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Research on Organic Carbon and Elemental Carbon Distribution Characteristics and Their Influence on Fine Particulate Matter ($PM_{2.5}$) in Changchun City

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Abstract: In order to understand the distribution characteristics of organic carbon (OC) and elemental carbon (EC) in $PM_{2.5}$ in Changchun; China; $PM_{2.5}$ samples were collected from April 2017 to December 2017 using the KC-120H particulate matter sampler; and the NIOSH 5040 method was used for determination. The results showed that the average concentration of $PM_{2.5}$ in Changchun was 45.92 μg/m^3 (45.92 ± 50.17), and the annual average concentrations of OC and EC ranged from 15.69 to 24.32 μg/m^3 and from 1.38 to 2.33 μg/m^3; respectively. The annual OC/EC ratio range was 8.08–15.44; with an average of 11.70. OC and EC concentrations in spring were the lowest; whereas higher levels of both OC and EC were found in winter. Significant correlations between OC and EC were found in the non-heating period; indicating that there was a consistent or similar source; whereas OC was non-significantly correlated with EC in the heating period; suggesting that contributions of OC were from unrelated combustion sources.

Keywords: $PM_{2.5}$; organic carbon; elemental carbon; secondary organic carbon

1. Introduction

Carbonaceous material is one important component of atmospheric particulate matter. Organic carbon (OC) includes primary organic carbon (POC) and secondary organic carbon (SOC); these are mainly created by the man-made process of fossil fuel combustion, with relatively little contribution from natural sources [1]. The structure of elemental carbon (EC) is similar to graphite, which is emitted directly into the atmosphere predominantly during incomplete combustion emissions, such as motor vehicle exhaust, fuel burning, and biomass burning [2]. Studies have shown that OC can cause or exacerbate many diseases, such as cardiovascular diseases, respiratory diseases, cancer, etc. [3]. According to the national environmental analysis of the People's Republic of China, released by the Asian Development Bank and Tsinghua University, air pollution in China has caused economic losses each year, based on the estimated cost of illness as equivalent to 1.2 percent of gross domestic product, and on willingness-to-pay estimates as high as 3.8% [4]. Elemental carbon can be measured as an index of fossil fuel combustion and transport [5]. It could lead to reduced visibility, global warming, and reduced crop yields [6]. Many foreign countries have conducted observational studies of OC and EC in $PM_{2.5}$. Viana et al. [7] analyzed the changes of OC and EC in three major European cities and inferred that secondary aerosols had a great influence on the composition of $PM_{2.5}$ from OC/EC. Srinivas et al. [8] analyzed the mass concentration of $PM_{2.5}$, mineral dust, and OC and EC from 10 November to 10 March in India, and the ratio of OC/EC was about 7.0 ± 2.2. Ham et al. [9] analyzed the pollution characteristics of EC and OC in atmospheric $PM_{2.5}$ in Seoul, Korea, from March to April 2016. The average concentrations of OC and EC during the study period were 4.4 ± 2.0 μg/m^3

and 1.4 ± 0.6 μg/m^3, respectively. The ratio was 3.4 ± 1.0 and the average concentration of $PM_{2.5}$ was 39.7 ± 19.8 μg/m^3. It was found that Seoul's air quality in spring was mainly affected by local pollutants and man-made pollutants. The range of domestic research is mainly concentrated in some large cities, such as the Pearl River Delta, the Yangtze River Delta, Beijing-Tianjin-Hebei and other economically developed cities; other areas have begun research in recent years. Jiajia Gao et al. [10] analyzed the sources and distribution characteristics of $PM_{2.5}$ in the Beijing-Tianjin-Hebei region and showed that OC and EC concentrations were lower in spring and summer than in autumn and winter, and vehicle exhaust emissions were the main source of OC and EC. This paper focuses on the carbon component variations of $PM_{2.5}$ in Changchun to explore the relationship between possible sources of carbon components and atmospheric components. Another goal is to provide a scientific basis for Changchun to carry out various air pollution prevention and control plans.

2. Methods

2.1. Sampling Sites

There are 10 automatic air monitoring stations and one super air monitoring station (SAMS) in Changchun, as shown in Figure 1: Daishan Park (DP), High-Tech Zone Management Committee (HZMC), Institute of Posts and Telecommunications (IPT), Children's Park (CP), Bus Factory Hospital (BFH), Labour Park (LP), Economic Development Zone Environment Sanitary Administration (EESA), Jingyue Park (JYP), Junzilan Park (JZP), Shuaiwanzi (SWZ), and one SAMS. Shuaiwanzi station is located in the Shuangyang district of Changchun; it is used as a controlling site to represent the background value of air pollution in Changchun. The other nine monitoring stations were set up according to the layout and air quality functional areas in Changchun, as shown in Table 1.

