RESEARCH ARTICLE

Key Points:
- The 56% OC and 36% EC were from nonfossil sources before APEC
- The 61% OC and 46% EC were related with the combustion of nonfossil sources during APEC
- Atmospheric visibility would be reduced by 20% if nonfossil sources were not controlled during APEC

Radiocarbon-derived source apportionment of fine carbonaceous aerosols before, during, and after the 2014 Asia-Pacific Economic Cooperation (APEC) summit in Beijing, China

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Abstract The Asia-Pacific Economic Cooperation (APEC) summit took place in Beijing, China, 5–11 November 2014, during which numerous measures were performed to control the air pollution, and consequently, the sky of Beijing was so clean that the public called it "APEC blue." The concentrations before, during, and after the APEC summit are $14.4 \pm 6.81\,\mu g\,C/m^3$, $6.66 \pm 2.99\,\mu g\,C/m^3$, and $32.3 \pm 10.6\,\mu g\,C/m^3$, respectively, for organic carbon (OC), and $2.27 \pm 1.17\,\mu g\,C/m^3$, $0.76 \pm 0.52\,\mu g\,C/m^3$, and $4.99 \pm 1.74\,\mu g\,C/m^3$, respectively, for elemental carbon (EC). We quantify the contributions of fossil and nonfossil sources to the OC and EC using radiocarbon. Results show that the contribution of nonfossil sources is $56 \pm 1\%$ (before APEC), $61 \pm 1\%$ (during APEC), and $48 \pm 1\%$ (after APEC), respectively, for OC, and $36 \pm 4\%$ (before APEC), $46 \pm 1\%$ (during APEC), and $33 \pm 4\%$ (after APEC), respectively, for EC. Comparing to the period before APEC, 70% and 60% of fossil EC and OC and 60% and 50% of nonfossil EC and OC are reduced, respectively, implying that the control on the nonfossil sources has considerable contribution to the good air quality in Beijing. Both EC and OC mass loadings during the APEC summit would have increased by 60% if the biomass-burning activities were not taken into account for control. In such a case, the atmospheric visibility would decrease 20% at least and the blue sky thereby would likely not have been visible during the summit.

1. Introduction

The atmospheric brown cloud (ABC), which evolved from long-lasting and large-scale air pollution events consisting of numerous particulate and gas pollutants, has many adverse impacts on air quality, climate, human health, and agriculture in our Earth (Project Atmospheric Brown Cloud, http://www-abc-asia.ucsd.edu/). Recently, studies concerning the ABC or regional air pollution in East Asia, and especially the ingredient critical to the ABC, so-called carbonaceous aerosols (CAs), have drawn great attention worldwide [*Alexander et al.*, 2008; *Chen et al.*, 2013; *Huang et al.*, 2014; *Kirillova et al.*, 2014a; *Zhang et al.*, 2015a], mainly due to the dense habitation and huge pollutant emissions in this region and the complicated sources and atmospheric process with respect of CAs. A latest study has comprehensively pointed out that 92% of the population of China experienced >120 h of unhealthy air [*Rohde and Muller*, 2015] and ~1.2 million die prematurely in 2010 owing to the severe air pollution in China [*Lim*, 2013]. While, most compounds in CAs cannot be isolated or identified at the molecular level by now. Therefore, CAs are generally subdivided into the fractions of organic carbon (OC) and elemental carbon (EC or black carbon), based on their physical and chemical behaviors. OC aerosols mainly scatter solar light and exert a net cooling effect on the climate [*Stocker et al.*, 2013], whereas EC aerosols strongly absorb solar light and make a great contribution to global warming [*Stocker et al.*, 2013; *Bond et al.*, 2013]. Diverse processes such as traffic exhaust, coal combustion, agricultural burning, and the domestic burning of biofuel emit a large amount of OC and EC aerosols in the atmosphere. The widely used bottom-up technique estimated that global contribution of fossil fuel, biofuel, and open burning are 38%, 20%, and 42% for EC and 7%, 19%, and 74% for OC [*Bond et al.*, 2004]. In North China, *Zhao et al.* [2012] believed that industry, straw burning, and traffic contributed 38%, 39%, and 15% for EC and 37%, 45%, and 10% for OC, respectively. Results basing a new emission inventory (PKU-BC (China)) revealed that

the major sources for EC in China were residential coal (28%), residential biomass (23%), coke production (18%), diesel vehicles (8%), and brick kilns (7%) [*Wang et al.*, 2012] and believed that ~50% of global EC were coming from Asia [*Wang et al.*, 2014]. However, the relative contribution from different sources to atmospheric OC and EC is still poorly constrained due to the large uncertainty derived by the bottom-up method, typically about a factor of 2 [*Bond et al.*, 2004; *Zhao et al.*, 2012] or more in some heavily polluted areas. It is reported that the uncertainty of EC derived from bottom-up technique in China could be up to a factor of 8 due to the highly variable fuels and combustion conditions [*Streets et al.*, 2001], such as coals with different thermal maturities for residential burning [*Chen et al.*, 2006]. Therefore, an accurate source apportionment for OC and EC is urgently needed to refine the understanding of regional air pollution, especially in the key region of East Asia, with the largest carbonaceous aerosol emissions in the world.

