ijar
ISSN NO. 2320-5407
Journal Homepage: - www.journalijar.com
INTERNATIONAL JOURNAL OF ADVANCED RESEARCH (IJAR)
Article DOI: 10.21474/IJAR01/8821
DOI URL: http://dx.doi.org/10.21474/IJAR01/8821

RESEARCH ARTICLE

CHEMICAL CHARACTERISTICS OF $PM_{2.5}$ DURING A TYPICAL SPRING FESTIVAL FIREWORKS DISPLAY IN RURAL AREAS IN WENZHOU, CHINA.

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Manuscript Info

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Manuscript History
Received: 05 February 2019
Final Accepted: 07 March 2019
Published: April 2019

Key words:-
$PM_{2.5}$; Water-Soluble Inorganic Ions; Trace Elements; Carbon Components.

Abstract

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To investigate the effect of fireworks on the quality of the atmospheric environment and changes in the chemical characteristics of fine particulate matter ($PM_{2.5}$), $PM_{2.5}$ samples were collected from rural areas in Wenzhou during the Spring Festival in 2018. Components loaded on $PM_{2.5}$ samples were analyzed, including water-soluble inorganic ions, organic carbon (OC), elemental carbon (EC), and trace elements. The $PM_{2.5}$ concentration during the fireworks period is twice that of normal times. Main components loaded on fine particles during the use of fireworks were K, NO_3^-, Cl^-, SO_4^{2-}, and OC, accounting for approximately half of the mass of $PM_{2.5}$. The ratio of total cations to total anions decreased, indicating that fireworks released a high amount of acidic substances. The concentrations of K^+, Mg^{2+}, Cl^-, NH_4^+, NO_3^- and SO_4^{2-} on $PM_{2.5}$ were significantly increased by 18.13, 8.25, 6.25, 4.97, 3.47 and 2.67 times, respectively, and the effect of fireworks was much higher on OC than on EC. PM released from fireworks was more harmful because the K, Cu, Pb, Cr, and Ni enrichment factor values of the samples obtained during the fireworks show increased by 9.36, 8.98, 7.40, 3.89, and 3.51 times, respectively.

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Introduction:-

Air pollution is considered a global environmental problem that poses a hazard to the comfort, health, and well-being of humans when pollutant concentrations or exposure durations reach a certain level. Atmospheric particulate matter (PM), which is one of the most important atmospheric pollutants, is regarded as a carcinogen by the International Agency for Research on Cancer (Loomis et al., 2013). Because of the rapid development of economy and industry, China has become the second largest economy in the world. Economic growth is accompanied by energy consumption, and the massive discharge of air pollutants has become one of China's most critical environmental issues (Meng et al., 2016). Studies have investigated the composition and source of fine particles by examining the chemical properties of fine PM ($PM_{2.5}$) samples from different regions and of different types (Zhang et al., 2017; Hao et al., 2018; Mi et al., 2018; Wen et al., 2018; Zhu et al., 2018). In recent years, many studies have focused on pollutants that cause short-term air quality degradation (Tsai et al., 2012; Zhang et al., 2015; Liu et al., 2016). Fireworks produce considerable amounts of gaseous and particulate contaminants, which can cause severe health hazards. Therefore, fireworks used in festivals are considered a crucial short-term anthropogenic source. Fireworks are used in popular festivals worldwide, including Chinese New Year's Eve, American Independence Day, France's Bastille Day, and India's Diwali. However, fireworks are one of the main sources of PM and

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precursors, such as carbon monoxide, nitrogen oxides, and sulfur dioxide, which can contribute approximately 30% to $PM_{2.5}$ (Tian et al., 2014). During the 2007 Montreal International Fireworks Competition, the highest $PM_{2.5}$ level reached nearly 10,000 ug m^{-3}, which is approximately 1000 times the background level (Joly et al., 2010). Sarkar et al. (2010) reported that fireworks can increase the levels of many harmful chemicals, including color-producing metals (e.g., Sr, Mg, Ba, K, Cu, and Pd) and oxidants (e.g., nitrates, perchlorates, and chlorates). The levels of NO_3^-, SO_4^{2-}, and Cl^- in the Lantern Festival were reported to be five times the normal levels (Wang et al., 2007). Cheng et al. (2014) showed that the K^+ level increased significantly by 4.97 times when fireworks were used in winter compared with when no fireworks were used. Fireworks release a large amount of fine PM that can be inhaled; thus, a comprehensive analysis of the chemical properties of fine particles present in fireworks is necessary.

