

# Seasonal Characteristics of Fine Particulate Carbonaceous Species in Taiyuan, North China

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**Abstract:** To characterize seasonal carbonaceous aerosol pollution in Taiyuan, a typical city in North China that mainly relies heavily on coal, a total of 124 PM<sub>2.5</sub> samples were collected from August 2018 to the next May. The annual mean PM<sub>2.5</sub> concentration was  $83.8 \pm 48.5 \mu\text{g m}^{-3}$ , with a seasonal rank of winter ( $117.4 \pm 47.6 \mu\text{g m}^{-3}$ ) > spring ( $79.2 \pm 34.3 \mu\text{g m}^{-3}$ ) > fall ( $67.3 \pm 34.7 \mu\text{g m}^{-3}$ ) > summer ( $31.8 \pm 6.5 \mu\text{g m}^{-3}$ ), suggesting that fine particulate pollution was still serious in cold seasons. Organic carbon (OC) and elemental carbon (EC) showed similar seasonal patterns with PM<sub>2.5</sub>. The mean concentration values of OC in summer, fall, winter, and spring were  $5.1 \pm 0.9$ ,  $11.8 \pm 6.4$ ,  $22.1 \pm 14.9$ , and  $12.2 \pm 6.7 \mu\text{g m}^{-3}$ , respectively. The mean concentration values of EC in summer, fall, winter, and spring were  $1.5 \pm 0.3$ ,  $2.5 \pm 1.6$ ,  $4.4 \pm 2.8$ , and  $2.4 \pm 1.5 \mu\text{g m}^{-3}$ , respectively. The proportion of total carbon aerosol (TCA) was about 31.7%, 33.8%, 30.0%, and 27.0% in PM<sub>2.5</sub> in summer, fall, winter, and spring, respectively. The good correlation between OC vs. EC and the high value of OC/EC suggests that coal and biomass combustion were the main emissions in cold seasons, aggravated by adverse meteorological conditions and the dustpan-shaped terrain. The mean annual secondary organic carbon (SOC) concentration was  $6.1 \pm 7.1 \mu\text{g m}^{-3}$ , representing 38.7% of the OC content. The present results presented the serious carbonaceous particulate pollution, which might affect haze pollution in cold seasons.

**Keywords:** Taiyuan; carbonaceous species; elemental carbon; organic carbon; PM<sub>2.5</sub>



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## 1. Introduction

Fine particulate matter (PM<sub>2.5</sub>) is currently one of the major pollutants which can essentially cause severe haze pollution and threaten public health [1,2]. In recent years, heavy haze episodes have occurred frequently in large areas of China [3–5], and numerous in-depth studies have been conducted in those urban agglomerations with those economically developed 1st/2nd-tier cities, such as North China Plain [3,6–8], Guanzhong Plain [9], Yangtze and Pearl River Deltas [10,11], and Sichuan Basin [12,13] in the context of the national government's vigorous air pollution control. As a result, air quality in many megacities have significantly improved in recent years [14,15]. However, there are still many areas where pollution is very serious, especially in some small/medium-sized cities [4]. This suggests that China still has a long way to go to combat air pollution.

In 2018, Fenwei Plain (i.e., the general name for Fenhe & Weihe Plains) was included for the first time in the main battlefield of “The Blue-Sky Defense War”, launched by the Ministry of Ecology and Environment of China (MEEC). This is mainly related to its fragile ecological environment, more coal, oil, and other chemical enterprises, and serious atmospheric pollution. Taiyuan is the provincial capital city of Shanxi Province and is located in the northernmost part of the Fenhe Plain. Strong emission sources, such as metallurgy and coking, and unfavorable topography and meteorological conditions, made urban ambient air pollution in the Fenhe Plain very serious. According to MEEC ([www.mee.gov.cn](http://www.mee.gov.cn), accessed on 1 June 2021), the annual average percentage of good/excellent days with an air quality index (AQI) less than 100 [14,16] in the Fenwei Plain was only

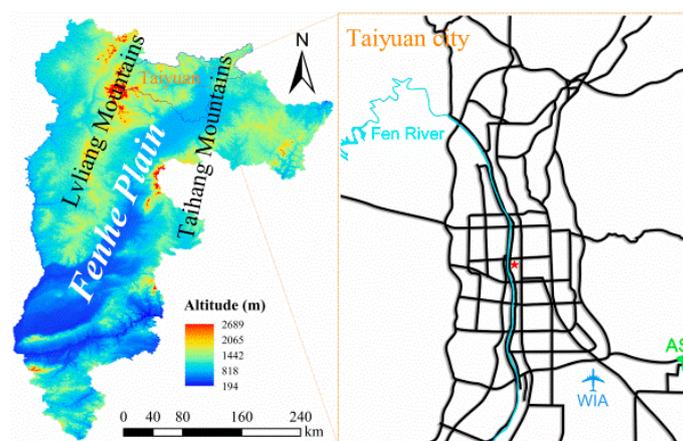
54.3%, and Taiyuan air quality ranked 7th from the bottom in cities across the country in 2018. However, comprehensive information on ambient carbonaceous aerosol pollution in the Fenhe Plain is relatively few.

