The Changes of $PM_{2.5}$, BC, Elements and Pollution Sources Using Nuclear Technology in Xinzhen, Beijing Over the Past Decade

Junkai Yang
China Institute of Atomic Energy
Yonggang Yao
China Institute of Atomic Energy
Hui Zhang
National Institute of Metrology PR China
Yangmei Zhang
China Academy of Meteorological Sciences
Caijin Xiao (cjshaw@ciae.ac.cn)
China Institute of Atomic Energy
Bangfa Ni
China Institute of Atomic Energy

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Abstract

The concentrations of $PM_{2.5}$, black carbon (BC), elements as well as sources of pollution in Beijing from 2003 to 2018 were investigated. The results show that the concentrations of $PM_{2.5}$ and BC had similar annual and seasonal trends, especially in autumn-winter, which with declining trends in recent years. The proportion of BC in $PM_{2.5}$ reduced from 13% in 2013(max) to 8.5% in 2018(min), indicating the reducing measure of replacing coal boiler with gas boilers worked well. In this study, annual trends of 15 elements were also discussed, it is found that the concentrations of S, K, Mn, Ca, Pb, Cu, Mn and Fe displayed remarkable decrease these years. Br, Zn and Cl was growing overall and Cl was more concentrated in $PM_{2.5}$ in autumn and winter. Moreover, the In detected by NAA with high sensitivity may be a new and crucial fingerprint element associated with coal combustion, industry emission or biomass combustion because of correlation with BC, Na, K, Cu and halogen elements well. Finally, 6-factor solution was identified during 2007 and 2016-2017 by EPA PMF, and the proportions of some pollution sources changed a lot in $PM_{2.5}$. Soil management in north China reduced the soil and dust source by 9.2%; The contribution of Industry-S or secondary S showed decrease from 27.5% to 22.5% due to industries relocating , gasoline with sulfur optimization and coal burning restriction; Banning straw burning and waste incineration in 2007 kept biomass and waste combustion out gradually. However, initial stage of some policies maybe the main reasons for a small increase of coal combustion source despite some steps taken.

1. Instruction

$PM_{2.5}$ (fine particulate matter, the aerodynamic diameter ≤2.5 μm) has an adverse effect on ecosystem and human health. There was a notably positive correlation between the concentration of pollutants and human mortality and morbidity(Atkinson, et al., 2014). Atmospheric particles with many harmful pollutants and heavy metal elements are non-degradability, bioavailability, toxicity persistence, etc. and oxidative stress usually causes inflammations, cardiopulmonary diseases, lung cancers and other diseases. (Limbeck et al.,2012, Wang et al., 2005, Bhatt, et al., 2013.) According to the report issued by World Health Organization(WHO) in 2013, for long-term exposure in heavy pollution, every 10μg/m^3 increase of $PM_{2.5}$ will result in a 6% and 11% increase of all diseases incidence and cardiovascular diseases, respectively. And for short-term exposure, an increase of 10μg/m^3 were associated with a relative of increase 1.05% in daily mortality rate.(Squizzato et al., 2018). In addition, SO_4^{2-}, BC and organic aerosol have direct or indirect effects on global climate such as frequent El Nino Phenomenon and global warming.(Chuang, et al., 1997)

As the capital of China, the API (air pollution index) has been at a high level over the past two decades in Beijing. For the sake of mitigate haze and environmental problems, Chinese and Beijing governments have taken scores of measures. Earlier in this decade, leaded gasoline was completely prohibited(Xia et al.,1999). From 2007 onwards, the government began to restrict the traffic flow in street. National III gasoline with sulfur content less than 0.015% was abolished after 2013 and national IV and V gasoline

(GB 18352.5-2005, GB 18352.5-2013, Wu et al., 2013)was promoted. Heavy pollution companies were relocated and closed gradually from 2014. Furthermore, coal heating has been replaced by gas heating in Beijing since 2017. Some achievements have been made these years throughout these regulations and policies. In 2017, the percentage of fine days in Beijing rose 7.8% compared with 2016, occupying 61.9% of the total. The average concentration of $PM_{2.5}$ was 58μg/m^3, which was 35% lower than that in 2013(http://www.cnki.com.cn/Article/CJFDTotal-HJJI201811008.htm). However, the figure still exceeded the new national secondary standard, i.e. 35μg/m^3 (the primary standard is less than 15μg/m^3) (Ban, 2012)

Last century, atmospheric pollution and source apportionment were mainly promoted by XRF(X-ray Fluorescence Analysis) and other nuclear analytic techniques, especially NAA and PIXE(Yang, et al., 2019). Liu et al., obtained concentrations of 41 elements and their trends such as In, Cl, Cu and As by NAA at three sampling sites in Beijing(Liu, et al.,2005). Jin, et al and Liu, et al determined 6 factors using PIXE including coal-fired, automobile exhaust, chlorine industrial sources, buildings, dust and secondary sulfur sources in Xinzhen area(Jin, et al.,2014, Liu, et al., 2018). In this work, we combined our historical data with recent results to present the trends of $PM_{2.5}$(from 2003 to 2018), BC (from 2007 to 2018) and various elements(from 2007 to 2017) in Liangxiang, Beijing. And EPA PMF was used to understand the pollution sources change in 2007 and 2016 ~2017.