Radiocarbon-based top-down (^{14}C, $\lambda_{1/2} = 5730$ a) measurements represent a state-of-the-art technique for the source apportionment of OC and EC, based on the simple fact that ^{14}C in fossil sources (coal, gasoline, and diesel) is completely depleted, but in grass, wood, crops, and other biomass materials (nonfossil sources), ^{14}C is maintained at a relatively stable level. Earlier studies performed on the European continent showed that nonfossil (~50–100%) and fossil sources (~65–95%) are the dominant contributors to OC and EC aerosols, respectively [*Bernardoni et al.*, 2013; *Dusek et al.*, 2013; *Szidat et al.*, 2004a, 2004b, 2006, 2007, 2009; *Zotter et al.*, 2014], predominantly depending on the sampling time and location. Given that the strict environmental standards and clean air in Europe, these results largely reflect the ^{14}C levels of CA$_S$ in an atmosphere that is less polluted by factories, traffic exhausts, and other anthropogenic sources. Yet the ^{14}C levels of CAs in the region of South Asia that is enveloped by the large-scale ABC display a distinct characteristic. It was reported that up to ~50–85% of EC and ~80–90% of water-soluble OC over South Asia were derived from nonfossil sources, implying the importance of domestic cooking and agricultural burning on regional air pollution in India and the surrounding areas [*Bosch et al.*, 2014; *Gustafsson et al.*, 2009; *Kirillova et al.*, 2014b]. Compared to South Asia, only ~20–30% of EC aerosols originate from nonfossil sources over East Asia [*Andersson et al.*, 2015; *Chen et al.*, 2013; *Zhang et al.*, 2015a], with the rest coming from fossil sources. Also, both fossil (~35–60%) and nonfossil (~40–65%) sources have comparable contributions to OC aerosols in East Asia [*Zhang et al.*, 2015a]. All of these ^{14}C-related results strongly suggest that the large-scale air pollution occurring in East Asia is not a single product of one emission source but a comprehensive outcome of multiple factors, including the intense use of coal that has almost been abandoned by many developed societies, the rapid increase in oil-fired vehicles, the extensively outdoor and indoor burning of wood fuel and other biomass, and relatively lower environmental standards. Consequently, the heavy air pollution in East Asia is unique from that experienced by Western countries during 1940s and 1950s [*He et al.*, 2014], and the lessons learned from that experience can therefore probably not be directly applied here.

To mitigate air pollution, the Chinese government has strictly updated its regulations on airborne pollutant emissions since 2012 (Chinese Ambient Air Quality Standards, GB 3095–2012). The new 24 h particulate matter (PM)$_{2.5}$ standard is first set to 75 μg/m^3. It is interesting to determine how the air quality and sources of pollutants would develop if the government instituted rigorous regulations on emission sources. This knowledge would provide valuably useful information and data set to the control of severe air pollution in China. To date, few study concentrated on the ^{14}C analysis in China, and all these studies [*Andersson et al.*, 2015; *Chen et al.*, 2013; *Liu et al.*, 2014; *Zhang et al.*, 2015a] were conducted in air that has had no manual intervention, leading to difficulties in assessing the performance of air pollution treatments and in finding the culprits of emission sources. The Asia-Pacific Economic Cooperation (APEC) summit, hosted by Huairou District, Beijing, China, 5–11 November 2014, provides an excellent opportunity to address this troublesome issue. In total, leaders from 21 countries and regions including United Sates, China, Japan, South/North Korea, Russia, Australia, Canada, Chile, Indonesia, Thailand, and New Zealand were present at this summit. In order to make a good air quality for this summit, almost all main emission sources in Beijing and adjacent areas had been strictly controlled during the APEC period. During this period, numerous regulatory control measures, including traffic movement restrictions, temporary cessations of operation for thousands of polluting factories, administrative holidays for civil servants and students, and strict limitations on the open/domestic combustion of biomass, were implemented by official order in Beijing and the surrounding provinces with the aim of improving air quality for this important summit. Consequently, the public coined the phrase, "APEC blue," to describe the artificially clear sky in Beijing during the summit. This good air quality—aerosol optical depth
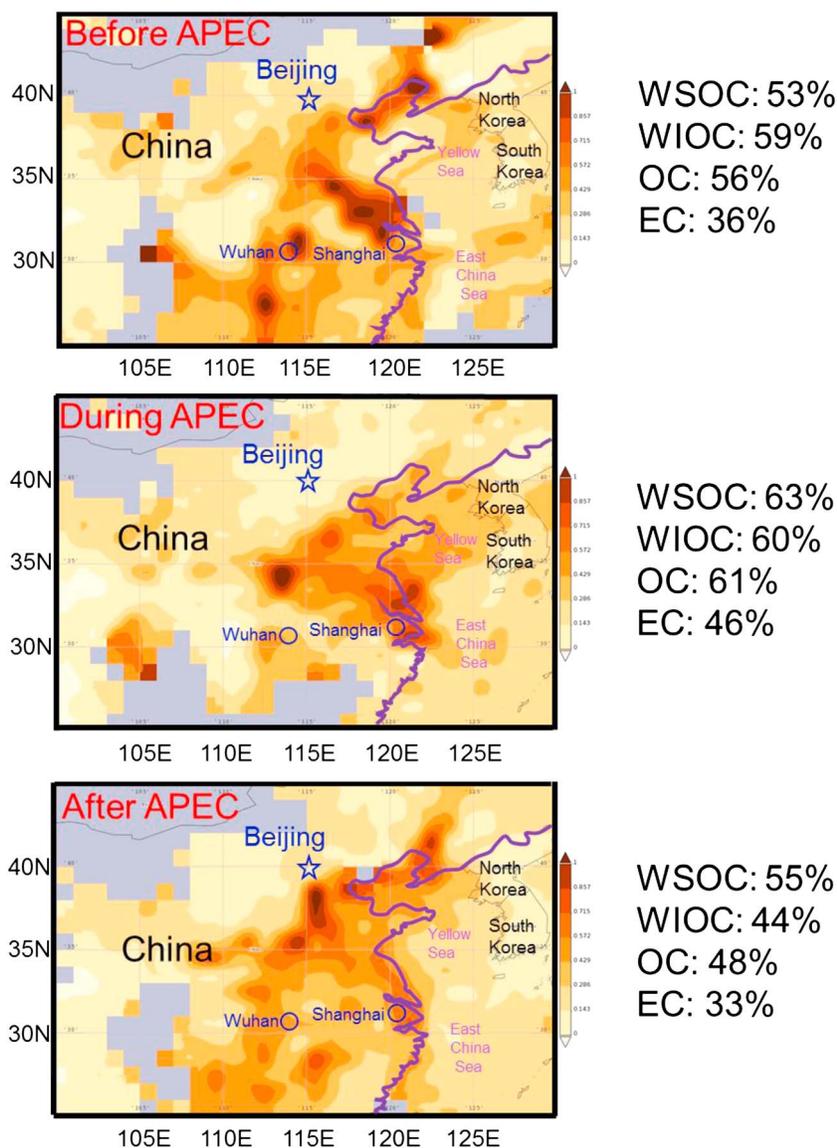
Figure 1. Map showing the geographic location of Beijing in China and the average aerosol optical depth (AOD) and the contribution of nonfossil sources to different carbon fractions (top) before, (middle) during, and (bottom) after APEC summit.