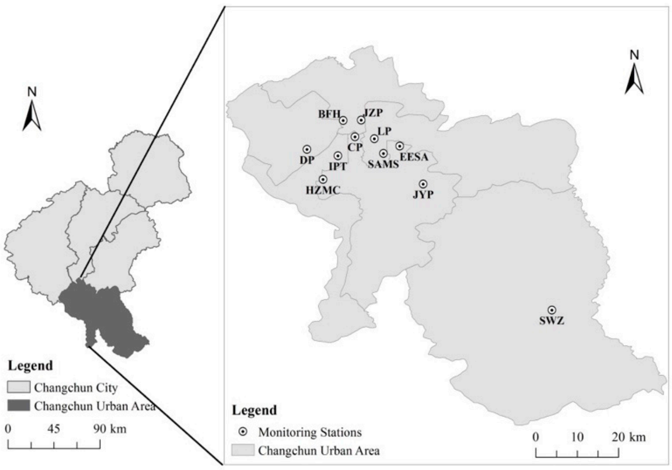

Figure 1. Air monitoring stations of Changchun.

Table 1. Sampling stations in Changchun.

Name	Environmental Air Quality Functional Area	Major Source of Pollution (Area)	Wind Direction
CP	II	Urban and rural	Upwind
BFH	II	Industrial discharge	Upwind
IPT	II	Densely populated	Upwind
LP	II	Densely populated	Downwind
HZMC	II	Urban and rural	Upwind
DP	II	Industrial discharge	Upwind
JYP	I	Natural reserve	Side wind
EESA	II	Densely populated	Downwind
JZP	II	Vehicle emission	Downwind
SWZ	I	Background	Side wind
SAMS	II	Normally populated	Center

2.2. Sample Collection

Sample collection was conducted from 15 April 2017 to 14 June 2017, 15 June 2017 to 24 August 2017, 25 August 2017 to 24 October 2017, and 25 October 2017 to 31 December 2017. Sampling was carried out at one-day intervals; 15 samples were collected every season, representing the four seasons of spring, summer, autumn, and winter of 2017. At each sample site, a KC-120H particle sampler manufactured by Qingdao Laoshan Electronic Equipment Co., Ltd. (China) was set. This aspect of the daily monitoring data was used to analyze the correlations between the OC and EC concentrations in the heating and non-heating periods of the nine sampling sites. The rest of the data analyzed in this paper are the hourly monitoring data from the super air monitoring station (SAMS). All filter membranes were scrutinized and baked in the oven for 24 h before sampling. Each sampling process lasted for 22 h, and the quartz filter membrane was replaced every 24 h. During the sampling procedure, we added some blank filter samples at a proportion of 10% for quality control, and we also added some standard samples during the sample analysis procedure for the uncertainty measurement. After sampling, the filter was folded in half (the sample of the collected particles was on the inside of the fold) and placed in a sealed bag. The sampling time was marked, and it was placed at a constant temperature (22 °C ± 1 °C) and humidity (40% ± 5%) in a room at constant temperature for measurement. The quartz filter membrane was accurately weighed before and after sampling by an electronic balance with an accuracy of 0.1 mg (Sartorius, WZA215-LC 210 g/0.01 mg, Gottingen, Germany), then data were recorded.

2.3. Sample Analysis

NIOSH method 5040 was selected for OC and EC analysis of 540 receptor samples. For 14 receptor samples, equipment failure occurred in the sampling process and were therefore not analyzed. This study referred to the national standard, "Determination of elemental carbon and organic carbon of atmospheric aerosols—thermal/optical analysis method (QX/T 70-2007)", for sample analysis. Before analysis, the instrument was calibrated with a sucrose solution at a standard concentration of 2.103 μgC/μl. Then, for the blank test, six consecutive measured and theoretical values were compared to the test results. Where the calibration factor of ±5% adjusted exceeded a predetermined instrument, the sample would be tested after calibration of the instrument. A blank experiment was performed for each of the 20 samples analyzed, and the standard curve was verified daily using the standard curve intermediate point standard. After the problem was found, the standard curve was re-made to ensure the accuracy of the results.

In this study, hourly monitoring data of OC and EC were obtained from the SAMS of Changchun.