2. Sampling And Experiment

2.1 sampling

The sampling site was located in Fangshan District in Beijing, between North China Plain and Taihang Mountains, bordered by Hebei Province to the southeast. There are mountains and hills on the western and northern sides, while the eastern and southern sides are plains; it is about 150 km to the Pacific, covering an area of 1990 square kilometers, with a population of 1.29 million in 2019 (http://www.bjfsh.gov.cn/zjfs/fsgk/201810/t20181018_199972.shtml)and influenced by temperate monsoon climate. Generally, spring is from March to May, summer is from June to August, autumn is from September to November and winter is from December to February of the second year.

The sampling site was situated on the roof of four-floor Building(116.184867E,39.719024N). It was close to living area, 500m away from a main highway, 12 km away from a petrochemical plant and about 40km away from center of city, which is an open and representative sampling position.

The Gent SFU (Stacked Filter Unit, Gent University) cascade sampler was used in this work(from 2003 to 2018). Two polycarbonate nuclear membranes of different pore size were put in the sampler to collect $PM_{2.5}$ and PM_{10}, respectively. Sampling time was from 8:00 to 18:00 ever sampling day(twice or third a week and more times could be employed in pollution days), the initial flow of 18 L/min and the termination flow of 14 L/min were used to obtain the ideal particles and results according to the requirement of Gent SFU.

2.2 Experiment

2.2.1 $PM_{2.5}$, BC and Elements

To meet the standard of temperature-humidity balance, collected filters were put in a balance chamber with temperature of 25±5°C and humidity of 50%±5% 24h before weighing. Moreover, filters were weighed strictly according to national standard HJ 618-2011(Determination of atmospheric articles PM_{10} and $PM_{2.5}$ in ambient air by gravimetric method, China), using MT5 ultra-micro balance (Mettler-Toledo Company, Switzerland). Meteorological data of this region including weather conditions, air pollution index, wind speed, wind direction, temperature, visibility and so on rigidly recorded based on China Weather Network (http://www.weather.com.cn/weather/101010100.shtml).

The BC concentrations from December 2007 to January 2018 were determined by EEL43D Black Carbon Meter (Diffusion Systems Ltd, UK). Its mathematical formula(equation 1) is as followed(Liu, et al., 2018) :

$$\mathrm{BC}_R(\mu g/cm^2) = [100/(2\varepsilon F)]\ln(R_0/R) \qquad (1)$$

more details about analyzing and calculation of BC can be found at Liu, et al.(2018).

The samples from 2016 to 2017 were analyzed by PIXE (GIC 417 tandem accelerator, Beijing Normal University). The proton energy was 2.5 MeV, the beam intensity was 0.1-20 nA, and the beam spot area was about 20 mm^2. The reference materials, a series of thin film standard samples(MicroMatter, Canada, the influence of sample matrix composition can be neglected) were used to meet the standard of quality assurance(Table S1), and the Si (Li) detector was used in this work. The resolution of K-alpha characteristic X-rays (corresponding energy 5.9 keV) of Mn was 159eV(Yu, et al.,2010). There were 15 elements ,including Mg, Al, Si, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, Br and Pb, were performed by GUPIXWIN software.

2.2.2 PMF

Positive Matrix Factorization (PMF) is a source unknown receptor model widely used in the analysis of air pollution sources, which was published by Paatero in 1997. After continuous improvement by Hopke and Paatero, EPA PMF software was launched on EPA (Environmental Protection Agency, EPA) website in the United States(Zhu et al,2012). This work used EPA PMF5.0 source analysis software, which was divided into PMF 2 and ME-2(Reff et al., 2007).

All of elements statistics were showed in Table S1, and S/N≥2 of species were considered as “strong”, while the species with 2≥S/N≥0 were regarded as “weak”. In this study, the uncertainty of elements were calculated as Error+MDL/3, where the MDL is the detection limit. The concentrations below MDL were set as 5*MDL/6 (Wang et al.,2018).The final solution depended on several standards, including: (1) The result of source profile(2) Temporal trends for each factor in the winter and non-winter(Because pollution is most serious in winter in Beijing) (3)Three error estimations. For Bootstrap(BS),100 runs were set and the ratio of mapping was more than 80%. For displacement (DISP), decrease of Q was <1% and no swap was observed for dQmax=4. For BS-DISP, if the decrease of Q was0.5%, we could think the result was stable(Norris et al., 2014).