Previous studies have shown that about 20–50% of  $PM_{2.5}$  mass concentration was carbonaceous aerosols in urban regions [17–20]. Carbon-containing aerosol usually exists in the form of EC and OC. EC is a primary pollutant usually emitted by incomplete combustion of fossil fuels and biomass fuels, while OC can be both a primary and a secondary pollutant [21]. Many studies have shown that carbon aerosols significantly affect the environment and human health. For example, EC has a strong heat and light absorption effect, and OC has a scattering effect on light. When combined, they will seriously damage the radiative balance of climate change in the ground-air system of atmospheric visibility. In addition, OC is a mixture of hundreds of organic compounds, some of which are mutagenic and/or carcinogenic and can seriously harm human health [22,23]. Therefore, to grasp the carbonaceous composition of atmospheric fine particles in Taiyuan and provide suggestions for the local government to formulate emission control policies, this study investigated the seasonal information of fine particulate, and its carbonaceous species has been conducted in Taiyuan from August 2018 to May 2019.

## 2. Experiments

### 2.1. Sample Collection and Analysis

The sampling site was located on the roof of a commercial building (~65 m above ground) in downtown Taiyuan city with some institutions, major traffic roads (Riverside Roads in the west and Yingze Street in the north) and Fen River (The second largest tributary of the Yellow River) adjacent to it (Figure 1).



**Figure 1.** The sampling site (red star) in downtown Taiyuan city with the Wusu international airport (WIA, blue plane) and the agrometeorological station (AS, green pin).

The 90 mm quartz membrane prebaked at 600 °C for 4 h was used for particulate matter collection using a mid-volume particle collector with a flux of 100 L min<sup>-1</sup> (KC-6120). A total of 124 effective membrane samples, of which 15 (1–15 August), 41 (16–20, 22–30 September; 9–23 October; 4–15 November), 36 (12–29 December 2018; 1–9 January 2019; 21–29 February) and 32 (15–22 March; 15–23 April; 1–15 May) in summer, fall, winter and spring, respectively, were collected and analyzed.

The mass concentration of  $PM_{2.5}$  was determined using an electronic balance (FA1004, Lichen, China) after equilibration in a chamber at constant relative humidity (~35%) and temperature (~23 °C) for 24 h. In addition, a DRI (2001A) thermal/optical reflectance carbon analyzer was used to analyze fine particulate carbonaceous species. The minimum determined limits of DRI were 0.20  $\mu\text{gC cm}^{-2}$  and 0.82  $\mu\text{gC cm}^{-2}$  for EC and OC, respectively [2].

### 2.2. Meteorological Parameters

The meteorological characteristics include ambient temperature (T), relative humidity (RH), wind speed (WS) and direction (WD), visibility (V) and rainfall (RF), solar irradiance (SI) and sunshine hours (SH). WS/WD and T/RH were monitored using a self-recording anemometer (ZCF-5, Sipeik, Shanghai, China) and hygrothermograph (SW-572, SNDWAY, Qingdao, China) at the sampling site. V and RF were obtained from the Wusu Airport (WIA, 12.9 km from the sampling site). SI and SH were obtained from the agrometeorological station (AS, 21.5 km from the sampling site) (Figure 1) [24].

## 3. Results and discussion

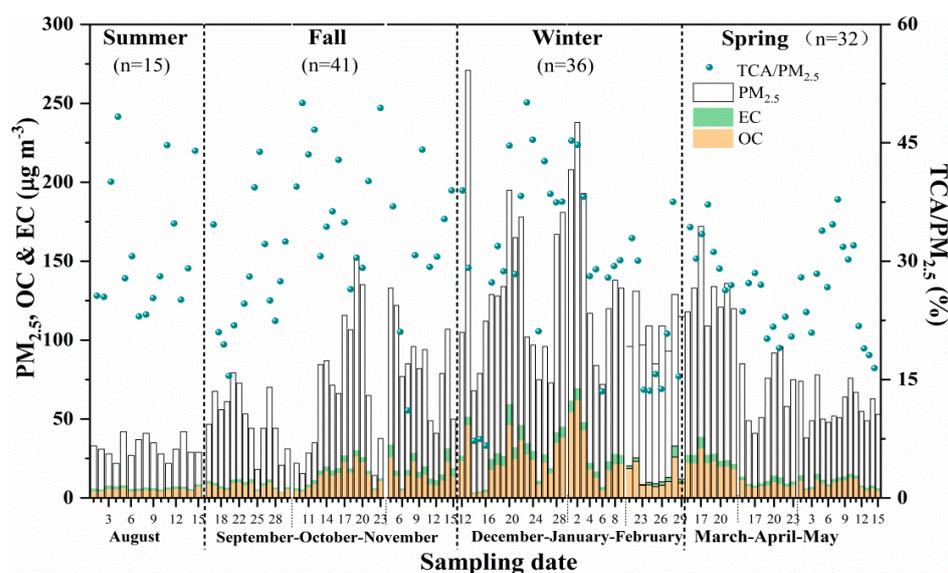
### 3.1. PM<sub>2.5</sub> Levels and Its Carbonaceous Species