Zhejiang is one of the wealthiest provinces in China. In this province, many fireworks and firecrackers are used every year, particularly in rural areas. The present study investigated the effects of fireworks on the quality of the atmospheric environment and the health of the population. $PM_{2.5}$ samples were collected from rural areas in Wenzhou during the Spring Festival in 2018. Particles and their components, namely water-soluble inorganic ions (WSIIs), organic carbon (OC), elemental carbon (EC), and trace elements (TEs), were analyzed to determine the effect of fireworks on the air quality of the rural areas.

Methods:-

Site description and sample collection

From February 11, 2018, to February 25, 2018, $PM_{2.5}$ samples were collected through sampling on the roof of a seven-story building (approximately 25 m above ground) in Wenzhou (28.07°N, 120.86°E) in eastern China (Fig. 1). The sampling point was located in a rural residential area with a busy street approximately 10 m to the south.

A quartz fiber filter with a diameter of 90 mm was baked at 450°C for 5 h to generate a $PM_{2.5}$ sampler (Tianhong, MA, China). Starting at 10 am on February 11, the sampling time for each sample was 24 h, and the flow rate was set at 100 L min^{-1}. Weighing filters before and after sampling need to be carried out at a constant temperature (20°C ± 3°C） and relative humidity (45% ± 5%) for 24 hours. All filters were wrapped in aluminum foil and stored in a refrigerator (−4°C) until analysis. Fireworks were found to exert a slight effect on samples collected on New Year's Eve. Thus, in this study, we hypothesized that the previous air features on February 15, 2018, could represent the winter of the sampling point.

Fig. 1 Location of the $PM_{2.5}$ sample point

Carbon component analysis

The concentrations of OC and EC were analyzed using a DRI-2001A thermal optical reflectance carbon analyzer (Model 2001A, USA) and the IMPROVE temperature program. First, in a fully anaerobic helium environment, the 0.52 cm^2 samples were sequentially heated at four temperatures 140°C(OC1), 280°C(OC2), 480°C(OC3), and 580°C(OC4); then, samples were heated in a 2% oxygen/helium atmosphere at 580°C(EC1), 740°C(EC2), and 840°C(EC3). The evolved carbonaceous gas was oxidized to CO2 by using a manganese dioxide catalyst and then reduced to methane, which was detected using a flame ionization detector. During heating, OC can be developed as pyrolytic carbon (OP). To correct for OC, light reflections were monitored during the reaction. After oxygen is added to the analytical atmosphere, the OP is determined when the reflected laser reaches its original intensity (Tang et al., 2016). OC and EC are calculated by the following formulas, respectively. OC=OC1+OC2+OC3+OC4+OP, EC=EC1+EC2+EC3-OP.

Water-soluble inorganic ion analysis

Four anions (F^-, Cl^-, NO_3^-, and SO_4^{2-}) and five cations (NH_4^+, Na^+, K^+, Ca^{2+}, and Mg^{2+}) were analyzed through ion chromatography (Dionex ICS-900, USA). First, one-eighth of a filter sample was cut and ultrasonically extracted with 10 mL of high-purity water (Millipore Milli-Q plus 185). Subsequently, the filtrate was passed through a membrane filter with a pore size of 0.22 μm. The anions were analyzed using a Dionex IPAS19 analytical column and a Dionex IPAG19 guard column. The cations were analyzed using a Dionex IPCS 12A analytical column and a Dionex IPCG 12A guard column. The anion eluent was 20 mM KOH, and the cation eluent was 20 mM methanesulfonic acid.

Trace element analysis

One-eighth of the sample filter was pulverized using ceramic scissors and added to a Teflon digestion tank. Then, 5 mL of concentrated HNO_3 and 2 mL of concentrated H_2O_2 were added, and the sample filter was finally digested at 170°C for 30 min in a microwave digestion instrument. After cooling, the filtrate was evaporated to 0.5–1 mL on a hot plate at 140°C and then diluted to 10 mL with 1% HNO_3. The concentrations of Ti, Ni, Co, Pb, Mn, Cr, Fe, Zn, K, Mg, Ca, Al, Na, and Cu were measured using inductively coupled plasma atomic emission spectroscopy (ICP-AES, JOBIN-YVON Company, France).

Results:-

Meteorological conditions and mass concentrations of particles

Meteorological conditions, namely relative humidity (%), temperature (°C), and wind speed (m s^{-1}), during the sampling period are shown in Fig. 2. During sampling, the wind speed was lower (≤ 2 m s^{-1}) and relative humidity was higher (>60%), which favored the accumulation of PM and the observation of atmospheric pollution at the sampling site (Deshmukh et al., 2011).