3. Results And Discussion

3.1. $PM_{2.5}$ and Black Carbon

The temporal trends of BC and $PM_{2.5}$ were showed in Fig.1, the annual mean of $PM_{2.5}$ (Fig.1(a) red line) firstly increased(2003-2015) and then decreased(2015-2018) over the past 16 years. From 2003 to 2015, consistent increase of $PM_{2.5}$ was caused by rapid urbanization, population explosion and increase in the number of private cars. According to statistics, in 2018, the car ownership in Beijing reached 6.08million, 2.85 times as much as that in 2005 (https://baijiahao.baidu.com/s?id=1643270530939456964&wfr=spider&for=pc) Moreover, in 2017 there were 21.71 million people in Beijing compared with 14.56 million in 2003, increased by 7.15 million (http://www.stats.gov.cn/). While many effective environmental protection policies and laws including coal-to-gas heating, relocation of heavy pollution industries and promotion of electric vehicles etc. reduced the concentration of $PM_{2.5}$ evidently in last several years. Autumn and winter can cover the whole heating season, usually from November to March of next year, in Beijing, so spring-summer and autumn-winter were divided into groups respectively to reveal more details. From Fig1.(b), $PM_{2.5}$ was much higher in autumn-winter than that in spring-summer except 2007, which was caused by the influence of bad atmospheric conditions(such as atmospheric circulation, sunshine radiation and dry weather)(Jin., 2014) ,coal burning, vehicle emissions and other pollution sources. The concentrations of autumn-winter declined year by year from 2015, also indicating there were achievements of the above policies.

The measurement of BC concentration in samples has begun since 2007. It is no secret that BC is the significant composition of $PM_{2.5}$ and mainly originate from coal combustion, vehicle emission or biomass burning(Mousavi, et al., 2019). The mean concentrations of BC(blue line in Fig.2(a)) also consistently increased from 2007, peaking in 2015(8120ng/m^3), and decreased from 2015 to 2018. Like $PM_{2.5}$ seasonal trends, the concentrations of BC were more severe in autumn-winter(Fig.1 (b)). From 2007 to 2015, there was an increase trend in the two seasons, and it also showed a decrease trend between 2015 and 2018. BC had a strong correlation with $PM_{2.5}$ (Fig.2.(b), R^2=0.676,slope=0.0718), occupying approximate 11% in total $PM_{2.5}$ in Liangxiang. However, in recent three years, its proportion in fine particle has shifted down significantly, and this figure was 8.5% in 2018, which may be due to cut down the capacity of coal burning and restrict some pollution industries by the government.

3.2 Common elements

Six years, 2007, 2009, 2013, 2014, 2016, 2017, has been selected because the elements' data of other years from 2007 to 2017 were incomplete or some samples were not analyzed. In Figure.3(a) and Figure S1. the soil and crustal elements like Si, Mg and Al had similar temporal trends, and the frequency of dust weather has decreased compared with before in recent years. The K is one of the fingerprint elements about biomass (Zheng et al., 2004), a continuous decrease of concentration showed the reduction of biological emission. S, Mn and Fe had the similar temporal trends, indicating that steel and heavy industries developed rapidly before 2013, and then they were controlled effectively in these years. Shougang Group, for example, had stopped production, and relocated in Hebei province in 2011. The of concentrations for Pb, Zn and Br had the overall upward trends, while the obvious decline trends in 2017 might be well caused by the coal to gas policy and pollution industries management. Cl had a clear upward trend and seasonal variations, the concentrations in autumn-winter were much higher than it in spring-summer. The reasons of aggregation of Cl in autumn and winter are as followed: Firstly, heating in winter caused a large number of coal burning in Beijing, which is the main source of Cl (Yu. et al.,2018.); secondly, most chemical compound with Cl is volatile in high temperature, while the environment conditions make it aggregation easily in winter and autumn. In addition, there were no clear law about Cr and Ni. Further study will be done in the next section of PMF. All measured elements and BC these years accounted for 30%-40% in total $PM_{2.5}$(showed in Fig. 3(b)), which was similar with the percentage(37%) in Tianjin, a city bordering on Beijing. Therefore, in most cases, inorganic salts (NO_3^-, SO_4^{2-}, NH_3^+ etc.), organic matter and other secondary pollutants may be the main component in $PM_{2.5}$.

3.3 Element In

70 samples from 2017.1 to 2018.6 were analyzed by NAA(Neutron Activation Analysis)(Kothai et al., 2008, D.D.Lestiani et al., 2011, Abel, et al., 2011.). The parameters of NAA and statistical data of various elements were showed in Table S2 and Table 1. One element, In, was special and had a strong relevance with other elements. According to the results, though the In was extremely slight content in $PM_{2.5}$, it had a good correlation, about 0.797, with the fine particles. Moreover, the In correlated with BC, and halogenated elements contents in $PM_{2.5}$, like Cl, Br and I obviously($R^2_{In\text{-}BC}$=0.879, $R^2_{In\text{-}Cl}$=0.399, $R^2_{In\text{-}Br}$=0.796, $R^2_{In\text{-}I}$=0.775), while there was little relevance between In and soil elements($R^2_{In\text{-}Dy}$= 0.074, $R^2_{In\text{-}Al}$=-0.138, $R^2_{In\text{-}Ti}$=0.134), explaining it maybe originate from combustion of fossil fuel or vehicle emission instead of crust or soil. In addition, anode of battery or some electrical materials also dope a few In element to improve the performance(Yu., 2018, Fen., et al, 2013). Obvious correlation with Cu, Na and K($R^2_{In\text{-}Cu}$=0.761, $R^2_{In\text{-}Na}$=0.708, $R^2_{In\text{-}K}$=0.751) also displayed that biomass combustion could be one of the origins of element In(Taghvaee, et al., 2018, Santoso, et al., 2008). Therefore, it was possible that the In could be a fingerprint and semi-fingerprint element to identify some specific sources.