The daily mass concentrations of PM<sub>2.5</sub>, OC and EC and total carbonaceous aerosol (TCA) contribution percentage to PM<sub>2.5</sub> are illustrated in Figure 2 and summarized by season in Table 1. The annual mean concentration of PM<sub>2.5</sub> was  $83.8 \pm 48.7 \mu\text{g m}^{-3}$ , with a range of 14.3–269.7  $\mu\text{g m}^{-3}$ , which was about 2.4-fold greater than the 2nd annual limit value ( $35 \mu\text{g m}^{-3}$ ) according to China National Ambient Air Quality Standard (NAAQS) (GB3098-2012). The seasonal mean PM<sub>2.5</sub> concentration was the highest in winter ( $117.4 \pm 47.6 \mu\text{g m}^{-3}$ ), followed by spring ( $79.2 \pm 34.3 \mu\text{g m}^{-3}$ ) and fall ( $67.3 \pm 34.7 \mu\text{g m}^{-3}$ ), and the lowest in summer ( $31.8 \pm 6.5 \mu\text{g m}^{-3}$ ). Compared to 2009–2010 in Taiyuan, reported by He et al. (2015) [25], the PM<sub>2.5</sub> concentration has dropped by 77%, 63%, 55% and 68% reduction in summer, fall, winter and spring. However, fine particle pollution in the present study was still relatively serious, especially in the heating seasons. It can be seen that only the summer PM<sub>2.5</sub> concentration was within the first order ambient air quality standard daily limit ( $35 \mu\text{g m}^{-3}$ ) of MEPC while that in winter, spring and fall were about 3.4, 1.9 and 2.3 times that of the limit.

**Table 1.** Seasonal Mean  $\pm$  SD values of PM<sub>2.5</sub>, carbonaceous species, and meteorological parameters.

Seasons	Summer (n = 15)	Fall (n = 41)	Winter (n = 36)	Spring (n = 32)	Annual (n = 124)
PM <sub>2.5</sub> $\zeta$	31.8 $\pm$ 6.5	67.3 $\pm$ 34.7	117.4 $\pm$ 47.6	79.2 $\pm$ 34.3	83.8 $\pm$ 48.7
OC $\zeta$	5.1 $\pm$ 0.9	11.8 $\pm$ 6.4	22.1 $\pm$ 14.9	12.2 $\pm$ 6.7	14.1 $\pm$ 10.9
EC $\zeta$	1.5 $\pm$ 0.3	2.5 $\pm$ 1.6	4.4 $\pm$ 2.8	2.4 $\pm$ 1.5	2.9 $\pm$ 2.2
TCA $\zeta$	9.7 $\pm$ 1.5	21.4 $\pm$ 11.6	39.8 $\pm$ 26.0	21.9 $\pm$ 12.1	25.5 $\pm$ 19.3
TCA/PM <sub>2.5</sub> $\xi$	31.7 $\pm$ 8.5	33.8 $\pm$ 10.7	30.0 $\pm$ 12.1	27.0 $\pm$ 5.8	30.4 $\pm$ 10.1
OC/EC	3.5 $\pm$ 0.7	5.1 $\pm$ 1.8	5.2 $\pm$ 2.9	4.8 $\pm$ 2.5	5.1 $\pm$ 2.4
SOC $\zeta$	1.1 $\pm$ 0.9	4.0 $\pm$ 3.7	10.4 $\pm$ 10.6	6.4 $\pm$ 3.9	6.1 $\pm$ 7.1
SOC/OC $\xi$	20.2 $\pm$ 14.3	31.1 $\pm$ 17.8	43.3 $\pm$ 21.0	52.0 $\pm$ 19.1	38.7 $\pm$ 21.4
SOC/PM <sub>2.5</sub> $\xi$	3.4 $\pm$ 2.9	6.0 $\pm$ 4.7	7.2 $\pm$ 5.1	8.2 $\pm$ 3.6	6.6 $\pm$ 4.6
SOA $\zeta$	1.7 $\pm$ 1.4	6.4 $\pm$ 5.9	16.7 $\pm$ 16.9	10.2 $\pm$ 6.3	9.8 $\pm$ 11.3
SOA/PM <sub>2.5</sub> $\xi$	5.5 $\pm$ 4.6	9.6 $\pm$ 7.5	11.5 $\pm$ 8.2	13.1 $\pm$ 5.8	10.5 $\pm$ 7.3
WS $\xi$	8 $\pm$ 5	6.9 $\pm$ 3.6	7.1 $\pm$ 4.6	10.3 $\pm$ 5.8	8.2 $\pm$ 4.8
T $\xi$	25.1 $\pm$ 2.6	10.6 $\pm$ 5.7	-2.8 $\pm$ 3.8	13.5 $\pm$ 6.0	9.0 $\pm$ 10.0
RH $\xi$	66.7 $\pm$ 7.0	56.5 $\pm$ 14.0	41.1 $\pm$ 13.3	35.9 $\pm$ 15.5	46.5 $\pm$ 17.6
RF $\xi$	32.8 $\pm$ 31.2	9.4 $\pm$ 22.5	2.2 $\pm$ 2.7	9.4 $\pm$ 22.5	9.6 $\pm$ 21.7
V $\xi$	10.8 $\pm$ 3.4	10.8 $\pm$ 4.4	9.5 $\pm$ 4.5	12.6 $\pm$ 3.8	10.9 $\pm$ 4.3
ITI $\xi$	0.17 $\pm$ 0.30	1.0 $\pm$ 1.3	1.2 $\pm$ 1.9	1.37 $\pm$ 1.25	1.1 $\pm$ 1.47
SH $\xi$	5.47 $\pm$ 1.83	4.07 $\pm$ 1.45	3.00 $\pm$ 0.86	6.13 $\pm$ 1.56	4.50 $\pm$ 1.90
SI $\xi$	175.5 $\pm$ 50.6	133.3 $\pm$ 51.2	108.2 $\pm$ 16.4	208.3 $\pm$ 50.3	151.7 $\pm$ 61.2
Solar irradiance	220.3 $\pm$ 36.9	153.9 $\pm$ 64.4	116.0 $\pm$ 35.1	241.4 $\pm$ 58.0	169.9 $\pm$ 72.2

