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RADIOCARBON CHARACTERIZATION STUDY OF ATMOSPHERIC PM2.5 IN BEIJING DURING THE 2014 APEC SUMMIT

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ABSTRACT. Radiocarbon (^{14}C) has become a unique and powerful tracer in source apportionment of atmospheric carbonaceous particles. In this study, the Asia Pacific Economic Cooperation summit (APEC) held in Beijing in 2014 was used as a demonstration to research the source apportionment of atmosphere PM2.5. We used a 200 kV single stage accelerator mass spectrometer recently completed at China Institute of Atomic Energy (CIAE). The PM2.5 samples related to above case were collected, and the characteristics of radiocarbon in organic carbon (OC) and elemental carbon (EC) in samples were analyzed using the AMS. The results show that the Before-APEC pollution emission mode is different from the During-APEC and After-APEC pollution emission modes. For Before-APEC, During-APEC and After-APEC, the average values of fossil carbon fraction of OC are 0.463, 0.431 and 0.615, respectively, and those of EC are 0.644, 0.561 and 0.687. The fossil source contributions of traffic activities using fossil fuels to OC and EC are 15.8% and 21.9%, respectively. The fossil source contributions of industrial activities to OC and EC are 38.0% and 8.2%, respectively. It is about 7–10 days that is needed to take to regenerate the PM2.5 pollution caused by human activities.

KEYWORDS: accelerator mass spectrometer, APEC, organic carbon and elemental carbon, PM2.5, radiocarbon.

INTRODUCTION

With the rapid development of industrialization and urbanization in the absence of effective measures for air pollution abatement, China has suffered from a serious air pollution problem over the past decades (Chan and Yao 2008). In order to improve air quality, the Chinese government has updated the Ambient Air Quality Standards and issued the 12th Five-Year Plan on Air Pollution Prevention and Control in Key Regions in 2012 (Central 2012). Subsequently, the State Council of China had released the Action Plan for Air Pollution Prevention and Control in 2013 (Central 2013). Nevertheless, air pollution is still getting worse in China, especially in northern China where haze episodes have happened frequently in recent years. Carbon-containing aerosols are important components of atmospheric particulate matter. Carbonaceous aerosols are now of worldwide concern for their direct and indirect impacts on global and regional climate change, visibility degradation, air quality, and public health (Mahowald 2011; Anenberg et al. 2012; Chung et al. 2012). Radiocarbon (^{14}C) has become a unique and powerful tracer in source apportionment of atmospheric carbonaceous particles, as ^{14}C measurements enable a distinction between non-fossil carbon and fossil carbon. This directly allows a differentiation between biogenic (non-fossil carbon) and anthropogenic (fossil carbon) origin, which is based on the simple model that biogenic sources emit aerosols on the present $^{14}C/^{12}C$ level and anthropogenic particulate matter only derives from fossil fuel extinct in ^{14}C. Especially in the study of radiocarbon characteristics in organic carbon (OC) and elemental carbon (EC) more valuable information can be obtained. EC and OC are good tracers for primary anthropogenic emissions that are formed by incomplete combustion (Bond et al. 2004, 2013). EC tends to be formed in larger amounts than OC when fossil fuels, such as oil, coal and diesel, are incompletely combusted. Conversely, when biomass fuels are incompletely combusted, OC is emitted in larger amounts than EC. Thus, the OC/EC ratio has become an important diagnostic ratio to estimate primary and

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secondary carbon pollution as well as the source type (biogenic or fossil). As a result of secondary organic aerosol (SOA) formation, ambient concentrations of OC, as well as the OC/EC ratios, increase. Therefore, OC/EC ratios that exceed the expected primary emission ratio indicate SOA formation (Cabada et al. 2004). It is a critical job to quantify these sources for the development of effective and efficient air pollution control measures, especially in areas with rapid economic growth and high population density where large amounts of those species are released into the air.

With Beijing hosting the 2014 APEC summit, Beijing and its neighboring regions took tough actions to reduce the emissions of air pollutants from industry, traffic, and construction sites. As a result, the emission reduction measures decreased the emissions of SO_2, NOx, VOCs, PM10, and PM2.5 by 39.2 %, 49.6 %, 33.6 %, 66.6 % and 61.6 % in Beijing, respectively (Beijing 2014), and as the air quality got better, the sky in Beijing maintained a long blue state, which was called APEC Blue. The event demonstrated that haze can be eliminated in the short term by a human-perturbing campaign although a permanent solution for air pollution is still a tremendous challenge, which provided opportunities to investigate the effect of pollutant emissions on the air quality of Beijing and the surrounding regions. Especially, the industrial activities were stopped before the meeting, and the traffic activities were strictly controlled during the meeting, however, both the traffic activities and industrial activities were resumed after the meeting, which provides an opportunity for researching the effect of human activities on the PM2.5.