(AOD) <0.1—can be monitored vividly from space during the APEC period (Figure 1). In this paper, we collected fine particle (PM$_{2.5}$) samples in Beijing before, during, and after the APEC period with the objectives of (1) determining a clear ^{14}C-derived top-down source apportionment for OC, EC, and other carbon fractions and (2) quantitatively assessing the performance of government-led air pollution controls during the APEC period. These fundamental data will greatly improve the understanding of complicated emission sources in East Asia and refine calculations of the radiative forcing values for OC and EC aerosols in this key region.

2. Methods

2.1. Sampling

The sampling campaign was performed from 25 October 2014 to 29 November 2014 at Yanqi Lake Campus of the University of Chinese Academy of Sciences, Huairou District, Beijing (40.4°N, 116.7°E), which is about 60 km away from Beijing urban center. PM$_{2.5}$ sampler (XTrust Instrument, Shanghai, China) was placed on
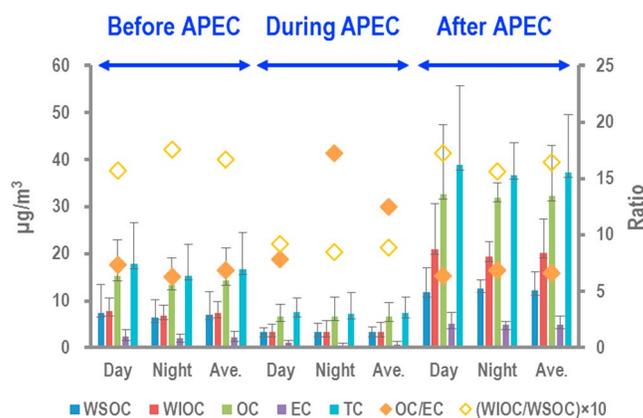
Figure 2. Concentrations of carbon fractions (left *Y* axis) and the ratios of OC/EC and WIOC/WSOC (right *Y* axis) during the sampling campaign.

the roof of one teaching building with ~20 m height. No obvious point emission sources were observed during the sampling campaign. Eighteen $PM_{2.5}$ samples were selected in total in this study, of which six samples for one period, that is, before, during, and after APEC summit. These selective samples were collected in day (09:00–21:00 h) and night (21:00–09:00 h) on precombusted quartz filters (8 × 10 inch; Pall, NY, USA). In order to get enough carbon amount (~15 μg) for ^{14}C measurement in our lab, all day or night samples were combined together for a given period. Table S1 in the supporting information presents the sampling time and meteorological parameters of these samples. After sampling, filters were folded in half, wrapped in aluminum foil, and stored at −20°C until analysis.

2.2. Analysis and ^{14}C-Derived Source Apportionment

We quantified carbon fractions using a commercial carbon analyzer equipped with a nondispersive infrared detector (Sunset Laboratory Inc., USA), following the manufacturer's instructions to measure the total carbon (TC) in the original filters. To eliminate the potential charring issue that arises from the water-soluble organic carbon (WSOC) fraction [*Yu et al.*, 2002; *Zhang et al.*, 2012], a portion of filter sample was cut and washed with 20 mL ultrapure water [*Liu et al.*, 2014]. After drying in a desiccator, water-extracted filter portions were analyzed for the concentrations of water-insoluble organic carbon (WIOC) and EC using the EUSAAR2 method [*Cavalli et al.*, 2010]. WSOC was calculated as TC − WIOC − EC. OC is estimated by the difference of TC and EC. The uncertainties of TC (5.9%), WIOC (6.7%), and EC (7.6%) were calculated for from four measurements of one sample in this study, while the uncertainties of WSOC (11.7%) and OC (9.6%) were estimated basing on error propagation formulas.