2.4. Data Analysis

The experimental data obtained were analyzed by Microsoft Excel and the functional modules of correlation analysis were obtained using IBM SPSS 19.0 (Chicago, IL, USA).

3. Results and Discussion

3.1. Annual Variations of OC and EC Concentration

According to the Changchun Statistical Yearbook of 2017 [11], the proportion of coal energy structure consumption in Changchun City is more than 62%, which means that the big OC and EC emission sources are industry usage and central heating in winter. Table 2 summarizes the average concentration levels of OC and EC in other urban areas, including Shanghai [12] and Nanjing [13], which are used for comparison with the results of this study. The results showed that the concentration of OC in Changchun was at the national medium level, whereas EC was lower than in other regions. The OC and EC concentrations in Sanya [14] were 3.3 $\mu g/m^3$ and 1.1 $\mu g/m^3$, respectively. OC and EC concentrations were only 15.2% and 59.5%, respectively, of those in Changchun, indicating that OC and EC pollution in Sanya $PM_{2.5}$ was relatively light. Conversely, OC and EC in Xi'an were higher than those in Changchun. This is because Xi'an is an inland city, vehicles are continuous sources of pollution, and there are agricultural activities such as autumn straw burning, which increase the carbon content in the atmosphere. Also, during winter, Xi'an has heating, air circulation is poor, and contaminants are not easily spread. Taiyuan has a high concentration of OC and EC, which is related to its primary industry, agriculture. This is related not only to straw burning but also to the city's vigorous industrial development. The cities with high OC concentration are all northern cities, and are associated with large amounts of OC produced by coal combustion.

Table 2. Average concentration of organic carbon (OC) and elemental carbon (EC) in $PM_{2.5}$ in Changchun and other regions.

Regions	OC (μg/m³)	EC (μg/m³)
Beijing [15]	19.7	2.3
Xi'an [16]	40.6	8.3
Guangzhou [17]	17.5	4.1
Huangshi [18]	11.89	2.28
Shanghai	8.6	2.4
Jinan [19]	16.98	5.81
Dalian [20]	31.85	6.38
Nanjing	11.8	2.9
Sanya	3.3	1.1
Tangshan [21]	28	11
Ningbo [22]	9	2.9
Wuhan [23]	19.24	2.9
Hangzhou [24]	13.7	15.9
Linan [25]	10.1	2.4
Xiamen [26]	17.7	30
Taiyuan [27]	65.2	23.5
Shenzhen [28]	8.3	4.7
Chongqing [29]	15.2	4.2
Changchun	21.7	1.85

Figure 2 presents the concentration trends of OC and EC as basically the same. After 5 am in $PM_{2.5}$, OC and EC concentrations continued to rise; after 9 am, the concentration of both first decreased and then increased, and the lowest concentration appeared at 3 pm. This could be due to the respective sampling sites in the urban center of the region: Human activities are more frequent after 5 am. Morning heating during the heating period and kitchen lampblack have an effect on its concentration. With an increasing vehicle population every year, vehicle exhaust has become a non-negligible source of air pollution. According to the research group's meteorological statistics, in Changchun city, the annual average temperature increased from 0 to 12 degrees Celsius during the hours from 9 am to 12 pm. A gradual increase means that convection is generated, which is conducive to the spread of pollutants. Moreover, the intensity of sunlight increases, photochemical reactions become active, and the SOC that may be generated supplements the concentration of OC.

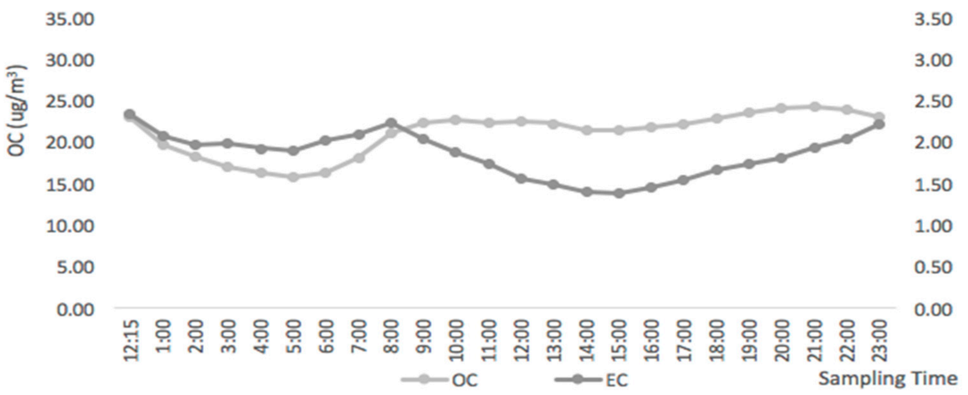

Figure 2. Annual variations of OC and EC concentration.