In order to meet the application requirement of ^{14}C in the field of environment such as PM2.5 monitoring, a 200 kV single stage accelerator mass spectrometer has been completed at CIAE. Based on this device, the characteristics of radiocarbon in organic carbon and elemental carbon in atmosphere during the APEC summit in Beijing in 2014 were analyzed. A series of PM2.5 samples from Chaoyang District in Beijing during the APEC summit were studied, the contents of OC and EC, including their respective radioactive carbon content were mainly analyzed. The objective of this study was to understand the carbon characterization of PM2.5 under the local pollution controls in Beijing before, during and after the 2014 APEC summit. In addition, this work may help researchers and local governments to better understand the mechanisms of PM2.5 pollution.

METHODS

Sample Location

Aerosol fine particulate matter (PM2.5) samples were collected from October 8 to November 24, 2014, in Chaoyang District, Beijing, China (116°35′E, 40°10′N), the sampler was installed at a site around 15 m height above the ground. Since the radiocarbon characteristics in the PM2.5 sample were analyzed, quartz filters (QMA 1851-865 100/PK, Whatman, UK) were used to collect the sample. All quartz filters were preheated at 450°C for 6 hr before sampling. The filters were wrapped in aluminum foil and sealed in polyethylene bags before and after the sample collection and the collected samples were stored in a freezer at –18°C and wrapped in aluminum foil to avoid light exposure. A high-volume sampler (Thermo-Andersen, Atlanta, GA, USA) was used to collect the aerosol samples on quartz filters (10×10 cm) at flow rate of 1.05 m^3/min. In total, we collected 48 samples during this period, and each sample was collected for 12 hr. The samples used for radiocarbon analysis

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were collected from October 28–31, November 8–11, and November 17–20, 2014, which represents periods before, during, and after APEC, respectively.

Sample Procedure

The contents of total carbon (TC), organic carbon (OC) and elemental carbon (EC) in the PM2.5 sample collected in a quartz film were measured using an OC/EC analyzer (Cavalli et al. 2010). We have explored a new OC/EC separation method by referring to the principle of Interagency Monitoring of Protected Visual Environment (IMPROVE) thermal/optical reflectance (TOR) protocol and combining with our laboratory ^{14}C sample preparation method. The OC and EC carbonaceous fractions were separately extracted by the method, and details are as follows. The flow chart of OC/EC in the PM2.5 sample preparation and device diagram are shown in Figure 1 (a) and (b), respectively. A proper amount of PM2.5 sample was put into a vacuum quartz tube filled with copper oxide, small copper beads and silver wire. The PM2.5 sample in the vacuum tube was heated to 120ºC, 250ºC, 375ºC, 450ºC, and 520ºC for 10 min, respectively. As for pretreatment sample size, the lower limit is estimated according to the EC content that calculated by the test results of the OC/EC analyzer should not be less than 500 μg, and the upper limit is calculated with reference to the quartz tube volume and the pressure in the quartz tube which must be about 10^3 Pa after heating during this process. In addition, it was necessary to shake the sample so as to release the organic carbon completely to ensure complete combustion. The gases generated from the above process were imported into the ^{14}C sample graphitization system, and then carbon dioxide in the gases was purified, collected and reduced to graphite which can be used to study the radiocarbon characterization of OC. The remaining solid powders from the above process were placed in a quartz tube filled with copper oxide again and heated at 850ºC for 2 hr under vacuum to collect the EC fraction, similarly to graphitization process of OC, the graphite was produced to study the radiocarbon characterization of EC.

