewhere (Offenberg and Baker, 1999).

2.3. Quality assurance

Replicate analysis of impactor substrates indicates good analytical precision, with relative percent variations

averaging 12% for OC analysis and 8% for EC analysis ($n = 4$ for each). Further comparisons among various sampling equipment and analytical methods was not possible within this study, as no concurrent samples were collected for this purpose. However, comparisons of this type have previously been performed in great detail (Hering et al., 1990; McDow and Huntzicker, 1990; Turpin and Huntzicker, 1995; Turpin et al., 1994; Cui et al., 1997,1998). While the EC/OC speciation is often quite different when analytical methods are compared, the precision of a single method is often quite good, with coefficients of variation between replicates being as low as a few percent (Turpin et al., 1994).

Procedural impactor substrate blanks processed concurrently with field samples are used to quantify operational detection limits. The total (sum of all stages) blank-based limits of detection (LOD), calculated for each species as three times the respective average field blank, are approximately 0.01 and 0.23 g m^{-3} for elemental and organic carbon, respectively. Both species were above the LOD in all samples collected.

3. Results and discussion

3.1. Particle mass

Particulate matter concentrations are presented elsewhere (Simcik et al., 1998; Offenberg and Baker, 1999) and briefly described here. Particulate matter concentrations (Σ -PM), calculated as the sum of all size fractionated particulate matter concentrations, ranged from 8.2 to 36.6 $\mu\text{g m}^{-3}$ in all samples collected. Aerosol concentrations collected by the Berner-type impactor were lower than those collected simultaneously by dichotomous samplers (10 μm inlet and 2.5 μm cut size; Simcik et al., 1998). On average, the impactor collected $59 \pm 17\%$ and $45.7 \pm 7\%$ (avg \pm S.D.) of the aerosol mass measured by the dichot over Lake Michigan and in Chicago, respectively. This is likely due to incomplete collection by the Berner-type impactor of large aerosols which often contain a significant portion of the particle mass, especially in urban atmospheres (Hinds, 1982). Direct comparison of simultaneous impactor results between locations is not possible due to non-coincident sampling. However, for all samples collected during the summer intensive, the urban atmospheric aerosol concentration measured by the dichot was significantly elevated relative to paired over-water samples and to the respective rural samples ($27.7 \pm 3.1 \mu\text{g m}^{-3}$ Chicago, $n = 19$; $17.4 \pm 1.8 \mu\text{g m}^{-3}$ Over-water, $n = 17$; $20.8 \pm 3.8 \mu\text{g m}^{-3}$ Rural, $n = 8$; avg \pm S.E.; Simcik et al., 1998). There was no significant difference between the dichot aerosol concentrations measured during summer ($n = 19$) and during winter ($n = 3$; $30.9 \pm 1.6 \mu\text{g m}^{-3}$).

Geometric mean diameters (GMD) of Σ -PM, as measured by the Berner-type impactor, averaged 1.71 μm a.e.d. and ranged from 0.72 μm a.e.d. on 19 July 1994 over Lake Michigan to 2.39 μm a.e.d. on 21 January 1995 in Chicago. There were no significant differences in Σ -PM GMD's between any of the sampling locations or periods. Geometric standard deviations (σ_g) of Σ -PM averaged 3.3 and did not depend significantly on location or sampling period. However, Σ -PM GMDs positively correlate with measured Σ -PM σ_g 's, such that 57% of the variability in the observed GMD can be explained by changes in the σ_g of particulate matter, indicating that increases in σ_g are due to additions of large aerosols which greatly increase Σ -PM. Resuspension of urban dust and soils is likely the source of the increase in large aerosols which fortify the Σ -PM and associated Σ -PM GMD. Accordingly, decreases in Σ -PM GMD correspond to decreases in Σ -PM σ_g , and are likely due to decreases in the concentration of largest particle concentrations due to dry particle deposition.

