

Elemental and Organic Carbon in PM₁₀ from Urban, Industrial and Background Areas in Ulsan, a Typical Industrial City, Korea

Van-Tuan Vu, Byeong-Kyu Lee^{*}, Ji-Tae Kim

Department of Civil and Environmental Engineering, University of Ulsan, Daehak-ro 93, Nam-gu, Ulsan, 680-749, Korea

^{*}Corresponding author: E-mail: bklee@ulsan.ac.kr Tel: 82(52)259-2864, Fax: 82(52)259-2629

Abstract — This study investigated the characteristics of carbonaceous species in airborne PM₁₀ at two urban areas (downtown and residence), an industry area and a background area of Ulsan, Korea. Daily PM₁₀ samples were collected on quartz fiber filters using high volume air samplers during winter periods. The elemental carbon (EC) and organic carbon (OC) were analyzed by the thermal-optical transmittance (TOT) method. The concentrations of total carbon (TC), which were calculated as OC + EC, ranged from 1.5 to 9.9 µg/m³ accounting for 7.0-36.1 % of PM₁₀ mass. The average levels of OC and EC at the urban areas were 5.5±2.0 and 1.3±0.5 µg/m³, respectively, which are around 2.5 times higher as compared to those at the background area. The highest concentrations of OC and EC were found at the residential area with an average of 5.5±1.8 and 1.3±0.6 µg/m³, respectively, followed by the downtown and industrial areas. The average OC/EC ratios in the urban, industrial and background areas were 4.1, 6.3 and 4.3, suggesting the presence of secondary organic carbon (SOC). The significant correlations between EC and PM₁₀ concentrations and between EC and OC concentrations were found at the background and urban areas.

Keywords —Elemental carbon; organic carbon; PM₁₀; urban area; OC/EC

1 INTRODUCTION

Carbonaceous species which are known as important components of airborne particles are of concerned because they could affect human health, visibility and even climate change. Main carbonaceous species are elemental carbon (EC) and organic carbon (OC). Cao et al. [1] reported that the carbonaceous aerosol accounted for 38.0 and 32.9 % of airborne PM_{2.5} and PM₁₀ mass, respectively, in four cities of the Pearl River Delta Region, China.

EC (also known as black carbon (BC)) is predominant primary airborne pollutants, mainly released into the atmosphere from the combustion processes of carbon containing materials such as coal, oil or biomass. Due to the absorption and scattering characteristics of the solar radiation, EC is one of the main causes of visibility reduction. EC or BC, having a longer atmospheric lifetime than OC, can greatly contribute to the Earth's radiative balance with estimated climate forcing range from 0.27 to 0.54 OC contains a mixture of organic compounds and a lot of OCs is classified as hazardous air pollutants (HAPs). In particular, some of OCs, such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs), are known as carcinogens or mutagens [3-5]. Also, OC

having estimated climate forcing range from - 0.04 to -0.41 W/m² can contribute to global cooling because aerosol particles contained in OC can reflect sun light back into space [2, 6]. Secondary organic carbons (SOCs) can be formed by the chemical reactions of organic matters or OCs, emitted directly from pollutant sources, with radicals in the atmosphere.

Ulsan is a typical industrial city which has many industrial complexes (ICs) with two national-scale ICs. Thus a lot of OC and EC will be emitted from industrial processes, traffic activities, and biomass burning or forest fire. However, there is no available information on OC and EC in airborne particulate matter (PM) in Ulsan resulting in difficulty for air quality control or management. This study is aimed to investigate the characteristics of carbonaceous species (OC and EC) from different areas with environmental characteristics in a typical industrial city of Korea.

2 METHOD

2.1 Sampling site description

Ulsan Metropolitan City, which has an area of 1.057 km², is located in the southeastern part of Korea and has coastal regions in the south and east areas. Ulsan is the largest industrial city of Korea with a population more than 1.1 million, and has many large industrial complexes, such as a non-ferrous metallic, petrochemical, mechanical and shipbuilding Ics. Elemental and organic carbon contained in airborne PM₁₀ samples were collected from three areas including a background area (BG), an industrial area (IC) and urban areas (UBs), which contain downtown and residential sites, in Ulsan as shown in Fig. 1.