Table1 The statistical data of elements analysed by NAA

Species	Mean(ng/cm^3)	STDEV(ng/cm^3)	Max(ng/cm^3)	Min(ng/cm^3)	R^2 (with In)
$PM_{2.5}$	67955	50851	245568	563	0.797**
BC	5315	3296	14874	8584	0.879**
Dy	0.053	0.084	0.683	0.032	0.074
Ba	15.2	10.7	70.0	2.47	0.351*
Ti	52.4	77.6	608	7.38	0.134
I	5.46	5.07	28.6	0.538	0.775**
Br	11.2	13.1	75.3	1.13	0.796**
Mn	24.9	13.8	73.9	5.62	0.597**
Mg	307	305	2324	56.1	0.054
Cu	12.6	8.41	33.7	1.10	0.761**
In	0.032	0.023	0.105	0.002	1
Na	284	270	2158	45.4	0.708**
V	2.43	2.61	14.9	0.481	0.473**
K	621	451	3142	130	0.751**
Al	874	1433	11708	102	-0.138
Cl	104	80.7	433	34.1	0.399
Ca	829	542	3523	140	-0.204

Note: "*"indicates a significant correlation at the 0.05 level (both sides)

"**" indicates a significant correlation at the 0.01 level (both sides)

3.3 Source apportionment

PMF analysis further defined the pollution sources in 2007 and 2016~2017, because the same methods (PIXE, Beijing Normal University) were used in the two periods. A total of 17 species (BC, 15 elements and $PM_{2.5}$) were used as the EPA PMF 5.0 input matrix, and the statistical data were showed in Table. S3. In the end, six factors were chosen in both periods and the PMF predicted results showed quite well correlation with observed results (Fig.S2. R^2_{2007}=0.928 slope=1.0466, $R^2_{2016\text{-}2017}$=0.931 slope=1.355).

Fig.4. showed there were three common sources, i.e., soil and dust(19.7% and 10.5%), coal combustion(16.3% and 18.8%) and industry-S(27.5% and 22.5%) in both two periods. Mg, Al, Ti, Si and Ca were the characteristic elements in soil and surface dust(Yu et al.,2018.), and mainly came from long-

range transport such as Loess Plateau, Mongolia and Kazakhstan.(Huang et al., 2017). Some elements rich in dust, like Pb, does not simply reflect in soil and crust, as it could also originate from vehicle emission or nearby construction sites. This factor contributed 19.7% and 10.5% in total $PM_{2.5}$ in 2007 and 2016~2017, respectively, showing a significant decrease. A large-scale afforestation project, for example “the Green Wall of China”, has been carried out in the Loess Plateau in the past two decades(Jing, et al., 2021). The frequency of dust weather in Beijing has decreased from 13 every year in the beginning of the new century to 2-3 in recent years, indicating the great achievements of these soil conservation projects.

Coal combustion, determined by Cl and BC(Yu et al.,2018), explained 18.8% in 2016~2017, which was a small increase compared with 16.3% in 2007. The factor in 2007 were involved in a little percentage of Cu, Zn, Br and Pb, all of which were characteristic elements of metal industries, e.g., dye industries for Cu(Chuang. et al., 2016) and were co-discharged with coal burning. In northern China, a large number of coal would be consumed for heating in winter every year. The sampling site was quietly close to Hebei province, there was still large amounts of coal-burning during winter heating. Moreover, northern and eastern China are the main region of coal combustion emission(Jin et al., 2014), the regional transport of pollutants exerted a negative influence on Beijing. In addition, due to policy of coal-to-gas in 2017, some polluting factories and enterprises were still relocating or plan to relocate. These reasons may cause a slight jump in percentage of coal combustion source in 2016~2017 than it in 2007 and some metal elements were also disappeared these years.

The third common pollution source, industry-S, accounted for 27.5% and 22.5% in their own samples, respectively. Beijing is the center of Beijing-Tianjin-Hebei industrial area, industries or fuel burning emissions are the most important source of SO_2 and BC, and then these SO_2 were transformed into SO_4^{2-} or secondary sulfur by photochemistry in the air (Mang, et al, 2018). Fe, Zn, Br and Pb were also involved in this factor in 2007, indicating some metal industries pose a threat to the environment(Tao J., et al, 2014). There were some small factories and workshops in Xinzhen ten years ago. While the decrease of this factor percentage in 2016~2017 proved relocating and shutting down industries had a good effect on local air.