3.2. Seasonal Variations of OC and EC Concentration

It can be seen from Figure 3a that the average concentration of OC and EC in spring varies. Unlike the annual trend, EC concentration did not increase significantly after 5 am. This result could be ascribed to the following: On the one hand, according to Shan et al [30], the surface temperature in spring increased, and the Mongolian high-pressure system weakened. At this time, the low-pressure system invaded from the Baikal area, forming a northeasterly low pressure and frequent transit. A strong southwesterly airflow often appeared in the low-pressure front, and a violent northwesterly airflow in the back. Furthermore, the maximum wind speed was up to 30 m/s, thus contributing to diffusion in the atmosphere. On the other hand, after 9 pm solar radiation disappeared, the temperature dropped, and the atmosphere was stable. These factors were not conducive to the spread of pollutants that led to the increase in EC concentration. At the same time, weak light and low temperature factors also limited the generation of SOC, and this was one of the factors that caused the OC concentration to decrease. Figure 3b shows that summer OC and spring EC trends were substantially the same, with no obvious peak or underestimation. This indicates that, although artificial activities weakened after noon, the intensity of sunlight during a summer afternoon is very high and temperature rises. This is the most intense period of photochemical reaction in one day, and it promotes a large amount of SOC generation.

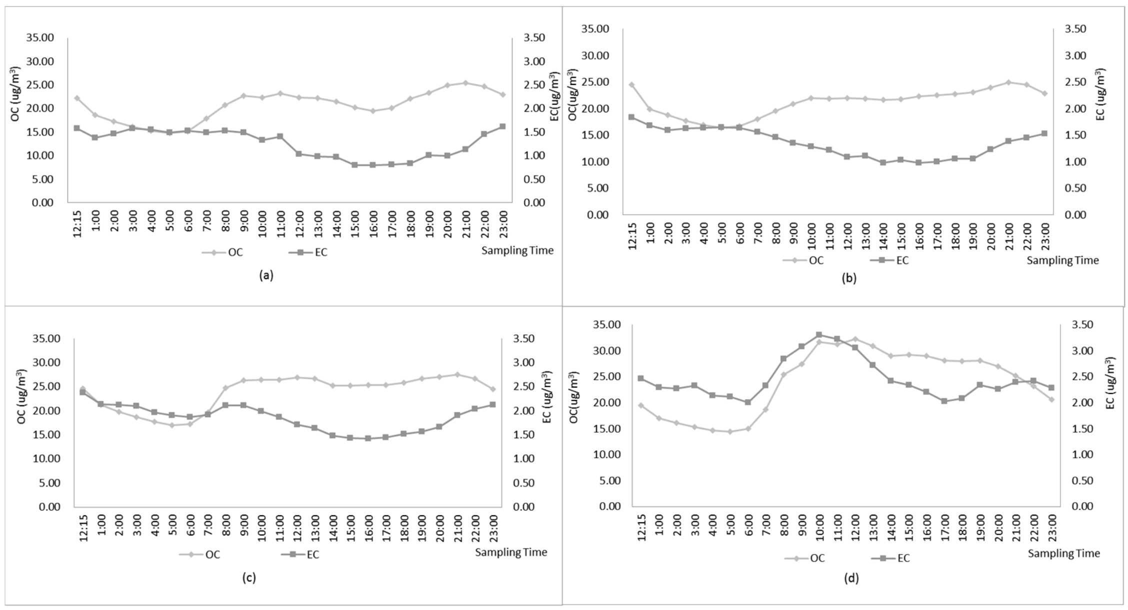

Figure 3. Seasonal variations of OC and EC concentration.

Autumn OC and EC concentration trends were substantially the same as the annual average. For EC, the concentration gradually increased from 6 am to 9 am, and reached a high level at 4 pm due to human activities. During autumn, Changchun often has low air pressure and wind speed. These meteorological conditions are not conducive to the diffusion of pollutants in the air, and the combustion of biomass (such as fallen leaves and straw) also makes the concentration of OC and EC relatively large.