Carbon fractions must be converted into pure CO_2 individually prior to their ^{14}C measurements, for which preparation approaches have been described in our previous studies [*Liu et al.*, 2013, 2014; *Zhang et al.*, 2010]. In this study, the isolated carbon amounts were typically in the range of 30–100 μg, depending on the samples. Briefly, carbon fractions were first combusted and isolated at distinct temperatures, and then all corresponding CO_2 fractions were cryogenically trapped and transformed into graphite targets for ^{14}C determination with the National Electrostatics Corporation compact accelerator mass spectrometry (AMS) facility at Peking University. All ^{14}C results are expressed as fractions of modern carbon (f_m) and corrected for decay between 1950 and the year of measurement. All AMS-derived f_m values were corrected by δ^{13}C values and are listed in Table S2 in the supporting information. The f_m uncertainty was ~1%. The f_m values for WSOC were calculated from the isotopic mass conservation. The f_m must be divided by the reference values (f_{ref}) to eliminate the bomb effect when studies are focused on the source apportionment of carbonaceous aerosols. Here f_{ref} for OC including WSOC and WIOC and EC are 1.052 ± 0.013 and 1.064 ± 0.010, respectively, based on the long-term time series of $^{14}CO_2$ at background stations [*Levin and Kromer*, 2004; *Levin et al.*, 2013] and with an assumption that the proportions of annual plants, 10 year old wood, and 20 year old wood in biomass burning were 80%, 10%, and 10%, respectively, in north China. The f_m values of biogenic emissions were the same as for $^{14}CO_2$ in 2014. Although different f_{ref} values may be selected in other studies, this uncertainty is generally <5%. f_{ref} of TC was the average of OC and EC.

3. Results

3.1. Mass Concentrations and Ratios

Figure 2 shows the concentrations of carbon fractions and the values of OC/EC and WIOC/WSOC. Before the APEC summit, the average TC, OC, and EC concentrations amounted to 16.6 ± 7.95 μg C/m³, 14.4 ± 6.81 μg C/m³,
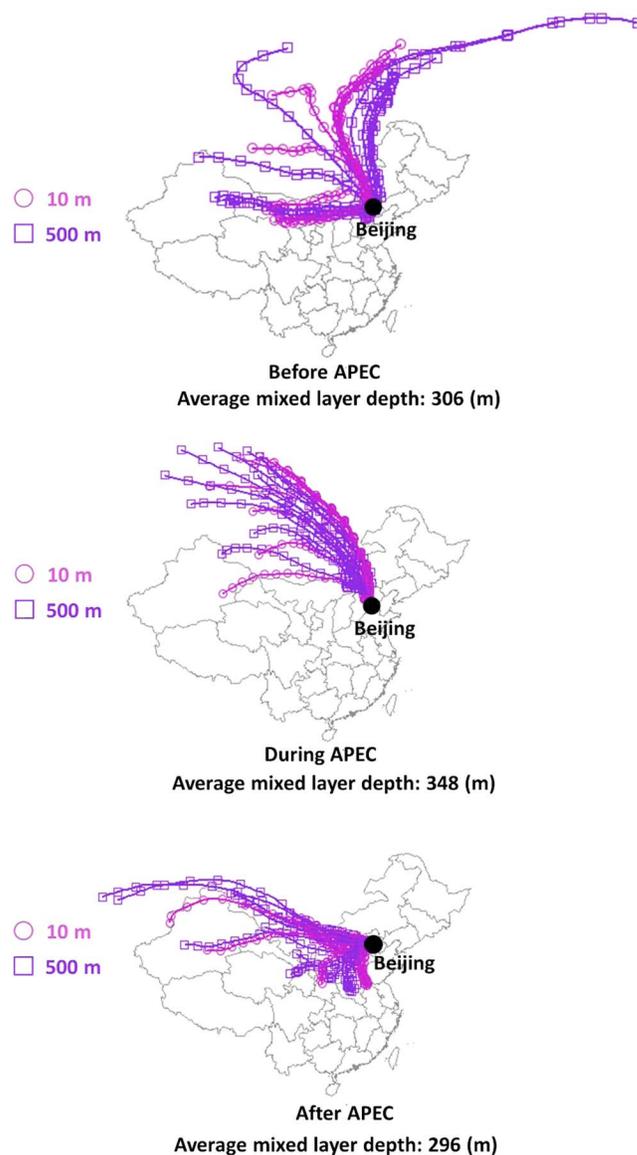
Figure 3. The 72 h back trajectories at 10 m and 500 m aboveground level and average mixed layer depth (top) before, (middle) during, and (bottom) after the APEC summit. All these data were obtained from Air Resources Laboratory, NASA.