Besides that, three pollution sources of the two periods were different from each other. Steel manufactory and vehicle accounted for 18.9% in $PM_{2.5}$ in 2007. Zn, Br and BC were mainly emitted from gasoline and diesels and Cu may come from car tie wear (Zheng, et al., 2004, Pant, et al., 2018). Also, some elements of steel industries like Cr, Mn and Fe were involved in this factor. In 2016 ~ 2017, industry-Zn and vehicles(also determined by Br, BC, Cu and crustal elements) contributed 19.8% to $PM_{2.5}$. Despite as an additive in two-stroke engine lubricating oils(Yu. et al.,2018.), high percentage Zn also associated with some electronical companies and industries (Zhang et al., 2014). There were many high-tech companies, e.g. new energy, electronic or material companies, located in Fengtai science and technology park about 20km away from sampling site. Moreover steel industries was also a serious pollution source with a portion of 18.3% in this period and most of it may sperate from area transmission. Element K and some microelements like Zn, Cu, were characteristic tracer for biomass burning which contributed 15.5% to the $PM_{2.5}$ in 2007(Taghvaee, et al., 2018, Santoso, et al., 2008). One decade ago, there were many farmland in Beijing suburbs, especially in Xinzhen, tons of straws and waste burning in late autumn and winter was the main reason of this pollution source emission. But as the law of prohibition of straws burning implemented in 2007, the proportion of this factor would decrease year by year. Therefore, there was no obvious factor with high level of K in 2016~2017.

Cement plant source was mainly determined by Ca and contributed small percentage(2.2%) to $PM_{2.5}$ in 2007. There was a cement plant not far from the sampling point in the past, but it was demolished several years ago. In 2016 ~ 2017, new kindergarten was built and the student dormitory was redecorated around the sampling site during this period. Therefore, construction, which was identified by high level of Ca and some Mg, Al, Si, Ti, Fe and Cu in concrete and cement, accounted for a relatively larger proportion, about 10.1%.

4. Summary And Conclusion

The concentration of $PM_{2.5}$ showed a upward trend from 2003 to 2015 and a downward trend from 2015 to 2018, indicating the change of air situation in both periods with the rapid development of city and environment treatment. BC contributed approximate 11% to the total $PM_{2.5}$, and had a similar annual trend of concentration with it. Basically, $PM_{2.5}$ and BC were more intensive in autumn and winter. For the elements S, K, Mn, Ca, Pb, Cu, Mn and Fe showed decrease trends these years, while Cl, Zn and Br had increase trends in general, and the concentration of Cl tend to aggregate in autumn and winter. Element In, less content in $PM_{2.5}$ and insufficient research about it, had stronger correlations with $PM_{2.5}$, BC, Na, Cu, K and halogen elements than soil elements, so maybe it is a fingerprint or semi-fingerprint element for some pollution sources like fuel combustion, biomass burning or industries emission. These elements and BC constituted only 30%-40% of the total $PM_{2.5}$. Therefore, paying more attention to inorganic salts, organic matter and secondary pollutants is crucial to have a deeper insight to $PM_{2.5}$.

6 factors were identified by PMF in both periods(2007 and 2016~2017), showing that some policies, like banning straw and waste burning, soil management, relocating heavy pollution industries etc., had made great achievements in the past ten years. Compared with some sources in 2007, the proportion of soil and dust source fell close to10%; biomass and waste source even disappeared in 2016~2017. However, the proportion of steel manufactory source was larger than 10 years ago. According the data of car ownership, vehicle emission might be a serve source now, though the government had a restriction about cars and diesels. Continuing to control the heavy pollution industries and vehicle emissions is still crucial to improve the quality of local air. Moreover, regional transmission also need to be considered because Beijing is next to Heibei where is the province with many heavy pollution industries.

Declarations

Ethical Approval

Not applicable

Consent to Participate

Not applicable

Consent to Publish

Not applicable

Authors Contributions

Caijin Xiao, the corresponding-author, gave me the idea about this research. I(Junkai Yang) was primarily responsible to sample and write this article, Yonggang Yao and Caijin Xiao helped me to collect and analyze the samples. Yonggang Yao, Hui Zhang, Yangmei Zhang, Caijin Xiao and Bangfa Ni gave me many useful advice and guidance to complete my article.

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Conflict of interest

The authors declare that there is no conflict of interest.

Availability of data and materials

The datasets used or analysed during the current study are available from the corresponding author on reasonable request.