Airborne PM₁₀ samples in the background area were obtained from a mountain surface (Bae-nae-gol) which is enclosed by tall mountains. Thus, the PM₁₀ collected from the background area could be derived from crustal source or pollen and composed of long range-transported particulate matter. Airborne PM₁₀ samples for the urban area were obtained from the roofs of small district offices which located in a typical urban residential area (Mugeo-dong) and a central downtown area of the city (Samsan-dong). As there are busy traffic roads nearby these two sampling sites, the PM₁₀ would be greatly affected by the traffic emissions and wear out emissions of tire and road surface. The sampling site for industrial area was located 500 m away from a petrochemical industrial complex area of Ulsan (Bukog-dong).

2.2 Sample Collection

Daily airborne PM₁₀ samples were collected on quartz microfiber filters (20.3 cm x 25.4 cm, Whatman, Inc., England) from four sites with different environmental characteristics (a background, an industrial, an urban residential, and an urban downtown areas) during winter periods (15th December, 2009- 17th February, 2010) using a high volume air sampler (Tisch Environmental, Inc., USA). The air flow rate for the PM₁₀ sampling was 1.13 m³/min and thus daily air samples collected had an average air volume of 1.627 m³/day-sample.

2.3 Sample Preparation

Quartz fiber filters were pre-cleaned to remove carbonaceous contaminants by firing for 8 h at 650

°C. Several such fired filters were analyzed as blanks for OC and EC. These filters were stored in dark sealed petri dishes at 0°C until use. The exposed filters were stored in a refrigerator at about 4 °C before chemical analysis to prevent evaporation of volatile components. Quartz fiber filters were analyzed gravimetrically for mass concentration using an electronic microbalance with an accuracy of 10⁻⁶ g before and after sampling, in triplicates. Weights were considered valid if triplicates weights on different times were within 3 µg of each other. These filters measured were conditioned for a minimum of 24 h in a controlled temperature/RH chamber at 25°C and 40% RH, both before and after sampling, to minimize the effects of water artifact.

2.4 EC/OC analysis

OC and EC concentrations in PM₁₀ samples were analyzed using a Thermal/Optical Carbon Aerosol Analyzer (Sunset Laboratory, Forest Grove, OR) which is operated according to the National Institute of Occupational Safety and Health (NIOSH) Method 5040. The detailed experimental parameters are presented in Table 1. Field blanks were obtained at each sampling site. Triplicate blank tests were also conducted by the same analytical procedures as the real air samples. The detection limit was 0.2 µg C. The OC and EC concentrations of all the samples were corrected by subtracting the their average values of blank samples from the values obtained from analysis of real air samples.

Table 1. The Experimental conditions for the Sunset Labs semi-continuous ECOC field analysers.

Step	Gas	Hold time (s)	Temperature (°C)
1	He	60	340
2	He	60	500
3	He	60	615
4	He	90	870
5	He	60	over off
6	He:O ₂	45	550
7	He:O ₂	45	625
8	He:O ₂	45	700
9	He:O ₂	45	775
10	He:O ₂	45	850
11	He:O ₂	120	900