3.2. EC/OC concentrations

Total particle bound concentrations (sum of all size fractions) of organic carbon ranged from 1.26 to 2.49 $\mu\text{g m}^{-3}$ in the urban atmosphere and from 0.94 to 3.04 $\mu\text{g m}^{-3}$ in the coastal atmosphere. Total elemental carbon concentrations ranged from 0.19 to 0.45 $\mu\text{g m}^{-3}$ in the urban atmosphere and from 0.15 to 0.96 $\mu\text{g m}^{-3}$ in the coastal atmosphere. The apparent similarity between over-water and urban carbon concentrations may in part be due to summer sampling of urban air during north winds, which resulted in lower than expected Σ -PM concentrations. Southwesterly winds increase total PAH concentrations by approximately 12-fold over regional background (north wind) concentrations (Simcik et al., 1997). It is likely that a similar increase in both EC and OC may occur when winds carry the urban aerosols out over the lake, but this can not be substantiated by this data set.

Since most analysis provide operationally defined carbon speciation, comparison of the of the EC and OC concentrations measured here to most of the available literature is not possible due to different analytical methods (Birch and Cary, 1996). However, total carbon (TC = EC + OC) is quite comparable between analytical methods, and as such comparisons follow. Generally, highest TC concentrations are observed in urban locations, with values often exceeding 25 $\mu\text{g TC/m}^3$ (Table 1; Pimenta and Wood, 1980; Kadowaki et al., 1990; Valaoras et al., 1988; Nunes and Pio, 1993; Chen et al., 1997). The lower limit of atmospheric concentrations is $< 2 \mu\text{g TC m}^{-3}$ and is found in rural/remote atmospheres (Ohta and Okita, 1984; Andreae, 1983). The TC concentrations measured herein ($2.2 \pm 0.7 \mu\text{g m}^{-3}$; range 1.1 to 3.8 $\mu\text{g m}^{-3}$) are lower than might be expected for

Table 1
Literature values of total carbon (TC) in aerosols from various atmospheres

	Location	PM $\mu\text{g m}^{-3}$	TC $\mu\text{g m}^{-3}$	$\mu\text{g g}^{-1}$	OC/TC	Reference
Urban	Estarreja, Portugal	124	27.7	0.22	0.57	Nunes and Pio (1993)
	Ping Tung, Tiawan	—	95.5	—	0.58	Chen et al. (1997)
	Chicago, IL	13.9	2.2	0.16	0.84	This work
Suburban	Aviero, Portugal	—	19.2	—	0.38	Nunes and Pio (1993)
	Academy, CA	35.9	10.8	0.30	0.84	Chow et al. (1996)
Rural	Yosemite, CA	23.7	12.2	0.51	0.86	Chow et al. (1996)
	Sequoia Lower Kaweah, CA	21.1	7.1	0.34	0.72	Chow et al. (1996)
	South Haven, MI	10.4	1.7	0.16	0.82	This work
Remote	Meadview, AZ	—	1.9	—	0.94	Turpin et al. (1997)
	Ivory Coast	—	10.1	—	0.85	Cachier et al. (1989)
Marine	Japanese Coast	—	3	—	0.67	Ohta and Okita (1984)
	Point Reyes, CA	7.3	1.7	0.19	0.72	Chow et al. (1996)
	southern L. MI	15.6	2.2	0.14	0.79	This work

an urban to rural transect, however, many of the other urban values were measured during the late 1970s to mid 1980s, or in areas of especially high total suspended particulate matter concentrations.

All size-segregated samples measured here exhibit a strong correlation between EC and OC concentrations (Fig. 1). Organic carbon is uniformly 3.25 times the concentration of elemental carbon, regardless of particle size, location or season. While the influence of manual attenuation on the EC/OC speciation cannot be excluded as a possible source of this high correlation, similar relationships have been previously reported. Turpin and Huntzicker (1995) suggest that samples with very high degrees of OC to EC correlation may be due to dominance of primary emissions. Alternatively, similar weather patterns in the areas of interest could mix the local and regional emission sources in a like manner, leading to reproducible OC/EC ratios (Turpin and Huntzicker, 1995). However, wind and weather patterns were not constant at these locations, as evidenced by the passing of pressure/temperature fronts and associated precipitation events (Offenberg and Baker, 1997). This indicates that the relative rates of EC and OC emission/production remain proportional to one another. The two species are behaving in the atmosphere nearly identically, with no significant change in the ratio between the two species. However, the relative importance of the two species does change with respect to particle mass, comprising a larger fraction of the total aerosol in small a.e.d. particles than in the large particles.