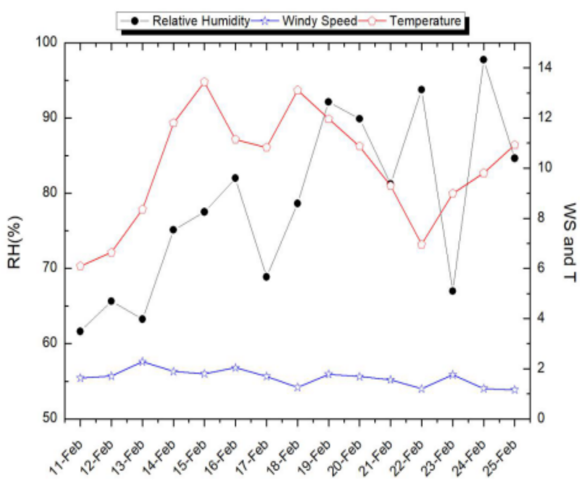

Fig. 2 Meteorological parameters in Wenzhou

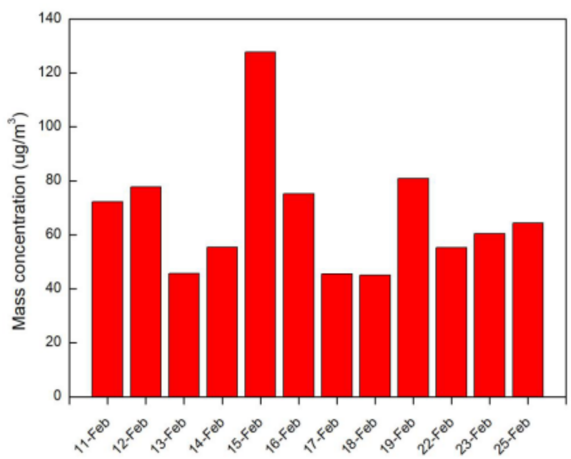

Fig. 3 Mass concentrations of $PM_{2.5}$ in Wenzhou during the Spring Festival

The mass concentrations of $PM_{2.5}$ during sampling are shown in Fig. 3. The concentration of $PM_{2.5}$ ranged from 25 to 127 μg m^{-3}, with an average concentration of 59 μg m^{-3}, and the highest concentration was observed on February 15 (Lunar New Year's Eve). The $PM_{2.5}$ concentration on February 15 was twice that on February 14 and then declined rapidly. According to anthropogenic activities, the sampling period in this study could be divided into three periods: (1)February 11–14 (4 days): this period (BNY) was before the Chinese New Year. During the During the BNY period, no fireworks were used, and the weather was fine without rain. The average mass concentration of $PM_{2.5}$ in winter is 55.9 ug m^{-3}, which is close to the average of 62.7 ug m^{-3} in the period (BNY).Therefore, the air condition during this period can represent the winter air condition in rural areas of Wenzhou; (2) February 15 (1 day): during this period (CNY), a considerable amount of fireworks were used to celebrate the Chinese New Year; and (3) February 16–25 (7 days): during this period (ANY), no fireworks were used. In the ANY period, no samples were collected on February 20, 21, and 24 because of rain.

The average concentrations of $PM_{2.5}$ for the three periods are listed in Table 1. The PM concentration was the highest during the CNY period, indicating that fireworks had a significant effect on the quality of the atmospheric environment (Zhang et al., 2010). In addition, the $PM_{2.5}$ concentration in the BNY period was slightly higher than that in the ANY period, indicating that industrial emissions had a certain impact on the quality of the atmospheric environment. Although the average $PM_{2.5}$ concentration during the BNY or ANY period was lower than China's air quality standard of 75 ug m^{-3}, that value was still higher than the US Environmental Protection Agency (USEPA) and WHO's air quality standard of 35 ug m^{-3}.

Water-soluble inorganic ions

The average mass concentrations of WSIIs in rural areas in Wenzhou are listed in Table 1. In $PM_{2.5}$, the mass concentrations of total WSIIs (TWSIIs) ranged from 15.90 to 56.46 μg m^{-3}, accounting for 24%–44% of $PM_{2.5}$ mass. Secondary water-soluble ions, mainly NH_4^+, SO_4^{2-}, and NO_3^-, accounted for 48%–75% of TWSIIs; these levels are lower than their corresponding levels in urban areas such as Tianjin (78%)(Gu et al., 2011), Shanghai (81%) (Zhou et al., 2016), and Beijing (83%) (Zhang et al., 2016). This difference might be due to the high sea salt content in coastal areas and particularly the contribution of Na^+ to TWSIIs (from 10% to 29%). The findings of USEPA'S backward trajectory analysis (Fig. 4) showed that air parcels arriving in Wenzhou were mainly from the East China Sea and might carry many sea salt components.