^{14}C Analysis

^{14}C measurements were performed at CIAE compact accelerator mass spectrometry (AMS) system, which was based on a 200 kV single-stage accelerator mass spectrometer (SSAMS) (He et al. 2019). Details of the target production, measurement parameters, and data evaluation for sub-milligram samples were reported (Pang et al. 2017). Measured f_C values were corrected for an over-all blank, which includes not only the contributions of the filters, but also the contributions of storage and chemical pre-treatment. In all cases, the EC concentrations on these blank filters were below the detection limit. For OC, the blank-correction was approximately 4.8 %. Therefore, the uncertainty of the calibration is negligible for the combined measurement uncertainty. The radiocarbon data were expressed as the fraction of modern carbon (f_C). The f_C was converted into the fraction of biogenic carbon (f_{bio}) with a correction factor of 1.052 ± 0.013 based on the long-term time series of $^{14}CO_2$ at the background station (Levin and Kromer 2004; Levin et al. 2013); thus, the f_{bio} was calculated as $f_{bio} = f_C / 1.052$. The f_{bio} can range from 0 (pure fossil carbon) to 1 (pure modern carbon) and directly reflects the relative fossil and biogenic contribution to carbon, so the fraction of fossil carbon (f_f): $f_f = 1 - f_{bio}$. The uncertainty of f_{bio} for PM2.5 samples was obtained from an error propagation, and included uncertainties in the carbon concentration, variability of the reference f_C, and measurement uncertainty of f_C.

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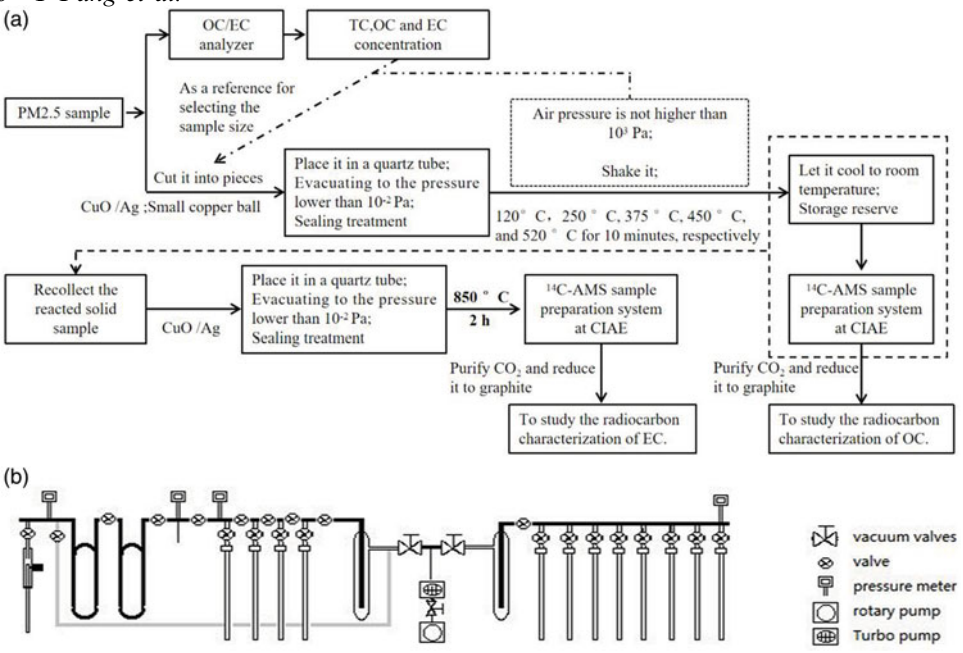

Figure 1 (a) the flow chart of OC/EC sample preparation and device diagram and (b) ^{14}C-AMS sample preparation system at CIAE.

RESULTS AND DISCUSSION

The carbon content of the sample must be known for sample preparation, which is provided by the OC/EC analyzer, and this information is crucial for atmospheric PM2.5 source apportionment based on the radiocarbon characteristics, as shown in Figure 2 and Table 1. Results from 12 samples were obtained for this study. The total carbon (TC) concentrations for Before-APEC, During-APEC, and After-APEC are 31.41–41.08 μg/m^3 (average: 37.73 μg/m^3), 7.53–32.31 μg/m^3 (average: 25.59 μg/m^3), and 25.75–106.68 μg/m^3 (average: 74.80 μg/m^3), respectively. Similar to that observed in Shanghai during winter (Qiao et al. 2015), but higher than that in the Pearl River Delta (PRD) region in southern China (Kuang et al. 2015). The OC/EC ratios in the Before-APEC, During-APEC, and After-APEC periods are 2.51–2.95 (average: 2.70), 2.89–5.17 (average: 3.62), and 3.23–3.66 (average: 3.44), respectively. The TC concentration for After-APEC is significantly higher than the previous two periods, and reaches up to 106.68 μg/m^3, while the TC concentration for During-APEC is slightly lower than that for Before-APEC and reaches the lowest value of 7.53 μg/m^3 on November 11, 2014. Comparing the TC/OC/EC concentrations of samples taken on November 11, 2014 and November 19, 2014 respectively, we found that the TC/OC/EC concentrations increased from the lowest to the highest through 8 days. Obviously, the preparatory work for the APEC meeting has indeed played a significant role in improving air quality.