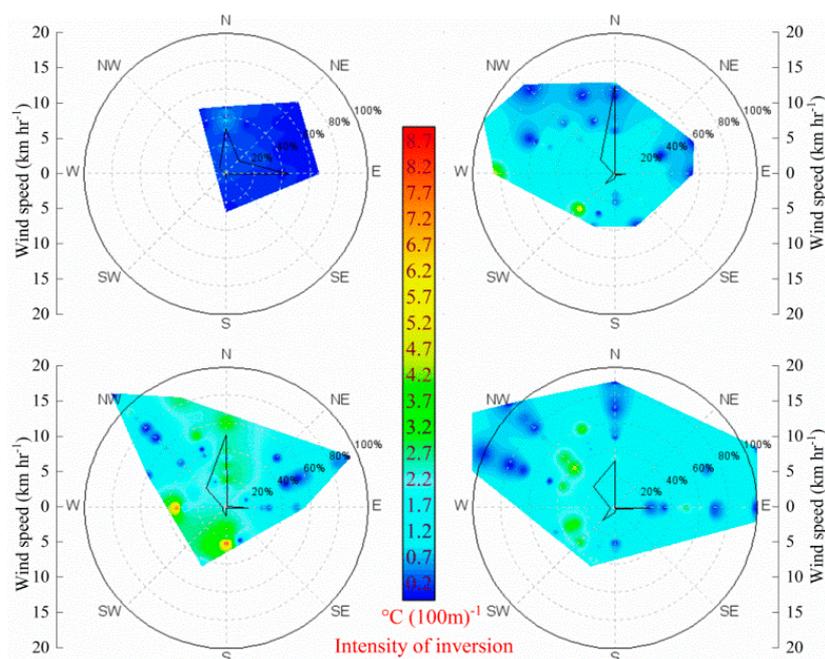
$\zeta$  and  $\xi$ : The units for  $\zeta$  and  $\xi$  are  $\mu\text{g m}^{-3}$  and %, respectively;  $\xi$ : WS ( $\text{km hr}^{-1}$ ), T ( $^{\circ}\text{C}$ ), RH (%), RF (mm), V (km), ITI ( $^{\circ}\text{C (100 m)}^{-1}$ ), SH (hours) and SR ( $\text{W m}^{-2}$ ) represent staged average wind speed, ambient temperature, relative humidity, rainfall, visibility, the intensity of temperature inversion, daily sunshine hours and average daytime solar irradiance during the sampling period, respectively.