and $2.27 \pm 1.17\,\mu g\,C/m^3$ and then decreased to $7.42 \pm 3.35\,\mu g\,C/m^3$, $6.66 \pm 2.99\,\mu g\,C/m^3$, and $0.76 \pm 0.52\,\mu g\,C/m^3$ during the APEC period, respectively. After the summit, the average TC concentration sharply increased to $37.3 \pm 12.3\,\mu g\,C/m^3$, up to ~3 times and ~5 times the values before and during the summit, respectively. The changes in the patterns of OC and EC aerosols were similar to those of TC (Figure 2). For the samples we selected in this study, air masses affecting Beijing were predominantly derived from the west and north before the APEC summit, the north during the APEC summit, and the west after the APEC summit, respectively (Figure 3). In North China Plain, severe air pollution occurs in the regions of southern and eastern Beijing [*Zhao et al.*, 2013; *Hu et al.*, 2015], which also can be observed vividly in Figure 1. Thus, air masses from the west and/or north would not result in a significant change on the sources of carbonaceous aerosols in Beijing and its adjacent areas, which, in fact, have been confirmed by a recent study [*Guo et al.*, 2014]. Moreover, the average mixed layer depth and wind speed before the summit were very close to that during the APEC (Figure 3 and Table S1 in the supporting information). Therefore, lowest concentrations of OC and EC during the APEC summit reflect the effective controls on the emission sources in Beijing and surrounding regions. The highest carbon concentrations in the period after the APEC summit can be attributed to both the uncommitted pollutant emissions in this period and its relatively weaker atmospheric dilution. The wind speed after the APEC period was 1.5 m/s, only ~50% of the other two periods (Table S1 in the supporting information). Additionally, central heating driven by coal combustion in Beijing, beginning on November 15, was another important factor for the sharp enhancement of CAs after the summit. This can be confirmed by the rapid increase of other chemicals during the heating period. For example, $PM_{2.5}$ sulfate and nitrate mass loadings during heating period in Beijing were 2–4 times those before heating in 2014 [*Yang et al.*, 2015]. In all, the concentrations reported in this study are consistent with a comprehensive observation in China: ~3–40 $\mu g\,C/m^3$ for OC and ~0.3–15 $\mu g\,C/m^3$ for EC, with a trend of background < regional < urban [*Zhang et al.*, 2008]. In fact, the OC and EC loadings during the APEC period were very close to those at Chinese background sites [*Zhang et al.*, 2008]. For all three periods, EC concentrations in daytime were only higher by ~0.4 $\mu g\,C/m^3$ than that in nighttime, demonstrating the limited changes of combustion sources for a given period. Note that nighttime OC during the APEC summit was higher than that in daytime, which is apparently different from the period before APEC and after APEC. As we discuss below, more secondary OC (SOC) probably was formed during the nighttime of the APEC summit.

Table 1. The Contributions of Nonfossil and Fossil Sources to the Carbon Fraction

Periods		WSOC			WIOC			OC			EC			TC		
		n^a	f^b	n/f^c	n	f	n/f	n	f	n/f	n	f	n/f	n	f	n/f
Before APEC	D^d	52 ± 6%	48 ± 6%	1.08	57 ± 4%	43 ± 4%	1.33	55 ± 5%	45 ± 5%	1.22	32 ± 2%	68 ± 2%	0.47	53 ± 3%	47 ± 3%	1.13
	N^e	53 ± 6%	47 ± 6%	1.13	61 ± 4%	39 ± 4%	1.56	57 ± 6%	43 ± 6%	1.33	40 ± 3%	60 ± 3%	0.67	55 ± 3%	45 ± 3%	1.22
	Ave^f	53 ± 1%	47 ± 1%	1.13	59 ± 2%	41 ± 2%	1.43	56 ± 1%	44 ± 1%	1.27	36 ± 4%	64 ± 4%	0.56	54 ± 1%	46 ± 1%	1.17
During APEC	D	62 ± 7%	38 ± 7%	1.63	58 ± 4%	42 ± 4%	1.38	60 ± 6%	40 ± 6%	1.50	47 ± 4%	53 ± 4%	0.89	57 ± 4%	43 ± 4%	1.33
	N	63 ± 7%	37 ± 7%	1.70	61 ± 4%	39 ± 4%	1.56	62 ± 6%	38 ± 6%	1.63	45 ± 4%	55 ± 4%	0.82	58 ± 4%	42 ± 4%	1.38
	Ave	63 ± 1%	37 ± 1%	1.70	60 ± 2%	40 ± 2%	1.50	61 ± 1%	39 ± 1%	1.56	46 ± 1%	54 ± 1%	0.85	58 ± 1%	42 ± 1%	1.38
After APEC	D	53 ± 6%	47 ± 6%	1.13	46 ± 3%	54 ± 3%	0.85	49 ± 5%	51 ± 5%	0.96	29 ± 2%	72 ± 2%	0.40	47 ± 3%	53 ± 3%	0.89
	N	56 ± 7%	44 ± 7%	1.27	42 ± 3%	58 ± 3%	0.72	47 ± 5%	53 ± 5%	0.89	36 ± 3%	64 ± 3%	0.56	45 ± 3%	55 ± 3%	0.82
	Ave	55 ± 1%	45 ± 1%	1.22	44 ± 2%	56 ± 2%	0.79	48 ± 1%	52 ± 1%	0.92	33 ± 4%	67 ± 4%	0.49	46 ± 1%	54 ± 1%	0.85
Total	Ave^g	57 ± 4%	43 ± 4%	1.33	54 ± 7%	46 ± 7%	1.17	55 ± 5%	45 ± 5%	1.22	38 ± 7%	62 ± 7%	0.61	53 ± 5%	47 ± 5%	1.13

[a]The contribution of nonfossil sources.
[b]The contribution of fossil sources.
[c]The ratio of a to b.
[d]Day.
[e]Night.
[f]The average of day and night.
[g]The total average.