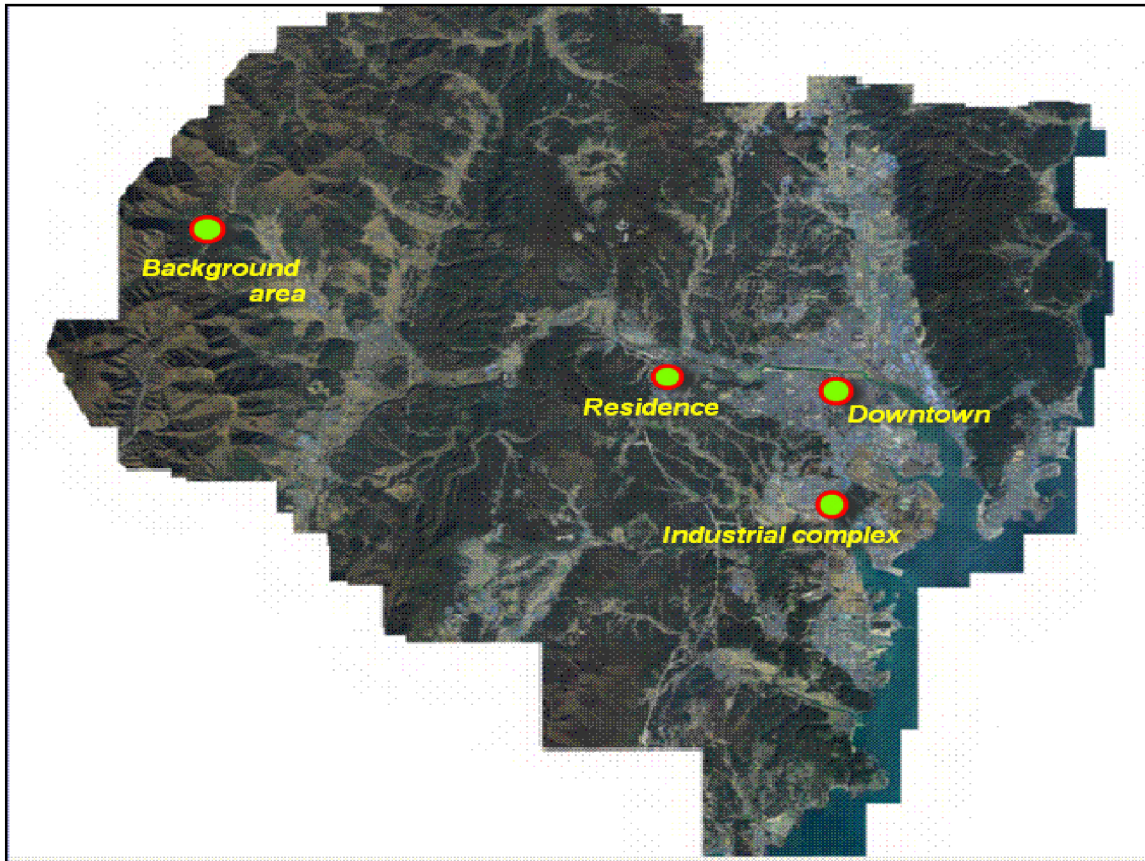


Fig.1. The sampling sites of airborne PM₁₀ in Ulsan, Korea

3 RESULTS AND DISCUSSION

3.1 Concentrations of PM

Table 2 shows the winter concentrations of PM₁₀ and carbonaceous species in PM₁₀ from the background, industrial, and urban residential and downtown areas in Ulsan, Korea. The average PM₁₀ concentration was found highest at the urban area with an average level of 52.2±13.1 µg/m³ which is 2.5 times higher than that at the background area. The average concentrations of PM₁₀ were 57.7±11.5 and 54.0±14.0 µg/m³ at the residential and downtown sites, respectively, which are exceeded the annual ambient air quality standards (AAQs) of PM₁₀, 50

µg/m³, in Korea. The concentration of PM₁₀ at the background site was ranged from 5.4 to 36.6 µg/m³, with an average level of 20.9±13.7 µg/m³. The average concentration of PM₁₀ from the industrial area was 44.8 ± 12.7 µg/m³, which is 2.1 times higher than that at the background area. The concentrations of PM₁₀ from the urban areas of Ulsan were slightly higher than those from the urban area of Busan (ranging from 37.9 to 54.0 µg/m³), however they were lower than those from the urban areas of Seoul (ranging from 46.0 to 91.0 µg/m³) [7].

Table 2. The concentrations of carbonaceous species in PM₁₀ from the background, industrial and two sites of urban areas of Ulsan, Korea.

Location	PM ₁₀ (µg/m ³)	EC (µg/m ³)	OC (µg/m ³)	TC (µg/m ³)	EC/PM ₁₀ (mass %)	OC/PM ₁₀ (mass %)	TC/PM ₁₀ (mass %)
Background	20.94±13.69	0.44±0.12	1.87±0.42	2.32±0.51	3.02±1.89	13.68±10.12	16.70±12.07
Industry	44.78±12.67	0.64±0.24	4.25±1.02	4.89±1.13	1.43±0.28	9.63±1.57	11.06±1.38
Urban Area	55.86±12.23	1.28±0.53	5.50±1.98	6.78±2.44	2.26±0.67	9.703±2.66	11.99±3.91
Residence	57.69±11.48	1.29±0.55	5.52±1.76	6.81±2.18	2.21±0.70	9.45±1.44	11.65±1.83
Downtown	54.04±14.01	1.28±0.58	5.47±2.39	6.75±2.95	2.31±0.72	10.02±3.70	12.33±4.38

average concentrations of PM₁₀, EC and OC in Ulsan with those in other cities over the world.