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Figures

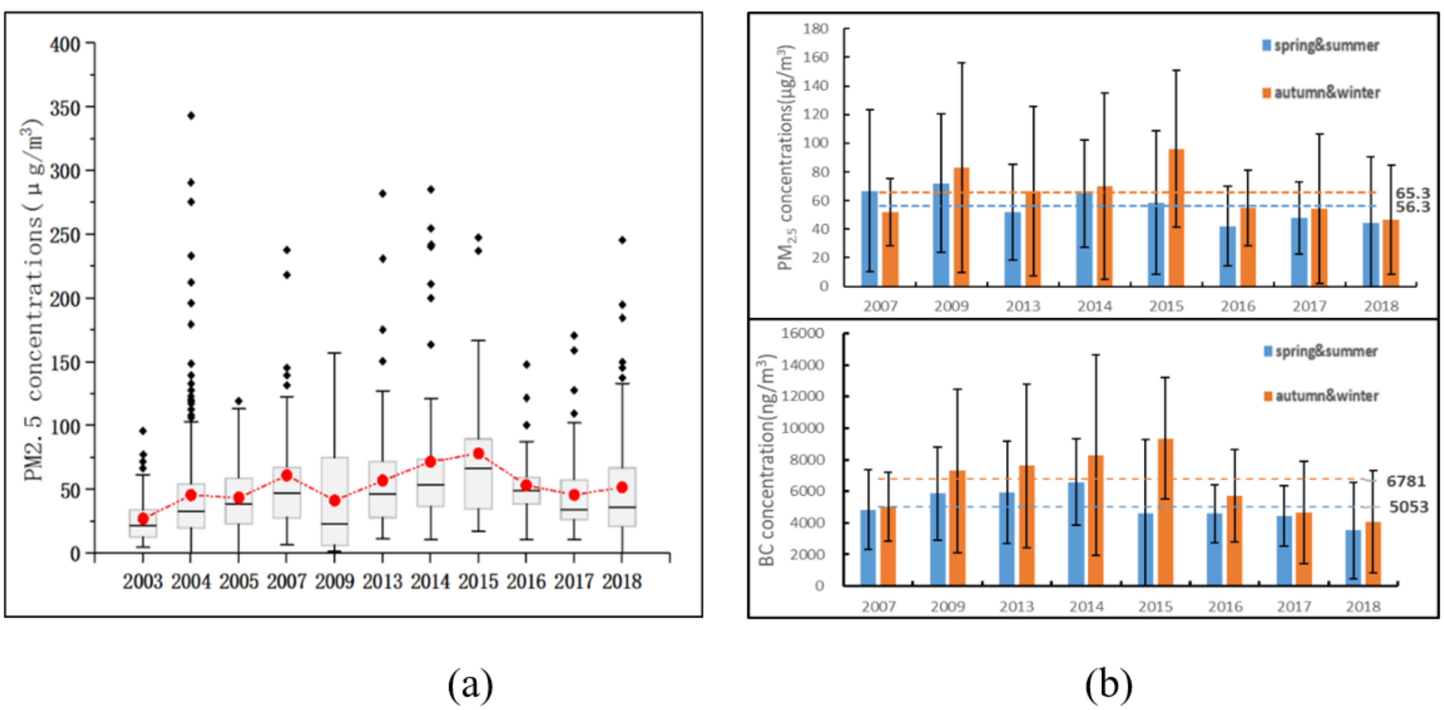


Figure 1

The concentration of PM2.5 and BC temporal trends: (a) Annual trend of PM2.5 (b) Seasonal trends of PM2.5 and BC(Horizontal lines were the average concentrations of PM2.5 and BC in different seasons)