The relationship between OC and EC concentrations can indirectly reflect the origin of a carbonaceous aerosol. If major fractions of OC and EC are emitted from a primary source, a correlation between OC and EC mass concentrations should exist, because the emission rates of OC and EC should be proportional to each other (Na et al. 2004). The correlations of OC and EC in all samples are described in Figure 3. The meteorological conditions

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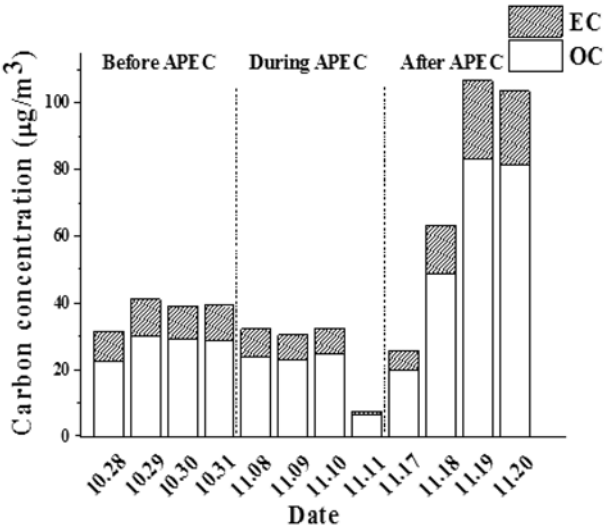

Figure 2 The OC or EC concentration for Before-APEC, During-APEC and After-APEC. The winter heating using fossil fuel was started on November 15, 2014.

examined in this research were similar in the three periods, therefore, the Before-APEC, During-APEC and After-APEC data were combined for analyses and the results are shown in Figure 3. Statistically significant correlations for the After-APEC ($R^2 = 0.996$) and During-APEC ($R^2 = 0.979$) are observed, which suggests that the OC and the EC concentrations came from stable emission sources. The correlation of Before-APEC ($R^2 = 0.757$) is not as significant as that of After-APEC and During-APEC, which indicates its emission sources were less stable than the other two, in addition, we can deduce the presence of mixed emission sources and a contribution of second oxidation aerosols by photochemistry and/or local or long range transport (i.e., the presence of aged aerosols). The slopes of the relationship between OC and EC concentrations for After-APEC, Before-APEC and During-APEC are 3.74, 3.11, 2.67, respectively. There is a correlation between the slopes and artificial control, as the artificial controls begun to make sense before APEC, and human activities were not banned completely until the APEC summit began, but the artificial control were completely relived after APEC. Here different slopes reveal different emission sources.

By studying the characteristics of radiocarbon contained in carbonaceous fractions of atmospheric particulate matter, it is possible to know how fossil and non-fossil sources contribute to air quality. Studies neglected in this simple model a few exceptions such as human-induced wood burning (Hildemann et al. 1994), which might contribute to a minor extent to the non-fossil sources.

As described above, the fraction of non-fossil carbon equals the fraction of biogenic contribution as defined by a simple source apportionment model. Therefore, the ^{14}C measured results can reflect the fraction of biogenic and fossil sources in atmospheric PM2.5. The ^{14}C measured results and error analysis in OC and EC are shown in Figures 4 and 5 and in Table 1.

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Table 1 Measurement results for PM2.5 samples.

Objects	Date	TC [a] (μg·m^{-3})	OC [a] (μg·m^{-3})	EC [a] (μg·m^{-3})	OC-f_{bio} [b] (%)	Average	EC-f_{bio} [b] (%)	Average
Before-APEC	2014-10-28	31.41	22.47	8.94	52.8 ± 1.95	0.537	35.4 ± 1.13	0.356
	2014-10-29	41.08	29.90	11.17	60.7 ± 1.12		31.2 ± 3.16	
	2014-10-30	39.20	29.29	9.92	46.2 ± 2.53		37.3 ± 1.13	
	2014-10-31	39.26	28.63	10.63	54.9 ± 4.06		38.6 ± 1.52	
During-APEC	2014-11-08	32.14	23.89	8.25	50.4 ± 0.90	0.569	46.2 ± 1.00	0.438
	2014-11-09	30.39	22.91	7.48	59.3 ± 3.60		46.1 ± 3.00	
	2014-11-10	32.31	24.85	7.46	60.0 ± 3.37		47.2 ± 3.33	
	2014-11-11	7.53	6.31	1.22	57.7 ± 3.35		35.9 ± 1.00	
After-APEC	2014-11-17	25.75	19.18	5.94	33.1 ± 1.85	0.385	28.0 ± 2.02	0.313
	2014-11-18	63.21	48.67	14.54	46.4 ± 0.93		36.9 ± 5.10	
	2014-11-19	106.68	83.14	23.54	36.1 ± 2.67		35.3 ± 4.07	
	2014-11-20	103.56	81.35	22.20	38.2 ± 1.44		25.1 ± 5.76	