The ratio of WIOC/WSOC was relatively constant in the two periods without pollution controls. Before the summit, WIOC/WSOC was 1.57 ± 0.77 in the day and 1.76 ± 1.17 at night, with an average of 1.67 ± 0.99. After the summit, these values were 1.72 ± 0.33, 1.56 ± 0.35, and 1.64 ± 0.35, respectively. Meanwhile, the average WIOC/WSOC was only 0.89 ± 0.46 during the APEC summit, with a value of 0.92 ± 0.35 in the day and 0.85 ± 0.60 at night. A majority of ambient WIOC is produced directly by the emission sources. It was reported that at least 90% of primary OC (POC) was water-insoluble, based on the measurements from semicontinuous instruments [*Miyazaki et al.*, 2006]. Thus, the relatively higher WIOC/WSOC ratios could, in principle, reflect the intense primary emissions before and after the APEC period. All OC/EC ratios maintained a relatively stable level (6–8), with an exception at night during the APEC period (Figure 2). In general, a rapid increase of the OC/EC ratio suggests (1) an invasion event of biomass-burning plumes or (2) that the formation rate of SOC is faster than the EC emission rate. Table 1 shows that the ^{14}C level of EC in day was almost the same as at night during the APEC summit, which strongly demonstrates that there was no invasion of biomass-burning plumes. Thus, much higher OC/EC at night during the APEC period was most likely due to a faster SOC formation rate relative to the emission rate of EC. In fact, during the APEC period, the temperature at night (3°C) was less than one third of that during the day (11°C; Table S1 in the supporting information), which would greatly facilitate the conversion of semivolatile OCs (VOCs) to OC, especially in conditions of lower wind speed and higher humidity due to the occurrence of aqueous-phase processing [*Sun et al.*, 2013].

3.2. ^{14}C-Derived Source Apportionment

Since the differences of ^{14}C levels between day and night among three periods were very small, we will discuss the average ^{14}C levels in the following text. Looking at all three periods surrounding the APEC summit together, the contributions of nonfossil sources to WSOC, WIOC, OC, EC, and TC were 57 ± 4%, 54 ± 7%, 55 ± 5%, 38 ± 7%, and 53 ± 5%, respectively, with the remainder coming from fossil sources (Table 1). The average ratio of the contribution of nonfossil source to that of fossil sources (n/f) was 1.33, 1.17, 1.22, 0.61, and 1.13 for WSOC, WIOC, OC, EC, and TC, respectively. Note that all n/f values exceeded one with the exception of EC, implying the large importance of nonfossil sources to the air pollution in Beijing and the surrounding regions, which probably is an important factor resulting in the severe air pollution in north China [*Zhang and Cao*, 2015]. While, comparing to South Asia that also is suffering air pollution, the reported ^{14}C levels in this study are much lower. In South Asia, up to 86 ± 5% of WSOC were derived from the nonfossil sources [*Bosch et al.*, 2014], 30% higher than our measurements. Similarly, the contribution of nonfossil sources to EC in South Asia was 59 ± 4% [*Bosch et al.*, 2014], 20% and 35–40% higher than that in this study and recent studies [*Zhang et al.*, 2015a, 2015b], respectively. This comparison largely suggests that air pollution covering East Asia is related more closely with the extensive usage of fossil fuel than South Asia. At the same time, we find

that the contribution of nonfossil sources to EC is typically higher than those some cities with good air quality. For example in Zurich, Switzerland, it was reported that the proportion of nonfossil sources is only ~6–25% [*Szidat et al.*, 2006], demonstrating that nonfossil sources play a more important role in the air pollution of Beijing comparing to those developed cities in Western Europe. All these results demonstrate that both fossil and nonfossil sources are key factors controlling the formation of air pollution in East Asia. In addition, we observed that the contribution of nonfossil sources to all EC aerosols was lower than other carbon fractions, as has been found independently at other times and locations [*Liu et al.*, 2014; *Szidat et al.*, 2004b; *Zhang et al.*, 2014], and thus probably is a universal phenomenon for ambient aerosols on Earth.

The ^{14}C content in carbon fraction shows a different characteristic among three periods. Nonfossil sources made its largest contribution to EC during the APEC summit, up to 46% with an *n/f* of 0.85, followed by the before (36%, *n/f* = 0.56) and after periods (33%, *n/f* = 0.49). Higher ^{14}C level during the APEC period means that the control strength of fossil sources were stronger than that of nonfossil sources. Even so, we find that the contribution of nonfossil sources to EC during the periods without pollution controls displayed a higher level than that in the winter season, during which nonfossil sources only accounted for ~15–25% of EC on average [*Chen et al.*, 2013; *Zhang et al.*, 2015a]. This result corresponds to the fact that agricultural residue/straw is burned actively around Beijing and peaks during mid-October to mid-November [*He et al.*, 2001]. In north China, these biomass materials with modern ^{14}C levels are frequently burned both in agricultural fields after harvesting and in rural stoves for cooking. While, satellites cannot monitor the obvious changes of open fire activities in Beijing and adjacent areas among the three periods we studied (Figure S1 in the supporting information). Thus, we speculate that these biomass-burning activities were mainly performed in indoor environment such as the burning of biofuels for heating and cooking.