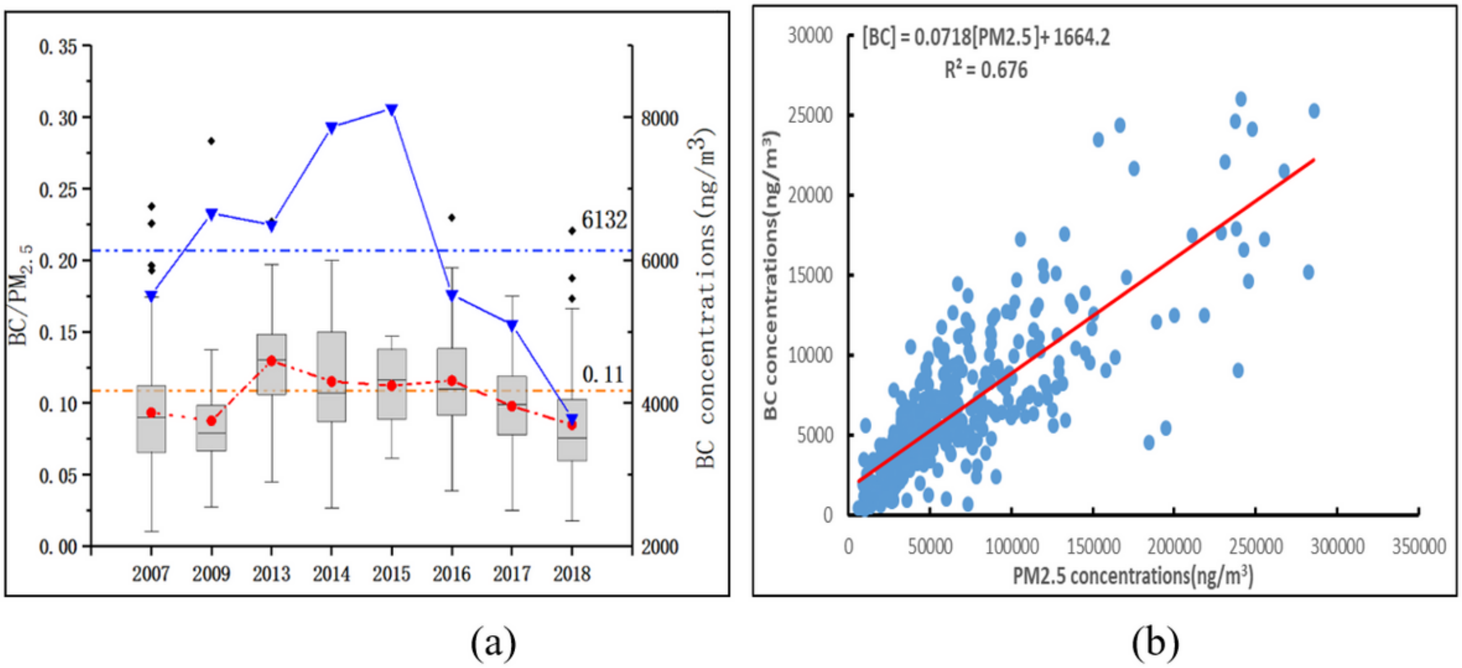
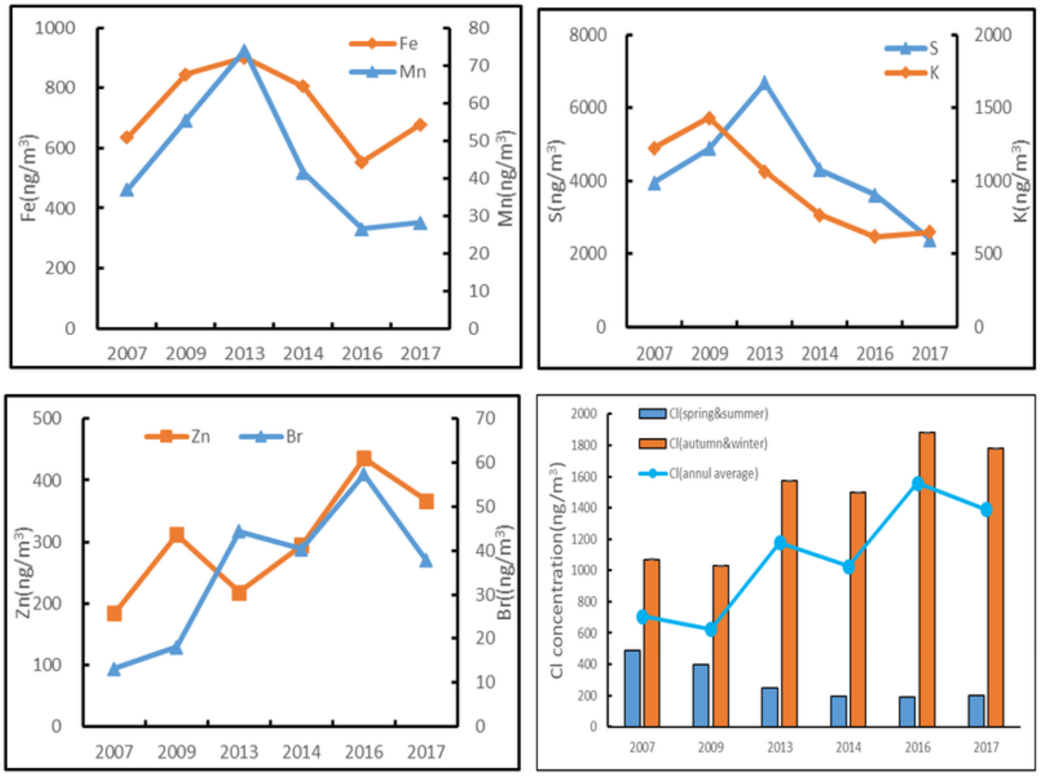
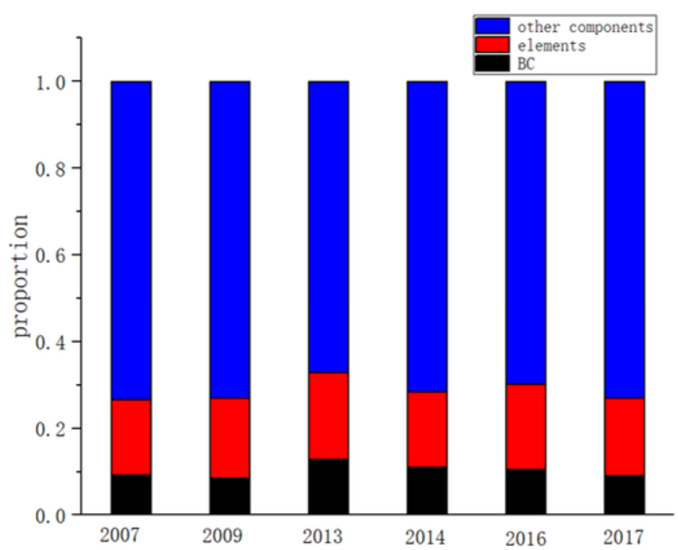