[a]The error of the TC/OC/EC concentration data can be ignored due to the sample content used in the experiment far exceeds the detection limit.

[b]The error was derived from the differences in the results of the six test knots taken from one PM2.5 sample. (In order to obtain accurate data and eliminate the differences in radiocarbon characteristic of the PM2.5 sample on the quartz sampling filter, we took 6 samples on the same quartz filter for ^{14}C-AMS analysis. Statistical error calculated using the number of counts measured from each AMS sample.); OC-$f_f = 1 - (\text{OC-}f_{bio})$; EC-$f_f = 1 - (\text{EC-}f_{bio})$.

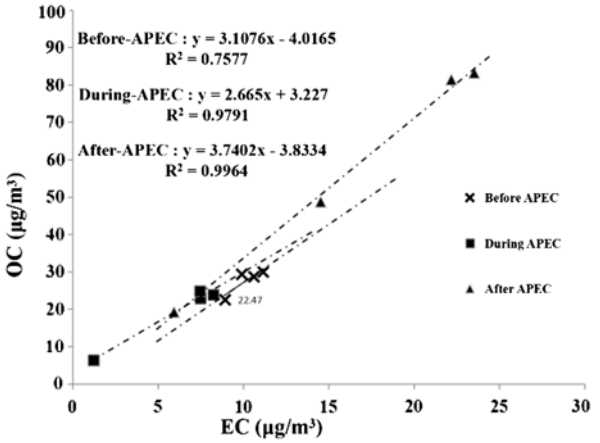

Figure 3 The correlations of OC and EC in all samples.

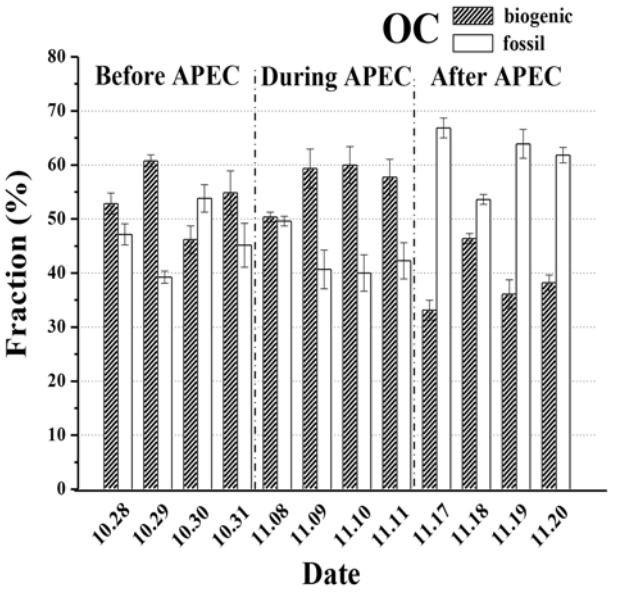

Figure 4 Radiocarbon characteristics of OC.

From the analysis of radiocarbon characteristics in OC of atmospheric PM2.5, it is known that the biogenic contribution for During-APEC is significantly higher than that for After-APEC, slightly higher than that for Before-APEC, which significantly reflects that the emission of the bio-source for OC is closely associated with the human activities. Combined with information in Figure 2, it can be concluded that human activities have significant impact on the pollution emission mode.