	PM ₁₀ (µg/m ³)	EC (µg/m ³)	OC (µg/m ³)	TC (µg/m ³)	References
Urban	20.94±13.69	0.44±0.12	1.87±0.42	2.32±0.51	
Industrial	44.78±12.67	0.64±0.24	4.25±1.02	4.89±1.13	This study
	55.86±12.23	1.28±0.53	5.50±1.98	6.78±2.44	
	-	8.39	11.1	-	Kim et al. [8]
Commercial	24.1-25.4	3.3-3.6	3.4-3.6	6.7-7.2	Kaneyasu et al. [9]
	83.52±16.74	6.86±1.16	9.45±2.01	-	
Residential/Industrial	73.11±22.40	5.09±0.95	10.16±2.59	-	Ho et al. [10]
Urban	80.01±30.06	1.48±0.36	5.52±1.13	-	
	110	6.1	14.5	-	Lin and Tai [11]
	172.6±98.3	8.9±5.1	21.2±1.16.0	30.2±20.4	Zhang et al. [12]
Region	161.7±114.4	10.4±6.8	29.4±22.2	39.8±28.9	Cao et al. [2]
Urban	182±48	12.4±5.1	37.3±10.5	52.2±16.3*	Venkataraman et al. [13]
	10.8-14.4	1.4-1.6	1.3-1.8	2.7-3.2	Roosili et al. [14]
	21.1-29.6	3.0-5.4	3.6-5.5	6.6-10.9	
Urban	5.8±0.7	0.1±0.1	2.2±0.5	2.3±0.4	
Forest	24.0±8.1	0.6±0.1	5.0±1.1	5.5±1.1	Zappoli et al. [15]
Commercial	38.7±12.2	1.0±0.4	6.2±2.0	7.2±2.0	
	52.5	5.7	19.7	25.4	Chow et al. [16]

The concentrations of PM₁₀ from Ulsan were lower than those in Hong Kong and Taiwan and cities in China and India [2, 10-13] as shown in Table 3. However, they were much higher than those in cities in Japan and European [14-15].

3.2 Concentrations of carbonaceous species

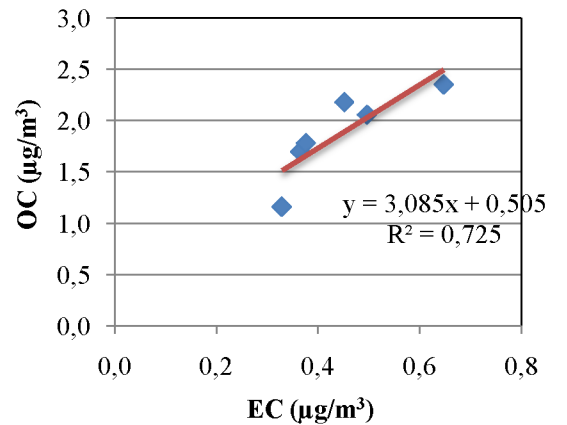
The winter average concentration of EC in Ulsan was $1.28 \pm 0.53 \mu\text{g}/\text{m}^3$ at the urban area, which is much higher than those at the industrial area ($0.64 \pm 0.24 \mu\text{g}/\text{m}^3$) and at the background area ($0.44 \pm 0.12 \mu\text{g}/\text{m}^3$). The winter average concentration of OC in Ulsan was also found highest at the urban area which has an average level of $5.50 \pm 1.98 \mu\text{g}/\text{m}^3$, followed by the industrial area ($4.25 \pm 1.02 \mu\text{g}/\text{m}^3$). The average concentration of OC at the background area was $1.87 \pm 0.42 \mu\text{g}/\text{m}^3$ which is much lower than those at the industrial and at the urban areas. The industrial area would be mainly affected by the emissions from industries in petrochemical complex. However, the highest concentrations of PM₁₀ and carbonaceous species were found at the urban area. It could be explained that this area is greatly affected both traffic and industrial emissions. In fact, the urban downtown areas are located in a short distance from the roads with high traffic volume and industrial complex. The urban areas are mainly and easily affected by traffic emissions from vehicles passing nearby the sampling sites. In addition, the urban areas were located in the downwind areas of the prevailing winds which could pass through the petrochemical industrial areas. Thus some of the industrial emissions could be transported to the sampling sites in the urban areas. Fang et al. [17] found that the order of OC and EC concentrations in Asia from highest to lowest was urban > industrial > island sampling sites which is a similar trend to this study result (urban > industrial > background).