Table 1 Average concentrations of OC, EC, and ions in $PM_{2.5}$ during the three periods in rural areas in Wenzhou.

Compound	BNY	CNY	ANY
$PM_{2.5}$	62.79±12.84	127.76	60.98±12.75
Na^+	3.74±0.05	5.56	3.11±0.85
NH_4^+	0.65±0.28	3.23	1.01±0.51
K^+	0.56±0.12	10.15	1.20±0.52
Mg^{2+}	0.16±0.03	1.32	0.21±0.07
Ca^{2+}	0.58±0.07	0.91	0.44±0.14
F^-	0.05±0.02	0.12	0.04±0.01
Cl^-	1.38±0.44	8.63	1.70±0.70
NO_3^-	3.79±1.03	13.18	5.79±2.23
SO_4^{2-}	5.00±1.63	13.37	8.54±1.61
OC	4.52±1.05	8.95	4.86±1.43
EC	1.75±0.37	1.83	1.40±0.40
OC/EC	2.60±0.52	4.90	3.65±3.65

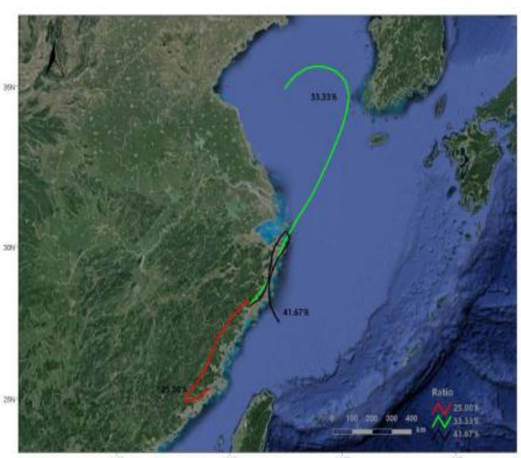

Fig. 4 Backward trajectory analysis in CNY

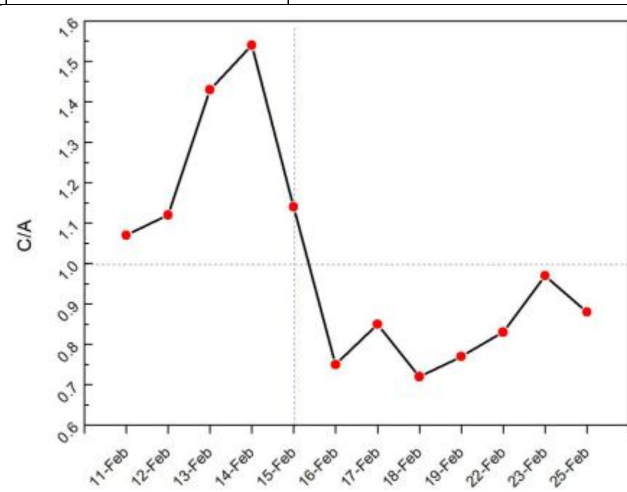

Fig. 5 C/A ratios in Wenzhou

In this study, the acidity and alkalinity of $PM_{2.5}$ were evaluated by calculating the ratio of total cations to total anions (C/A ratio) by using the following equation:

$$C/A = \frac{Na^+/23 + NH_4^+/18 + K^+/39 + Mg^{2+}/12 + Ca^{2+}/20}{Cl^-/35.5 + NO_3^-/62 + SO_4^{2-}/48}$$

Fig. 5 shows C/A ratios during all the sampling periods. Prior to Chinese New Year's Eve, the average C/A ratio was greater than 1 (range: 1.1–1.5), indicating that $PM_{2.5}$ was alkaline, and these results are consistent with C/A ratios reported for other cities in China, including Beijing (1.14) (Zhang et al., 2016), Shanghai (1.1) (Feng et al., 2012), Jinan (1.16) (Zhang et al., 2014), and Tianjin (1.35) (Gu et al., 2011). After Chinese New Year's Eve, the C/A ratio decreased significantly to less than 1, indicating that fireworks released a large amount of acidic substances.