The f_f of EC for During-APEC is significantly lower than the other two, and the other two have no obvious difference, therefore, the emission levels of EC are approximately consistent before and after the meeting, which indicates that the emission mode of EC is stable. Moreover, the

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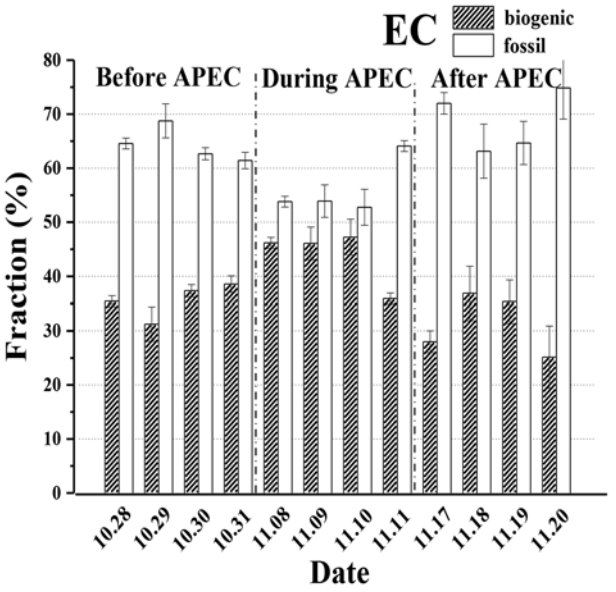

Figure 5 Radiocarbon characteristics of EC.

artificial control of the EC for improving the air quality will not have a major impact except to stop traffic activities. From June 1, 2014, the control of air pollution in Beijing began with the implementation of specific measures including the closure of some coal-fired power plants, the elimination of heavily polluting vehicles, and the use of clean energy alternatives, especially, the traffic was forbidden completely during APEC summit. However, the industrial activities and traffic activities in Beijing and its neighboring regions were resumed immediately as the summit concluded. Particularly worth mentioning is the winter heating using fossil fuel started at November 15, 2014. In terms of the fossil source contribution, comparing with the emission levels of EC, there exists obvious difference between Before-APEC and After-APEC for those of OC. It is well known that OC is produced along with EC, thus, the difference is due to the oxidation and accumulation of OC from fossil sources.

As described in the introduction, OC is associated with biomass burning origin and human activities, and EC is associated with insufficient combustion of fossil sources. The APEC Blue best reflects the impact of short-term artificial controls on atmospheric environment, especially the lowest value of EC-f_f is the best proof, so the lowest value was used as the reference value of During-APEC. However, as for After-APEC and Before-APEC, the average value of EC-f_f could better reflect the actual situation of daily life and was considered as the reference values. The origin of EC in atmospheric PM2.5 is directly related to direct discharge from fossil sources. Figure 5 shows the fraction of fossil of EC for Before-APEC is about 21.9 % higher than that for During APEC. The obvious difference between them was attributed to traffic activities forbidden completely during APEC, but not completely stopped before and after APEC. Therefore, the traffic contribution to EC in daily atmospheric PM2.5 is approximately 21.9 %. Figure 4 shows the f_f of OC for During-APEC is 15.8 % lower than that for Before-APEC and 53.8 % lower than that for After-APEC. This difference may be mainly caused by industrial activities. It can be concluded that the contribution from traffic activities and industrial

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activities to OC in daily atmospheric PM2.5 is 15.8 % and 38.0 %, respectively. Similarly, the contribution of fossil sources caused by industrial activities to EC is about 8.2 %. In addition, the f_f of EC and OC reached a relatively high value in about 7–10 days after stopping the artificial control as shown in Figure 4 and Figure 5, and this inference is consistent with the information reflected in Figure 2. We can use these data to deduce how long it is needed to achieve the state of balance between human activities and atmospheric disperse dilution. However, considering Beijing's winter heating increases the fraction of fossil of After-APEC, this may complicate the estimate.

CONCLUSIONS

The APEC meeting held in Beijing in 2014 is an excellent event to study the causes of atmospheric pollution due to the interruption of normal traffic and industrial activities during that time. The concentration characteristics of OC/EC in atmospheric PM2.5 samples collected during the APEC meeting were analyzed. The radiocarbon characteristics of OC and EC in the samples were analyzed by the CIAE-SSAMS. Based on the above study, the effects of traffic activities and industrial activities on carbon in atmospheric PM2.5 for Before-APEC, During-APEC and After-APEC in Beijing were quantitatively analyzed. The contributions to OC and EC are 15.8 % and 21.9 %, respectively, from traffic activities using fossil fuels. The fossil source contributions of industrial activities to OC and EC are 38.0 % and 8.2 %, respectively. We estimate that about 7–10 days is needed to take to achieve a state of balance between human activities and atmospheric disperse dilution. Through the analysis of radiocarbon characteristics in this typical case, it helps people understand the causes of air pollution and lays the foundation for improving the atmospheric problems.

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