The OC fractions were the dominant parts of the identified total carbons, which contribute 82.34 % of the total carbon mass. The concentrations of the total carbons (TC) which calculated by summing up the EC and OC concentrations were 6.78 ± 2.24 , 4.89 ± 1.13 , and $2.32 \pm 0.51 \mu\text{g}/\text{m}^3$ for the urban, industrial, and background areas, respectively. However, the total carbons only contributed 16.7, urban, and industrial areas 16.7, 12.0, and 11.1 % to the PM₁₀ mass at the background, the industrial, and the urban areas, respectively. Shwarz et al. [18] reported that OC accounted for 16.0 and 15.0 % of PM₁₀ mass and EC accounted for 2.3 and 3.2 % of the PM₁₀ mass for the suburban and downtown sites in Prague, Czech, respectively. The percentage of the total carbons to the total mass of PM₁₀ at the background area in Ulsan was higher than those at the industrial and the urban area. It could be explained by the transport of OC and EC emitted from urban traffic to the background area. Ho et al.

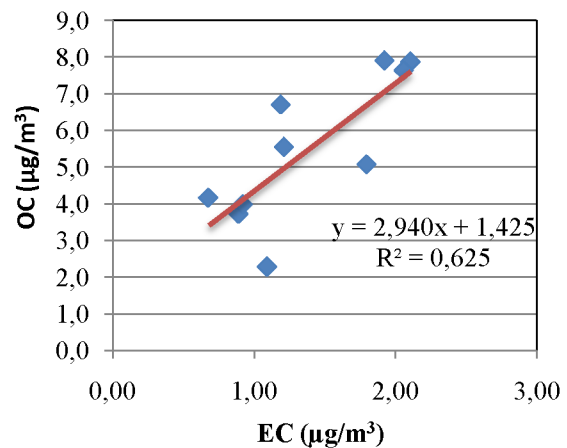
[10] reported that the concentration of the total carbons, particularly OC emitted as primary aerosol, from the urban areas was higher than that at the non-urban areas. As the background has much lower PM₁₀ mass concentration as compared other areas, the even small increase in carbon mass (due to the OC and EC transported from traffic areas or urban areas) can increase the carbon fraction to the PM₁₀ mass in the background area in Ulsan. The concentrations of OC and EC in Ulsan were much lower than those in Seoul of Korea, Hong Kong, Taiwan, Beijing and Guangzhou in China, and Mumbai in India (as shown in Table 3). They showed similar ranges to those in European cities.

3.3. Relationship between EC and OC and PM₁₀

There were significant correlations between the OC and EC concentrations at the background area ($R^2 = 7.3$, $p < 0.01$) and at the urban areas ($R^2 = 6.3$, $p < 0.01$) in Ulsan (Fig. 2).



(a)

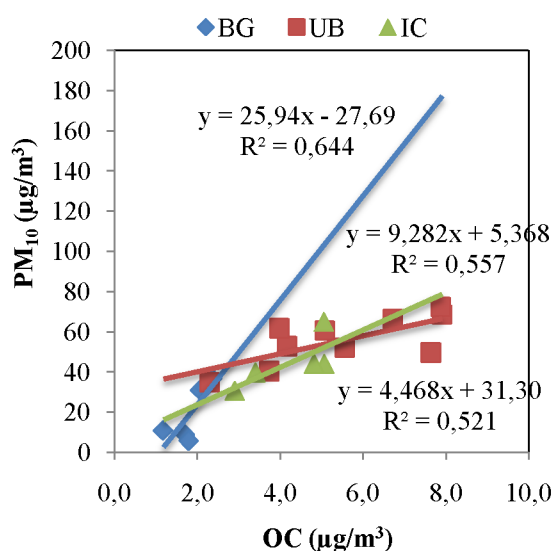


(b)