**Figure 2.** Daily  $PM_{2.5}$ , OC, EC, and percentage of TCA in  $PM_{2.5}$  during the sampling period.

OC and EC generally presented similar patterns of changes with  $PM_{2.5}$  on a timescale. The daily OC concentrations in summer, autumn, winter, and spring were  $4.1\text{--}7.1\ \mu\text{g m}^{-3}$ ,  $3.3\text{--}26.5\ \mu\text{g m}^{-3}$ ,  $2.9\text{--}62.0\ \mu\text{g m}^{-3}$ , and  $2.8\text{--}14.8\ \mu\text{g m}^{-3}$ , with the seasonal mean values of  $5.1 \pm 0.9\ \mu\text{g m}^{-3}$ ,  $11.8 \pm 64\ \mu\text{g m}^{-3}$ ,  $22.1 \pm 14.9\ \mu\text{g m}^{-3}$ , and  $12.2 \pm 6.7\ \mu\text{g m}^{-3}$ , respectively. The daily EC were  $1.0\text{--}2.1\ \mu\text{g m}^{-3}$ ,  $0.9\text{--}3.9\ \mu\text{g m}^{-3}$ ,  $0.4\text{--}12.9\ \mu\text{g m}^{-3}$ , and  $0.6\text{--}6.7\ \mu\text{g m}^{-3}$ , with the seasonal mean values of  $1.5 \pm 0.3\ \mu\text{g m}^{-3}$ ,  $2.5 \pm 1.6\ \mu\text{g m}^{-3}$ ,  $4.4 \pm 2.8\ \mu\text{g m}^{-3}$ , and  $2.4 \pm 1.5\ \mu\text{g m}^{-3}$ , respectively. This seasonal variation was in line with other studies [21,26–28]. The OC and EC concentration levels were four times and twice as high in winter compared to summer and other seasons. Compared to 2009–2010 ( $13.2\text{--}62.7\ \mu\text{g m}^{-3}$  for OC,  $8.8\text{--}25.4\ \mu\text{g m}^{-3}$  for EC) [25], the OC and EC concentration levels in Taiyuan decreased by about 59–68% and 83–89%, respectively, suggesting significant pollutant reduction have been achieved.

Exacerbation of the pollutants in cold seasons may be due to the source strength and topographic and prevailing meteorological conditions in Taiyuan. The higher levels of  $PM_{2.5}$ , OC and EC concentrations in cold seasons can be attributed to the heating period from November to March. In addition, the adverse meteorological conditions in cold seasons and the dustpan-shaped terrain of Taiyuan, with the mouth in the south and mountains in other directions, were not conducive to the advective diffusion of atmospheric pollutants [29]. Figure 3 shows seasonal wind roses with the intensity of atmospheric temperature inversion. It can be seen that the dominant wind usually came from west-north-east, of which only north wind with fast speed may dilute the pollutants under the condition of the dustpan terrain. Furthermore, the intensity of atmospheric temperature inversion was strong in cold seasons (Figure 3 and Table 1) and was liable to accumulate pollutants.



**Figure 3.** Seasonal wind roses with the intensity of atmospheric temperature inversion.

TCA represents the sum of EC and the sum of organic matter which is about  $1.6 \pm 0.2$  times that of OC in urban atmospheric aerosols [17,23,30]. In this study, TCA showed a similar seasonal change pattern to  $PM_{2.5}$ , OC, and EC (Table 1), while asynchronous variation was presented by the percentage of TCA in  $PM_{2.5}$ . TCA accounted for averaged 33.8%, 31.7%, 30.0% and 27.0% of  $PM_{2.5}$  mass in fall, summer, winter, and spring, respectively, with an annual mean TCA contribution percentage of 30.4% to  $PM_{2.5}$  mass. The present seasonal rank of TCA/ $PM_{2.5}$  was partly inconsistent with that in Taiyuan about a decade ago (winter > spring > fall > summer) [25]. The lowest TCA/ $PM_{2.5}$  percentage in the present work may be affected by frequent dust storms in spring, especially in dry spring seasons, such as April and May [23,31]. TCA contribution to  $PM_{2.5}$  in this study was in line with that in Chongqing [32], the Western Taiwan Strait Region [21] and Taiyuan a decade ago reported by He et al. (2015) [25], but much less than that in Beijing-Tianjin-Hebei integration [33].

OC and EC concentrations in this study were compared with those measured in other Chinese cities [23,32,34,35] in Table 2. In spring, the OC concentrations in Taiyuan were lower than in Shanghai and Beijing but higher than in Chongqing and Lvliang. However, the EC concentrations were lower than that in the remaining 4 cities. In summer, the concentrations of carbonaceous species in  $PM_{2.5}$  in Taiyuan were lower than in other Chinese cities. In the fall, the concentrations of carbonaceous species in  $PM_{2.5}$  in Taiyuan were higher than that measured in Lvliang but lower than that measured in Chongqing, Shanghai, and Beijing. In winter, the OC concentration was lower than that in Chongqing and Beijing but higher than that in Shanghai and Lvliang. However, the EC concentration was lower than that in Chongqing, Beijing, and Lvliang but higher than that in Shanghai. Therefore, several conclusions could be summarized: (1) In summer and fall, the concentrations of carbonaceous species in  $PM_{2.5}$  in Taiyuan were at a much lower level; (2) For the full year, the OC concentrations in Taiyuan were at a moderate level, and the EC concentrations of  $PM_{2.5}$  were at a lower level.