Similar to EC, the highest contribution of nonfossil sources to OC was observed during the APEC period. Specifically, the average contributions of nonfossil sources in the periods before, during, and after the APEC summit were 53%, 63%, and 55% for WSOC and 59%, 60%, and 44% for WIOC, respectively. The ^{14}C level of WSOC between the periods before and after APEC is almost the same, although the period after APEC summit was impacted by the coal combustion due to the central heating as we mentioned above. This is because ambient WSOC is a mixture of secondary OC (SOC) and biomass-burning primary OC (POC) that is emitted by biomass burning and fossil fuel combustion barely produces water-soluble POC [*Weber et al.*, 2007; *Dai et al.*, 2015]. Conversely, the contribution of nonfossil sources to WIOC after APEC summit is lower by 15% comparing to other two periods, which also suggests that coal combustion predominantly release the WIOC fraction. Concerning to whole mass closure of CAs, the largest carbon fractions before, during, and after the APEC summit were nonfossil WIOC, nonfossil WSOC, and fossil WIOC, respectively (Figure S2 in the supporting information). Nonfossil EC was the smallest fraction (4–6%) of the whole TC particles for all three periods, which has also been observed in remote and urban sites [*Liu et al.*, 2014; *Zhang et al.*, 2014]. As discussed, Beijing began to be intensively impacted by coal combustion due to central heating beginning on November 15, which led directly to the largest proportion of fossil WIOC after the APEC summit in this study. Nevertheless, the fraction of nonfossil WIOC still made up a large proportion (>20%) of the TC source apportionment pie chart (Figure S2 in the supporting information), which was also observed before and during the APEC summit (>25%). Given that the low contribution of plant debris to OC (~1%) [*Guo et al.*, 2012] and the few emission of biogenic VOCs during winter in Beijing [*Wang et al.*, 2003], this result strongly indicates the importance of biomass burning on Beijing's air quality, especially as a large fraction (>50%) of fresh biomass-burning OC probably is water-insoluble [*Iinuma et al.*, 2007].

3.3. The Importance of Nonfossil Source to Atmospheric Visibility

It is well known that Chinese government-led air quality improvement measures successfully resulted in a clear sky in Beijing during the APEC period. This is reflected in our measurements of the concentrations of CAs before, during, and after the APEC summit. Some previous studies have pointed out the importance of biomass burning on air quality in China. Using positive matrix factorization model, *Chen et al.* [2013] found that ~50% of EC and OC aerosols in Beijing are associated with biomass-burning activities. *Cao and Zhang* [2015] suggested making more efforts on the controls of nonfossil sources to slow down the severe air pollution in China. Yet the views of the government and recent studies seemingly suggest that the reduction of fossil sources, such as traffic- and industry-related emissions, was practically the only factor contributing to the blue sky during the APEC summit. The Beijing Municipal Environmental Protection Bureau pronounced
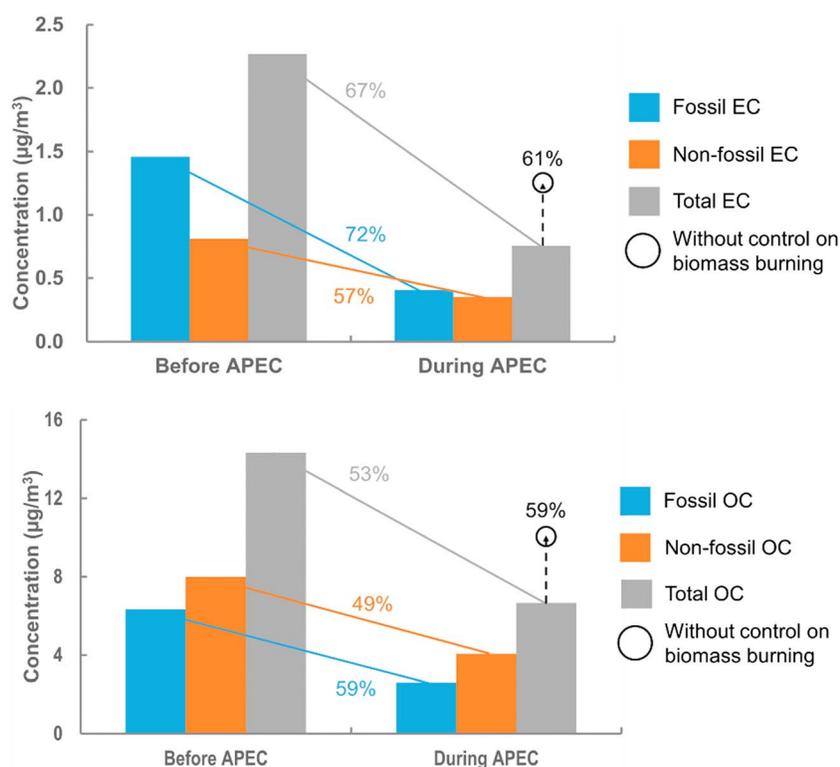
Figure 4. Fossil and nonfossil (top) OC and (bottom) EC concentrations (*Y* axis) and their reduction rates (percentage numbers) during the APEC summit comparing to the period of before APEC. The dashed lines mean the hypothetical cases without the control of biomass-burning activities.

that the reduction of $PM_{2.5}$ during the summit could be fully attributed to controls on vehicles (39.5%), coal combustion and industry (17.5%), building construction (19.9%), street cleaning (10.7%), and to administrative holidays (12.4%), respectively (http://www.bjepb.gov.cn). In another study, *Li et al.* [2015] found that the largest contributor to ambient VOCs was fuel combustion, followed by vehicular exhaust and liquefied petroleum gas during the APEC period. Both reports completely neglected nonfossil sources, whose importance in air pollution has been clearly confirmed by our study. Nonfossil sources exerted considerable impacts on all carbon fractions during the sampling campaign—that is, before, during, and after the APEC period.