According to Fang et al., [31], the main sources of OC in Changchun $PM_{2.5}$ are motor vehicle exhaust, secondary particles, biomass burning, and urban dust. The main sources of EC are motor vehicle exhaust and coal-fired dust. Figure 3d shows that the OC and EC concentrations in Changchun had obvious seasonal characteristics. The higher mean concentrations of EC and OC in winter were likely related to the influence of emissions from residential heating (in addition to traffic sources) and, on the other hand, to the unfavorable meteorological conditions leading to greater dispersion of pollutants in the atmosphere during this season.

Table 3 shows that the concentration in winter was higher than in other seasons, and the concentration in spring was lower than in other seasons. According to the previous meteorological statistics, in Changchun City winter sunlight and turbulence are weak; furthermore, heating in Changchun also increases pollutant emissions. In autumn, one month belongs to the heating period, and combustion of biomass (such as deciduous leaves and straw) in this season also affects the concentration of OC and EC. In spring and summer, the light duration is long, the ground receives more energy, and vertical diffusion of turbulence is stronger, which is conducive to the diffusion of pollutants, so the concentration of pollutants in spring and summer is lower. Compared to summer, wind speed in spring is greater and thus increases the spread of pollutants.

Table 3. OC and EC contents in different seasons in Changchun.

Period	OC (μg/m³)				EC (μg/m³)			
	Max	Min	Mean	SD	Max	Min	Mean	SD
Spring	25.45	14.72	20.62	20.62 ± 3.19	1.62	0.79	1.26	1.26 ± 0.29
Summer	24.91	16.41	21.15	21.15 ± 2.56	1.84	0.98	1.35	1.35 ± 0.27
Autumn	27.48	16.99	23.85	23.85 ± 3.52	2.37	1.42	1.85	1.85 ± 0.27
Winter	32.23	14.38	24.03	24.03 ± 6.21	3.3	2.07	2.45	2.45 ± 0.38

Notes: SD is defined as the standard deviation.

3.3. *Heating and Non-Heating Period Variations of OC and EC Concentration*

The heating period calculated in this paper includes two parts, 1 January 2017 to 14 April 2017 and 25 October 2017 to 31 December 2017; the non-heating period began on 15 April and ended on 24 October 2017. According to Figure 4a, the Changchun area heating period temperature is lower than in other regions, light intensity is weak, and the weather is often calm, with aggregation of particles in the air. This results in an increase of contaminant concentration within a limited space, but also promotes the formation of SOC.

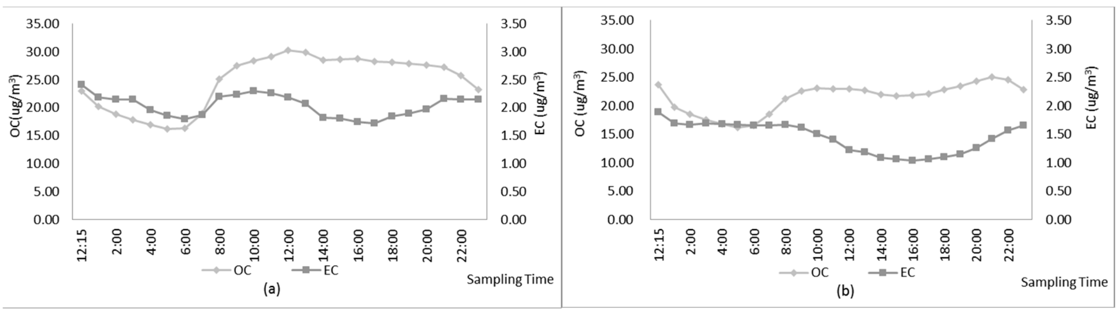

Figure 4. Heating and non-heating period variations of EC and OC concentration.

Figure 4b shows the concentration trend during the non-heating period. As opposed to the trend of the whole year, EC concentration did not increase significantly after 5 am, which was due to the high temperature and good air circulation in the non-heating period of Changchun, which promoted atmosphere diffusion. Afternoon OC and EC concentrations showed a significant downturn; essentially, afternoon solar radiation disappears gradually, temperature drops, and the atmosphere is stable against contaminant diffusion. The concentration of EC gradually increased after 4 pm, which means that the late peak gradually begun in the evening; pollutants produced in the evening also accumulate as human activities increase. OC and EC concentrations during the heating period of $PM_{2.5}$ in Changchun were higher than those during the non-heating period, which is made clear in Table 4. Compared with the non-heating period, OC was 0.15 times higher and EC concentration was 0.42 times higher during the heating period. This was caused by the large amount of coal burning during the heating period.

Table 4. Average OC and EC concentration in $PM_{2.5}$ during heating and non-heating periods.