**Table 2.** Compare OC and EC concentrations in PM<sub>2.5</sub> in Taiyuan with those in other Chinese cities.

City	Period	Site Description	OC <sup>ζ</sup>	EC <sup>ζ</sup>	OC/EC	Reference
Taiyuan	Spring 2019	Urban commercial traffic	12.2	2.4	4.8	This study
	Summer 2018		5.1	1.5	3.5	
	Fall 2018		11.8	2.5	5.1	
	Winter		22.1	4.4	5.2	
	Annual		14.1	2.9	5.1	
Wanzhou, Chongqing	Spring 2013	Urban commercial-residential-cultural	9.9	4.9	2.1	[32]
	Summer 2013		7.9	4.2	1.6	
	Fall 2013		13.9	5.1	3.3	
	Winter 2013		62.8	20.5	3.0	
	Annual		23.6	8.7	2.5	
Zha-bei district, Shanghai	Spring, 2006	Urban traffic-residential-commercial	14.1	3.1	4.5	[23]
	Summer, 2006		7.2	1.9	3.4	
	Fall, 2005		21.8	3.9	5.3	
	Winter, 2005		16.7	2.3	6.8	
	Annual		14.7	2.8	5.0	
Beijing	Spring, April–May 2016, March 2017	Olympic Forest Park	28.4	9.9	2.9	[34]
	Summer 2016		26.4	12.6	2.1	
	Fall 2016		30.4	9.9	2.8	
	Winter, November 2016, January to February 2017		37.7	8.7	2.7	
	Annual		30.7	10.2	2.6	
Xiaoyi, Lvliang	Spring, 2019	Suburban	6.2	2.8	2.3	[35]
	Summer, 2019		5.5	2.6	2.1	
	Fall, 2018		3.7	1.9	2.2	
	Winter, 2018		15.0	10.7	1.5	
	Annual		7.5	4.4	2.0	

<sup>ζ</sup>: The unit for <sup>ζ</sup> is  $\mu\text{g m}^{-3}$ .

### 3.2. Relationship between OC and EC

The OC/EC ratio can indicate the source of carbonaceous matter and its transformation information [21,25]. The mean OC/EC ratios were 3.5, 5.1, 5.2 and 4.8 in the four seasons (Table 1), representing the dominant emissions from gasoline and diesel vehicles in summer while coal and even biomass combustion in other seasons [36,37]. The current OC/EC ratios were in the ranges of those in the 14 Chinese cities reported by Cao et al. (2007) but much higher than those in Taiyuan (1.5~2.5) a decade ago [25]. Seasonal correlations between OC and EC in PM<sub>2.5</sub> in Figure 4 may initially distinguish the main sources of carbon elements in the atmosphere and can partly reflect the relationship between both sources [38]. As shown, OC had high correlations with EC in spring ( $R^2 = 0.78$ ), fall ( $R^2 = 0.74$ ), and winter ( $R^2 = 0.61$ ), which indicates that they mainly came from similar sources. In spring, the low temperature and particulate, chemical activity, and weak secondary generation, and frequent dust storm pollution input could make the OC mass concentration more affected by the primary emission source.

The highest OC/EC in fall with a strong correlation should be ascribed to staged biomass burning after harvest [25,36,39], (He et al., 2015; Huang et al., 2018b; Schaap and Denier, 2007) and rural heating ahead of urban, which might increase OC/EC ratios because of combustion of raw coal [2]. The emission sources of OC and EC in winter were also homologous, mainly should be influenced by coal soot during the heating season. The lowest OC/EC value and poor correlation between them ( $R^2 = 0.2$ ) occurred in summer, which should be significantly influenced by photochemical quadratic sources and gaseous pollution emissions [40,41].