Figure 4 shows the reduction rates of fossil and nonfossil sources for OC and EC aerosols during the APEC period compared to before it. Since the wind speed after the APEC summit was much lower than during the other two periods and the invasions of intensive emissions from coal combustion began on 15 November, we did not calculate the reduction rates after the APEC period. Meanwhile, the samples we selected for before and during the APEC summit had the similar wind speeds and mixed layer depths, and thus, the differences in atmospheric dilution effects among these samples should be minimal. Compared to the period before the APEC summit, fossil EC and nonfossil EC were reduced by 72% and 57%, respectively, during the APEC period, indicating that nonfossil source controls contributed a great contribution to the good air quality during the APEC summit. The total EC concentration during the APEC summit have increased from 0.76 C $\mu g/m^3$ to 1.22 C $\mu g/m^3$ (Figure 4) with an enhancement rate of 61% if nonfossil sources were not taken into account and controlled before APEC. We observed similar changes for OC aerosols. Given that fine CAs are the key ingredients that lead to the lowering of atmospheric visibility, a rapid increase of OC and EC would likely have destroyed the blue sky during the APEC period. Aerosol light extinction coefficient (b_{ext}, in Mm^{-1}), which is a function of sulfate, nitrate, ammonium, organics, EC, and the relative humidity, has a negative correlation with the atmospheric visibility (Figure 5). We analyzed these ion concentrations to make a comparison for the b_{ext} values with and without nonfossil control during the summit. During the APEC period, the b_{ext} was ~118 Mm^{-1} with an atmospheric visibility of ~13.8 km. However, the b_{ext} value would increase
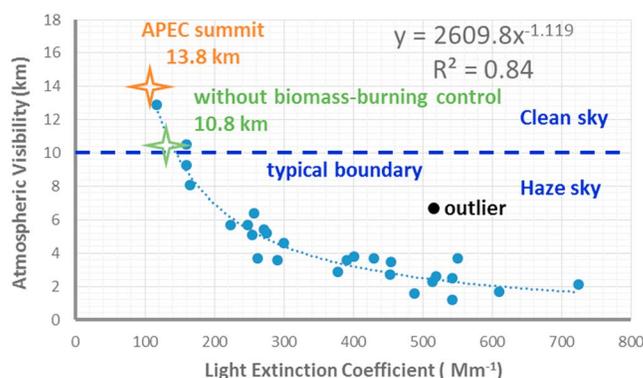
Figure 5. Atmospheric visibility during APEC summit with and without biomass-burning control. The established correlation between atmospheric visibility and light extinction coefficient was based on the data set in *Tao et al.* [2009]. One outlier datum was excluded.

to ~135 Mm^{-1} if the nonfossil OC and EC that had been reduced during the summit were added to the calculation of b_{ext} value, and the corresponding atmospheric visibility would decrease to ~10.8 km, very close to the so-called typical boundary of haze-clear sky (Figure 5). On other hand, in addition to emitting CAs, biomass-burning activities would also produce substantial loadings of sulfate, nitrate, and ammonia, which could reduce atmospheric visibility as well [*Tao et al.*, 2009]. The APEC summit provided a hard-won opportunity to gain the experience in air pollution control over such a large region, and the APEC blue has demonstrated clearly that Chinese government actually is fully capable of controlling the severe air pollution. The presented data set in our study reveals that the control of nonfossil sources activities—most of which probably from the adjacent rural and suburban areas in Beijing—has a great contribution to the good air quality of Beijing. Thus, we call on the government and society to pay more attention to the biomass-burning pollution in rural and suburban, rather than solely focusing on emissions from vehicles and industry in urban region.

4. Conclusion

Source apportionment of different carbon fractions in $PM_{2.5}$ collected in Beijing before, during, and after Asia-Pacific Economic Cooperation (APEC) summit were constrained using radiocarbon (^{14}C) measurement in this study. Before the APEC summit, the average concentration of water-soluble organic carbon (WSOC), water-insoluble OC (WIOC), and elemental carbon (EC) were 6.95 $\mu g\,C/m^3$, 7.38 $\mu g\,C/m^3$, and 2.27 $\mu g\,C/m^3$, respectively. These values decreased to 3.33 $\mu g\,C/m^3$, 3.33 $\mu g\,C/m^3$, and 0.76 $\mu g\,C/m^3$, respectively, during the APEC summit, showing the evidence that the control measures on the emission sources during the APEC period were very effective. The ^{14}C measurements showed that 60–70% of fossil sources and 50–60% of nonfossil sources were reduced during the APEC summit relative to that before it. After the APEC summit, the WIOC ^{14}C level decreased due to the coal combustion for central heating in Beijing.

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