Measured OC concentrations comprise a significant portion of total suspended particulate matter in all samples. An average of $13 \pm 4.6\%$ (\pm S.D.) of the observed mass of ambient aerosol is organic carbon and $3.0 \pm 1.2\%$ is elemental carbon. When a factor of 1.4 is

applied as an OC mass to organic matter (OM) mass ratio (Turpin et al., 1997), an average of $18 \pm 7\%$ of the aerosol mass can be attributed to organic matter. A seasonal difference is seen at the urban location, with organic matter comprising $26 \pm 4\%$ and $12 \pm 2\%$ of the Σ -PM during summer and winter, respectively. This difference is related to the lower Σ -PM measured during the summer samples, apparently driven by changes in non-carbonaceous material concentrations. This may also be influenced by collection of summer urban samples during periods of northerly winds. However, at the over-water location, the relative importance of organic matter in the total aerosol composition follows a similar trend, with OM accounting for an average of $21 \pm 1\%$ of the aerosol during summer, and $11 \pm 0.1\%$ with during winter. The similarity of this trend at both locations may be indicative of a difference in the sources of aerosol mass during the two seasons.

Concentrations of species were determined on particles of varying size (Table 2). When the above fractional composition analysis is performed by particle size, largest particles tend to have the smallest fraction of carbon composition, while smallest particles contain the greatest proportion of carbonaceous material. Organic matter ranges from 3 ($> 12 \mu\text{m}$ over-water, winter) to 49% (0.15–0.45 μm , urban, summer) of the particulate matter mass collected in the respective size class. Organic matter comprises of 23 ± 12 , 16 ± 7 , 17 ± 7 , 27 ± 11 and $10 \pm 5\%$ (average \pm S.D.) of the particulate matter in each of the five measured size fractions, smallest to largest, respectively.

The greatest seasonal differences are seen in the smallest size fractions at both the over-water and urban locations. In both cases the summer samples contain a greater fraction of organic matter than do the winter