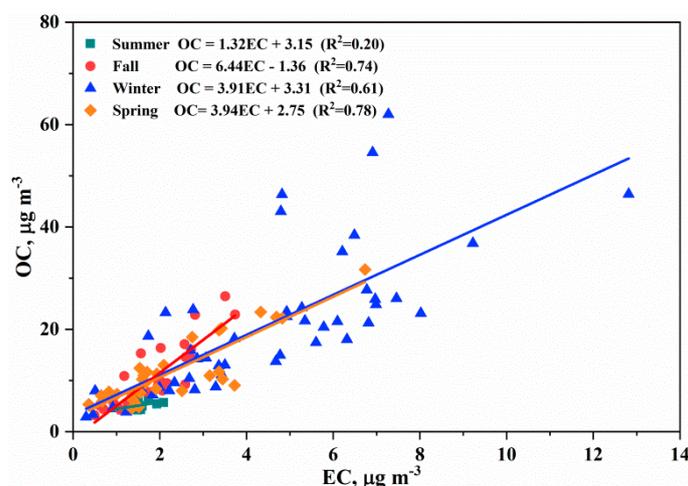


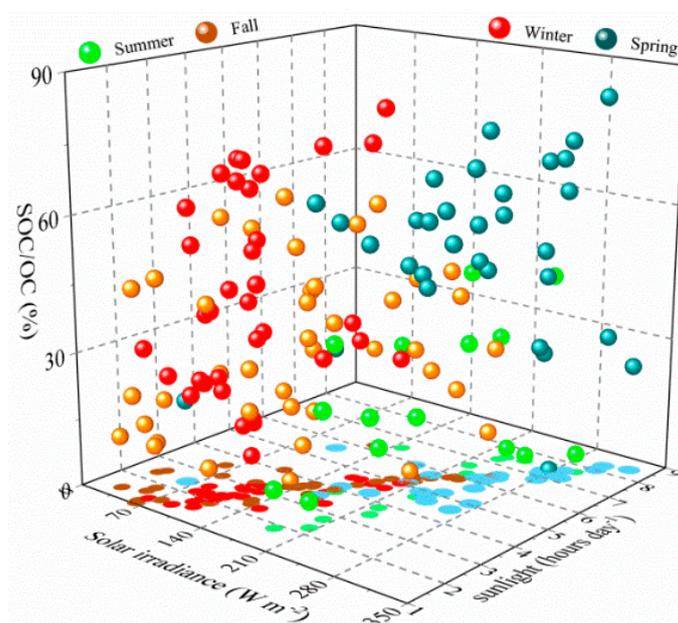
Figure 4. Seasonal correlations between OC and EC in  $PM_{2.5}$ .

### 3.3. Estimation of SOC and SOA Levels

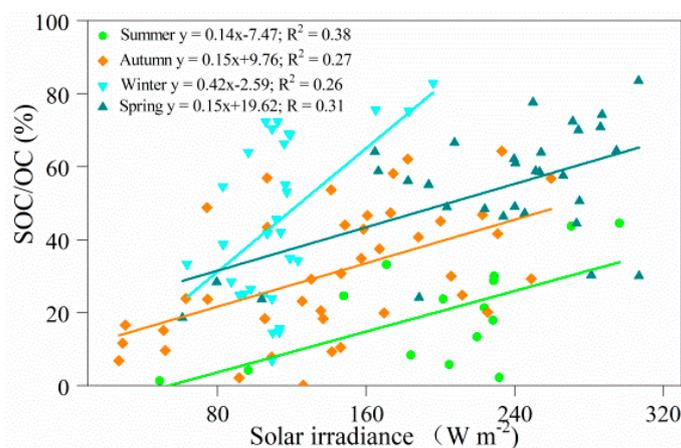
The presence of a second OC (SOC) is usually determined by using an OC/EC ratio higher than 2.0 [21,42,43]. However, the current OC/EC ratios were all higher than this value, and SOC was estimated via the current minimum value of OC/EC [44] (Table 1). The minimum values of OC/EC observed in the four seasons were 2.7, 3.5, 2.6 and 2.43. Thus, secondary organic aerosol (SOA) concentration can also be estimated at 1.6 times the SOC (Table 1).