Period	OC (μg/m^3)	EC (μg/m^3)
Heating period	24.68	2.04
Non-heating period	21.39	1.44

3.4. Relationship between Organic Carbon and Elemental Carbon

The OC/EC ratio is an important guide to determine the conversion characteristics of pollutant emissions and carbonaceous particles. Studies have shown that by studying the correlation between OC and EC, the source of carbonaceous aerosol can be distinguished [32]. If the correlation between OC and EC is significant, then the pollution sources are roughly the same. Therefore, the correlation between OC and EC can qualitatively analyze the source of carbon aerosol. Figure 5 shows the correlation analysis of OC and EC in the heating period and the non-heating period. Studies have shown that the correlation coefficient between OC and EC in the heating period of Changchun is very low (r = 0.042), indicating that the source of OC and EC is relatively complex. During the heating period, the temperature fluctuates greatly, and the concentration of pollutants changes greatly due to snow and wind. Changchun is located in the north and has a heating period of 5 months throughout every year. Coal burning is the main source of OC and EC, far exceeding pollution caused by vehicle exhaust and biomass combustion. Correlation analysis was based on data from nine sample sites, and the correlation coefficient of the non-heating r = −0.435. At the same time, Excel and SPSS were used for correlation analysis. For this study, we set a significance level α = 0.05 and a significant correlation was shown, indicating that, for the non-heating period, Changchun OC and EC mostly came from the same source. OC and EC mainly come from vehicle exhaust emissions, carbon burning for heating, and biomass burning. For the non-heating period, the coal combustion contribution rate is small, mainly affected by diesel and gasoline vehicle exhaust emissions.

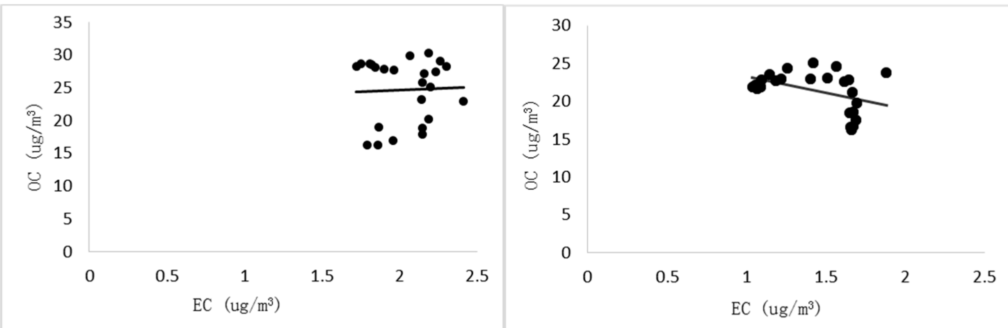

Figure 5. Correlation between OC and EC during heating and non-heating periods.

4. Conclusion

Based on the sampling data of the receivers in Changchun and the SAMS, the concentrations of OC and EC in the heating period were 16.27–30.27 μg/m^3 and 1.72–2.41 μg/m^3, respectively, and those in the non-heating period were 16.18–25.02 μg/m^3 and 1.04–1.88 μg/m^3, respectively. Similar trends were generally observed during the year. The OC concentration over the whole year increased continuously after 5 am and reached a very high level at 4 pm; the EC concentration generally increased from 4 pm. In spring, the OC and EC concentrations were the lowest, and the winter OC and EC concentrations were higher than the other three seasons. The correlation between OC and EC in the heating period was weak, indicating that the source of OC was not only the burning of fossil fuels. On the contrary, the correlation between OC and EC in the non-heating period was relatively good, indicating that OC and EC in the non-heating period had the same or similar sources. The OC/EC ratio was used to estimate the amount of secondary organic aerosols, and a relatively high value was found in the non-heating period. Thus, the major factors influencing the air quality in Changchun were vehicle exhaust emissions, coal burning, and biomass burning. Therefore, stricter countermeasures should be taken, such as controlling the growth of the vehicle population, reducing vehicle pollution, adopting natural gas instead of coal combustion in urban areas, etc, doing so will make an important contribution to improving the air quality in Changchun.

Author Contributions: J.W. was working for project administration. A.Y. was working for formal analysis and application of software. C.F. was working for date curation and methodology. Teacher C.F. and J.W. helped analysis of raw data and review of origin draft. Also, teachers worked as a supervisor and director of this study. L.Y. helped to the main author to collect the raw data of monitoring stations.

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Conflicts of Interest: The authors declare no conflict of interest.

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