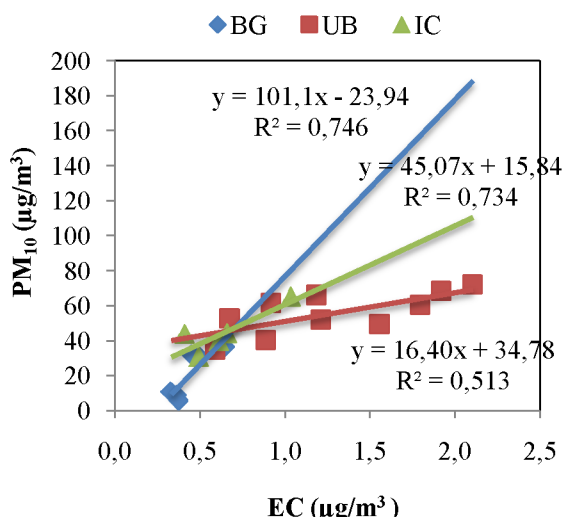
Fig. 2. The correlation between the OC and EC concentrations at the background area (a) and the urban area (b).

This indicates that the OC and EC obtained at the background and urban areas have common or similar emissions sources such as traffic emissions. The strong correlations between OC and EC from urban areas were also found in previous similar studies [1, 10, 19-20]. The significant correlation identified between EC and OC from the background area may represent that the EC and OC originated from traffic sources or urban areas would be transported to the background area. A significant correlation was not identified between OC and EC at the industrial area. It could be explained the EC and OC from the industrial area could be affected by the complicated sources such as traffic, ship and industrial emissions.

Fig. 3 shows the correlations between the PM₁₀ and the EC and OC concentrations in Ulsan. The correlations between OC and PM₁₀ concentrations were 0.64, 0.52 and 0.56 at the background, urban and industrial areas, respectively. In addition, the correlations between EC and PM₁₀ concentrations were 0.74, 0.51 and 0.73 at the background, urban and industrial areas, respectively. The correlation between EC and PM₁₀ at the background and industrial areas were higher than those between OC and PM₁₀. These high correlations between the PM₁₀ and the EC (or OC) show the consistent contribution of carbonaceous species to PM₁₀ [21-22]. Ye et al. [21] also found the significant correlation between PM₁₀ with the OC ($r = 0.82$, $p < 0.01$) and EC ($r = 0.45$, $p < 0.05$).



(a)



(b)

Fig. 3. The correlation between PM₁₀ mass with the OC concentration (a) and EC concentration (b) from background (BG), urban (UB) and industrial areas (IC).

3.4 OC/EC ratios and sources of carbonaceous

Novakov et al reported an extensive review of OC/EC ratios, based on the previously published papers, in many countries and regions over the world [23]. The origin and transported sources of carbonaceous particles can be estimated based on the ratio of OC to EC. Park et al. [24] reported that the lower OC/EC ($OC/EC < 2$) in the urban area of Seoul may be evidence for primary organic local sources such as traffic emissions of organic carbon. Cao et al. [1] also reported that effects of motor vehicle exhaust was approximately 2 in ratio of OC/EC. In contrast, the OC/EC exceeding 2.0 was trace of the presence of secondary organic aerosols [25-26]. The OC/EC ratios were found 4.3, 6.3 and 4.1 for the background, industrial and urban areas of Ulsan. The OC/EC ratio in the industrial area was higher than those in the urban and background areas. The OC/EC ratios in Ulsan were much higher than the average value (2.3) in OC/EC ratios in PM₁₀ at cities in Asian countries [17]. However, Lee et al. [27] reported the OC/EC ratios measured at two Korean background areas (Kosan and Kwangwha), which are located far away 500 km, were 9.94 ± 2.36 and 6.77 ± 1.70 , respectively. They used different analytical method, which utilizes MnO₂ as oxygen donor for carbon oxidation (MNO) method, from this study. In addition, Park et al. [24] and He et al. [21] reported that the OC/EC ratios by using a MNO method in an industrial city (Shiwa) and a metropolitan general city (Gwangju) in Korea were 5.06 and 4.67, respectively. The high OC/EC ratio (4.98) was also reported by Castro et al. [28]. The high ratios of OC/EC concentrations from the

background, industrial and urban areas in Ulsan (much exceeding 2.0) suggests that there might be the presence of secondary organic carbon in PM₁₀ [10].