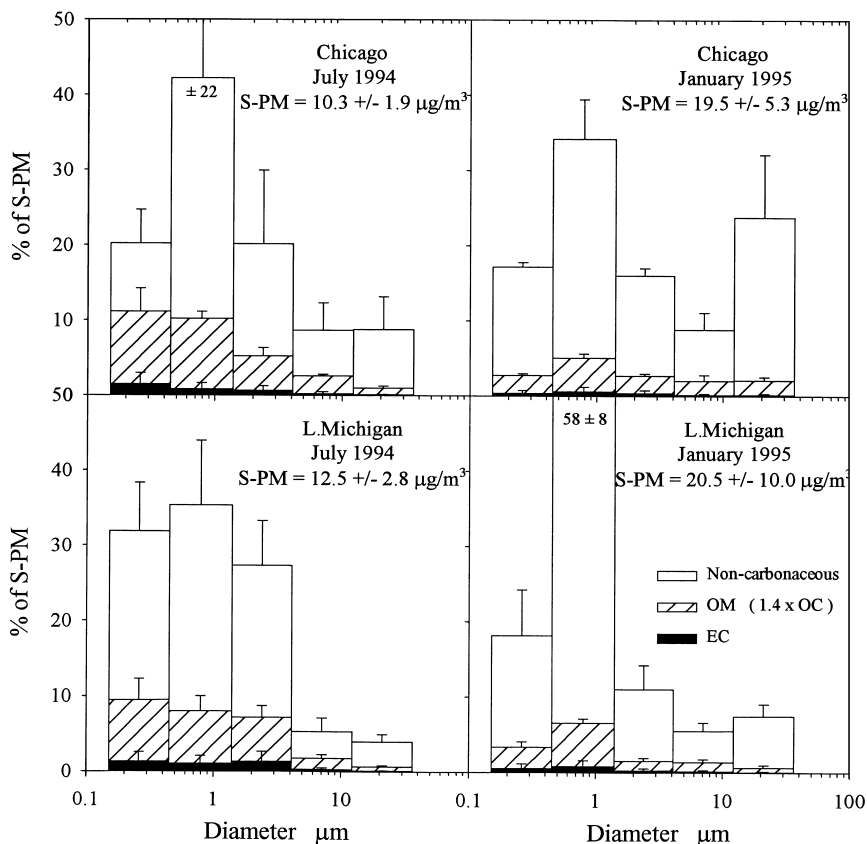


Fig. 1. Elemental and organic carbon concentrations ($\mu\text{g m}^{-3}$) in all size fractions of samples collected at urban, over-water and rural sites around southern Lake Michigan during July 1994 and January 1995.

Table 2

Average concentrations ($\mu\text{g m}^{-3}$), geometric mean diameters (GMD; μm) and geometric standard deviations (σ_g) of size-segregated suspended particulate matter (PM), organic carbon (OC) and elemental carbon (EC) in urban and over-water atmospheres during summer and winter conditions

Cutoff diameter (μm)	Chicago July 1994			January 1995			S. Lake Michigan July 1994			January 1995		
	N winds			SW winds			SW winds			SW winds		
	PM	EC	OC	PM	EC	OC	PM	EC	OC	PM	EC	OC
0.15	2.17	0.16	0.69	3.34	0.08	0.30	3.82	0.16	0.72	3.31	0.12	3.35
0.45	4.09	0.09	0.68	6.53	0.12	0.58	4.23	0.13	0.55	12.2	0.18	0.81
1.4	2.16	0.07	0.33	3.17	0.08	0.31	3.35	0.16	0.46	2.38	0.07	0.17
4.1	0.93	0.02	0.17	1.65	0.03	0.24	0.65	0.03	0.13	1.09	0.03	0.15
12.2	0.95	0.01	0.07	4.86	0.04	0.25	0.47	0.01	0.04	1.52	0.01	0.07
Sum of 5 stages	10.3	0.35	1.92	19.5	0.34	1.68	12.5	0.49	1.90	20.5	0.42	1.55
GMD μm	1.27	0.81	0.88	2.07	1.32	1.76	0.98	0.96	0.88	1.04	0.91	1.02
σ_g	3.56	3.28	3.27	4.79	3.84	4.26	3.03	3.04	3.08	3.16	3.15	3.25

samples. The smallest urban particles collected here (0.15–0.45 μm) contain an average of $45 \pm 5\%$ OM during the summer and $13 \pm 1\%$ during the winter. Like-

wise, smallest particles are comprised of $24 \pm 7\%$ OM during summer and $14 \pm 1\%$ OM during winter. Contrastingly, largest particles in the city are comprised of

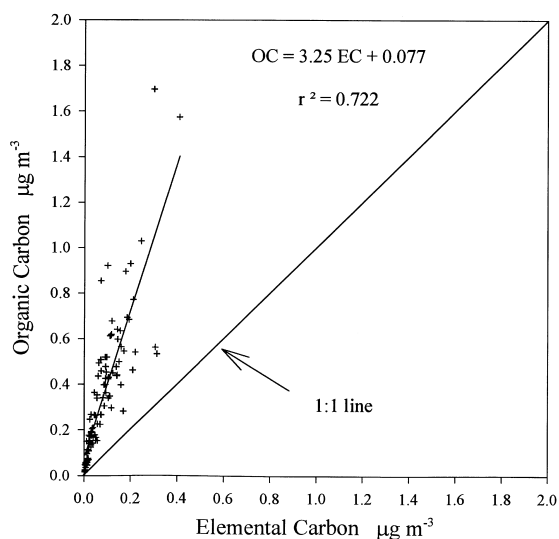


Fig. 2. Average percent contribution (\pm S.D.) of elemental carbon, organic matter ($1.4 \times$ organic carbon) and non-carbonaceous matter to particulate matter concentrations by aerosol aerodynamic equivalent diameter in samples collected from urban and over-water atmospheres during summer and winter conditions.