As shown, Taiyuan's average annual SOC concentration was  $6.1 \pm 7.1 \mu\text{g m}^{-3}$ , representing 38.7% of the OC content. The mean SOC concentrations in the four seasons were  $1.1 \pm 0.9$ ,  $4.0 \pm 3.7$ ,  $10.4 \pm 10.6$  and  $6.4 \pm 3.9 \mu\text{g m}^{-3}$ , contributing about 20.2%, 31.1%, 43.3% and 52.0% to OC, respectively, which suggested SOC was an important component of OC in  $PM_{2.5}$  in Taiyuan. The present contribution percentage of SOC to OC was in line with that reported in the Western Taiwan Strait Region (23.8–39.4%) [21], Baotou (24.4–43.2%) [1], Beijing-Tianjin-Hebei integration (38–42%) [33]. Compared to 2009–2010 (22.9%, 42.8%, 56.2% and 25.9% in summer, fall, winter and spring, respectively) in Taiyuan [25], the percentage of SOC/OC in this work was almost equivalent to that in summer but much higher in spring and much lower in fall and winter, which again suggested the air atmospheric oxidation was enhanced in recent years. Notably, in contrast to the phenomena of higher SOC in summer than in other seasons [17,45], the highest concentration of SOC, as well as the SOC percentage of OC in this work, were not occurred in the expected summer [46] but in spring, which was similar to the SOC/OC percentage seasonal variation at an adjacent coal mining city in the North China Plain [27]. This phenomenon indicated enough secondary transformation occurred in the present sampling periods (especially in April and May) with conducive conditions to the photochemical reaction of semi-volatile and volatile organic compounds in Taiyuan. Figure 5 shows the dependence of the percentage of SOC in OC in the four seasons on solar radiation parameters. The increase in the average solar irradiance and daytime hours may be one of the key factors contributing to the increase in SOC/OC percentage. The average solar irradiance during daylight hours was  $220.3 \pm 36.9 \text{ W m}^{-2}$ ,  $153.9 \pm 64.4 \text{ W m}^{-2}$ ,  $116.0 \pm 35.1 \text{ W m}^{-2}$  and  $241.4 \pm 58.0 \text{ W m}^{-2}$  in the four seasons, respectively (Table 1). The present average light intensity during the current sampling period was greater in spring than in summer. Additionally, the higher SOC/OC percentages occurred in cold seasons, and the low positive associations between factors of SOC/OC percentages and solar irradiance (Figure 6) indicated that SOC production was limited by solar radiation and other causes [37]. Although winter has not provided favorable meteorological conditions for SOC production, the relatively higher SOC/OC percentage occurred in winter, which should be attributable to the emission of Volatile Organic Compounds (VOCs) increased for heating in Taiyuan and the stable atmospheric

condition and dustpan-terrain condition [25,46]. Additionally, higher rainfall in summer (Table 1) may limit the formation of SOC [39,47].



**Figure 5.** Seasonal SOC/OC percentage as a function of daily hours of sunlight and average solar irradiance during daylight hours.



**Figure 6.** Scatter plots and linear regression equations of SOC/OC percentage vs. solar irradiance.

From Table 1, the concentrations of SOA were  $1.7 \pm 1.4 \mu\text{g m}^{-3}$ ,  $6.4 \pm 5.9 \mu\text{g m}^{-3}$ ,  $16.7 \pm 16.9 \mu\text{g m}^{-3}$  and  $10.2 \pm 6.3 \mu\text{g m}^{-3}$ , with the corresponding contribution of 5.5%, 9.6%, 11.5% and 13.1% to  $\text{PM}_{2.5}$ , in summer, fall, winter and spring, respectively, which showed the same seasonal rank as SOC/OC. The annual mean percentage of SOA in  $\text{PM}_{2.5}$  was 10.5%, indicating that, as was the case a decade ago (12.2%) reported by He et al. (2015) [25], SOA accounted for a relatively small fraction of the  $\text{PM}_{2.5}$  mass in Taiyuan. However, the concentration of SOA in this study ( $9.8 \pm 11.3 \mu\text{g m}^{-3}$ ) was about one-third of that of a decade ago ( $27.5 \pm 30.3 \mu\text{g m}^{-3}$ ) [25].

#### 4. Conclusions

Although significant air pollutant reductions have been achieved during this decade,  $\text{PM}_{2.5}$  pollution was still very serious in cold seasons. Only the summer  $\text{PM}_{2.5}$  concentration was within the first order ambient air quality standard daily limit, while that in winter,

spring and fall were about 3.4, 1.9 and 2.3 times that of the limit. The mass of OC and EC in PM<sub>2.5</sub> presented synchronous variation with PM<sub>2.5</sub> with the seasonal mean concentrations of  $5.1 \pm 0.9 \mu\text{g m}^{-3}$ ,  $11.8 \pm 6.4 \mu\text{g m}^{-3}$ ,  $22.1 \pm 14.9 \mu\text{g m}^{-3}$ , and  $12.2 \pm 6.7 \mu\text{g m}^{-3}$ , for OC and  $1.5 \pm 0.3 \mu\text{g m}^{-3}$ ,  $2.5 \pm 1.6 \mu\text{g m}^{-3}$ ,  $4.4 \pm 2.8 \mu\text{g m}^{-3}$ , and  $2.4 \pm 1.5 \mu\text{g m}^{-3}$  for EC in summer, fall, winter and spring, respectively. The mean OC/EC ratios of 3.5, 5.1, 5.2 and 4.8 in the four seasons and well correlations between OC and EC indicated that coal and biomass combustion was the dominant emissions in cold seasons, aggravated by the adverse meteorological conditions and the dustpan-shaped terrain. The present higher SOC concentration and SOC/OC percentage suggested that the air atmospheric oxidation was enhanced in recent years, and the production of SOC was influenced by solar radiation, heating in cold seasons, stable atmospheric condition, rainfall, etc. Overall, carbon aerosol pollution has reduced significantly in the last decade, but there is still a long way to go for local air pollution control.

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