3.5 Estimate of secondary organic carbon (SOC)

Secondary organic carbon (SOC) which formed from the reaction of volatile organic matter or semi-volatile organic matter with the radical gases, is one of the dominant forms of organic carbon in the atmosphere. However, because of complex atmospheric reactions, it is very difficult to determine the exact amount of SOC concentration directly. Due the similarity in sources of EC and primary OC, Turpin and Huntzicker [29] suggested the equation to calculate the SOC as follows:

$$OC_{\text{sec}} = OC_{\text{tot}} - OC_{\text{pri}} = OC_{\text{tot}} - EC \times (OC/EC)_{\text{pri}}$$

where OC_{sec} is the secondary OC, OC_{tot} is the total OC; $(OC/EC)_{\text{pri}}$ is the ratio between primary OC and EC.

However, the $(OC/EC)_{\text{pri}}$ were affected by a variety of sources and by temporal, locational and meteorological conditions. Thus, it is not easy to estimate exact $(OC/EC)_{\text{pri}}$. Castro et al. [28] found that the minimum value in OC/EC ratios could be exclusively primary carbonaceous compounds in many cases. Therefore, this study applied the following equation:

$$OC_{\text{sec}} = OC_{\text{tot}} - EC \times (OC/EC)_{\text{min}}$$

where OC_{sec} is the secondary OC, OC_{tot} is the total OC; $(OC/EC)_{\text{min}}$ is a minimum value in the ratio of OC to EC.

In the estimate of the SOC concentration in Ulsan, the $(OC/EC)_{\text{mi}}$ values obtained for the background, industrial and urban areas were of 3.4, 2.9 and 2.1, respectively. The SOC concentrations estimated for the background, industrial and urban areas in Ulsan were shown in Table 4.

Table 4. The SOC concentrations from the background, industrial and urban areas of Ulsan.

Area	$(OC/EC)_{\text{min}}$	SOC ($\mu\text{g}/\text{m}^3$)	SOC/OC %
Background	3.56	0.30	15.81
Industry	2.90	1.80	42.46
Urban areas	2.10	2.58	47.01

The calculated concentrations of SOC at the background area ranged from 0.04 to 0.81 $\mu\text{g}/\text{m}^3$ with an average concentration of 0.30 $\mu\text{g}/\text{m}^3$, accounted for 15.8% of the total organic carbon

(TOC). The average SOC concentration was 1.8 $\mu\text{g}/\text{m}^3$, accounted for 42.5 % of the TOC at the industrial area. The highest concentration of SOC was found at the urban area with an average level of 2.6 $\mu\text{g}/\text{m}^3$. The estimated SOC concentrations contributed to 25.7-66.0 % of the TOC at the urban area. Therefore, the SOC could be sometimes dominant contributor to the TOC. Cao et al. [1] reported that SOC accounted for 17.9-53.4 % of the TOC in Pearl river delta region, China. The estimated SOC contribution to the TOC in Ulsan seems to match with the study results reported by Cao et al. [1].

4 CONCLUSION

This study investigated the characteristics of carbonaceous species in airborne PM₁₀ from the background, industrial and urban areas with different environmental conditions in Ulsan, a typical industrial city, Korea. The highest concentrations of PM₁₀, elemental carbon, and organic carbon were found from the urban areas followed by the industry and background areas in order. The total carbons (EC+OC) contributed to 11.1, 12.0 and 16.7 % to the PM₁₀ mass concentrations at the industrial, urban and background areas, respectively.

Significant correlations between OC and EC concentrations were found at the background and urban areas. Also, high correlations between PM₁₀ and OC (or EC) concentrations were identified from all the sampling sites in Ulsan. This represents carbonaceous species are important constituents to PM₁₀ and their emission reduction strategies are vital for reduction of ambient particle concentrations. The high OC/EC ratios, ranging from 4.1 to 6.1, in Ulsan indicate SOC greatly contribute to the TOC. The estimated SOC concentrations were 0.30, 1.80 and 2.58 $\mu\text{g}/\text{m}^3$, which accounted for 15.8, 42.5 and 47.0 % of the TOC at the background, industrial and urban areas in Ulsan, respectively.

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