$11 \pm 3\%$ OM in July and $9 \pm 5\%$ in January as compared with $13 \pm 6\%$ in summer and $8 \pm 3\%$ in winter at the over-water location.

3.3. EC/OC size distributions

Elemental carbon geometric mean diameters ranged from $0.55 \mu\text{m}$ a.e.d. on 26 July 1994 at Chicago to $1.4 \mu\text{m}$ a.e.d. on 21 January 1995 at Chicago. Organic carbon GMDs ranged from $0.60 \mu\text{m}$ a.e.d. on 18 July 1994 at LM1 to $1.9 \mu\text{m}$ a.e.d. on 21 January 1995 at Chicago. Over-water and rural samples were nearly uniformly unimodal for both species, with the modal peak most often in the $0.45\text{--}1.4$ and $0.15\text{--}0.45 \mu\text{m}$ size classes (Fig. 2). Urban samples often exhibited a smaller secondary mode in the $> 12.2 \mu\text{m}$ size class, however, with the limited number of size cuts in this impactor a detailed analysis of this secondary mode is not possible. Furthermore, urban GMDs were significantly larger during the winter than all other collection times and locations for both carbon species. Additionally, geometric standard deviations (σ_g) about these GMDs are significantly larger ($p < 0.05$) in the city for both species during both winter, indicating the mass size distribution is broader for these moieties in the urban atmosphere than over-water locations.

A limited number of field studies have reported the concentrations of elemental and/or organic carbon in aerosols as a function of particle diameter. These size distributions were obtained using various collection and

analytical methods, but will be summarized here for comparison. In a series of field experiments, Venkataraman and Friedlander (1994a,b) showed that primary vehicular emissions contained unimodal EC distributions in the fine fraction ($< 0.5 \mu\text{m}$) while ambient air in Los Angeles shows bi-modal distribution of EC. This multi-modal distribution of carbon has also been seen by other researchers in urban settings, such as Ping Tung, Taiwan (Chen et al., 1997), and in the American desert southwest, in ambient measurements of OC (Turpin et al., 1997). This remote aerosol contains between 0.5 and $2.5 \mu\text{g m}^{-3}$ of total carbon (EC + OC) and the EC/OC concentration ratio averages $1 : 9.3$ for fine aerosols collected as measured with a MOUDI (Turpin et al., 1997). Furthermore, in a field campaign in southern Taiwan, EC and OC were both bimodal, with peaks < 0.32 and $> 5.6 \mu\text{m}$ (Chen et al., 1997). Chen et al. (1997) found that 70.3 and 75.1% of the EC and OC mass was on particles less than $1 \mu\text{m}$, with 81.9 and 84.9% of EC and OC on particles less than $2.5 \mu\text{m}$. They attributed these measurements to fresh primary emissions which dominated the air surrounding the traffic intersection studied.

The size distributions measured here suggest that resuspended particles increase the GMDs and σ_g 's of urban aerosols during southerly winds. This might lead one to believe that urban GMDs and σ_g 's should be larger than those observed at the over-water location during the summer similar to that which was found during the winter. This hypothesis remains untested during this field experiment due to weather conditions during sampling periods.

4. Summary

Size-segregated organic and elemental carbon concentrations in the Chicago-Lake Michigan coastal atmospheres show a strong correlation between OC and EC. No significant difference in the OC to EC ratio was observed between seasons or locations. Organic matter ($1.4 \times$ OC) accounts for an average of 18% of the total aerosol mass collected, but ranges from 3 ($> 12 \mu\text{m}$ over-water, winter) to 49% ($0.15\text{--}0.45 \mu\text{m}$, urban, summer) on size-segregated aerosols. Larger particles contain less carbonaceous material than do the smaller particles, both relative to particulate matter concentrations in each size fraction and on an absolute scale. Finally, an insignificant seasonal difference in concentrations at the over-water location suggests that the atmosphere over lake Michigan experiences nearly steady EC and OC introduction from the neighboring urban atmosphere during periods of southwesterly winds.

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