Figure 2

(a) BC annual trends from 2007 to 2018 (b) Correlation between BC and PM2.5



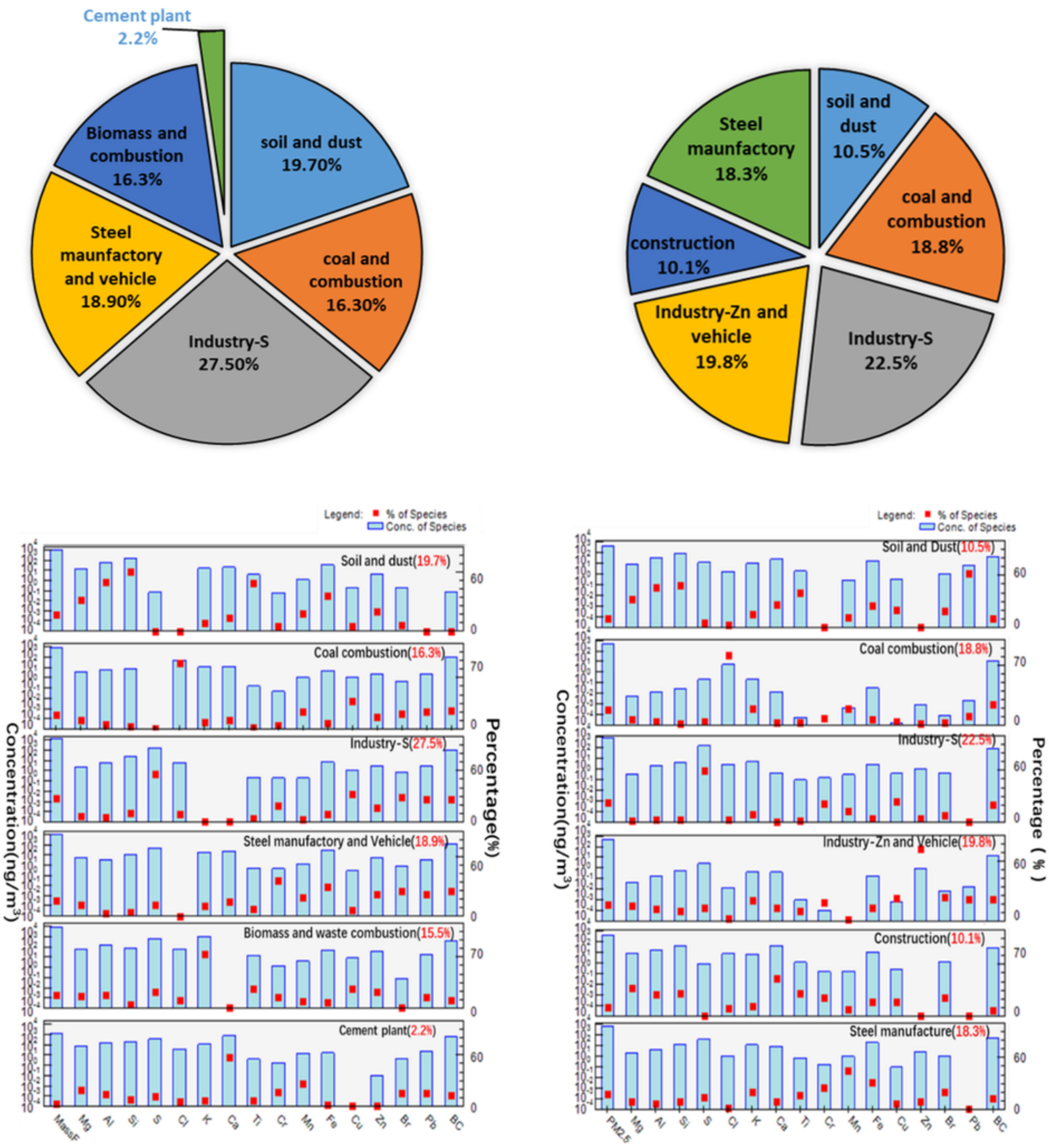
(a)



(b)

Figure 3

(a)The concentration of elements temporal trends. (b) The percentage of various components in PM2.5



(a)2007 (b) 2016-2017

Figure 4

Source apportionment(ng/m3) in :(a) 2007 (b) 2016-2017

Supplementary Files

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