A comparison of the chemical compositions of $PM_{2.5}$ in the three periods (BNY, CNY, and ANY) facilitated the understanding of the pollutant emission characteristics of fireworks. Fig. 6 shows the concentrations of the nine WSIIs measured during the three periods. As shown in Fig. 6, the concentrations of NH_4^+, K^+, Cl^-, NO_3^-, and SO_4^{2-} increased sharply when fireworks were used and then decreased rapidly, indicating that fireworks explosions a significant effect on the increase in these WSIIs. During the CNY period, K^+ showed the largest increase among WSIIs, and the K^+ concentration on New Year's Eve was 18.13 times higher than that on the previous day. Compared with the BNY period, the concentrations of Cl^- and NO_3^- during the CNY period increased by 6.25 and 3.47 times, respectively, indicating that fireworks released large amounts of Cl^- and NO_3^-. The significant increase in the concentrations of K^+, Cl^-, and NO_3^{2-} during the CNY period might because $KClO_4$, KNO_3, and $KClO_3$ are the main components of fireworks (Wang et al., 2007). In addition, the concentrations of SO_4^{2-}, Mg^{2+}, and NH_4^+ were increased by 2.67, 8.25 and 4.97 times, respectively. Moreno et al. (2007) proposed that the increase in the SO_4^{2-} concentration is related to the orange flame, and Mg is one of the essential components of fireworks because the combustion of magnesium powder releases strong light. In this study, the increased concentration of K^+ Wenzhou was much higher than that in Tianjin (5.78 times) (Tian et al., 2014) and Kaohsiung (2.99 times) (Tsai et al., 2012), and this might be related to the large amount of fireworks used in Wenzhou. These findings indicate that fireworks contribute significantly to air pollution, and that fireworks should be subject to more stringent control.

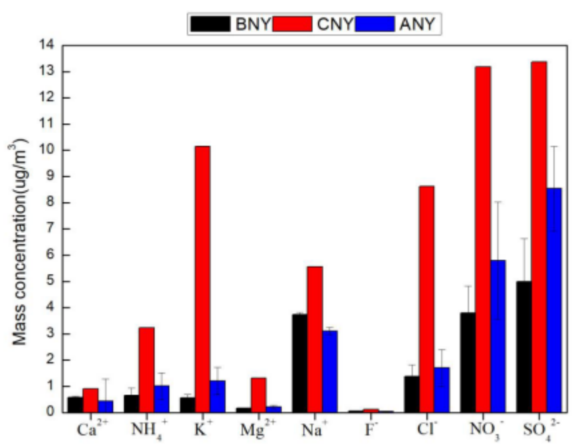

Fig. 6 Average concentration of nine types of ions in Wenzhou during the three periods

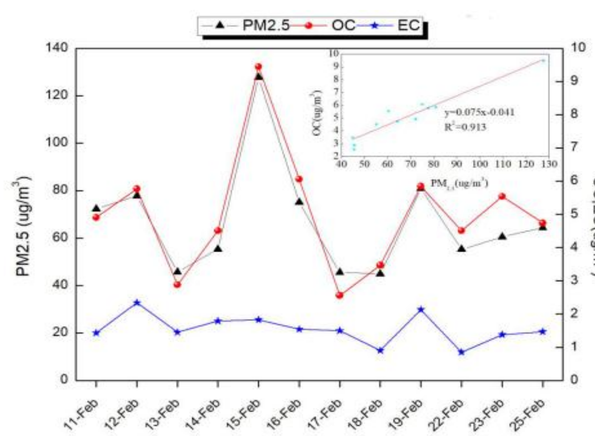

Fig. 7. Mass concentrations of $PM_{2.5}$, OC, and EC in Wenzhou.

Carbonaceous matters

During the BNY period, the average mass concentrations of OC and EC were 4.52 and 1.75 μg m^{-3}, respectively (Table 1). Although these mass concentrations are lower than those reported in megacities, such as Tianjin (12.9 μg m^{-3} for OC and 3.4 μg m^{-3} for EC)(Liu et al., 2016) and Beijing (14.0 μg m^{-3} for OC and 4.1 μg m^{-3} for EC) (Ji et al., 2016), they are higher than the values reported in rural areas in other countries, such as Czech Republic (3.96 μg m^{-3} for OC and 1.17 μg m^{-3} for EC) (Vodicka et al., 2015) and Gwangju in Korea (3.80 μg m^{-3} for OC and 1.0 μg m^{-3} for EC) (Park et al., 2018). Thus, carbon aerosol pollution in the rural areas of Wenzhou was severe.

The concentrations of OC and EC are shown in Fig. 7. During all the sampling periods, the OC concentration was significantly affected by the $PM_{2.5}$ concentration. The R^2 value was 0.914, indicating that OC had a strong correlation with $PM_{2.5}$ (Fig. 7). The mass concentration of OC was higher when fireworks were used than when no fireworks were used (Table 1), indicating that fireworks exerted a direct effect on the increase in the OC concentration.

To assess the effect of fireworks on the concentrations of OC and EC in $PM_{2.5}$, the growth rates of OC and EC were calculated. The growth rate of OC was 2 times and that of EC was only 1.05 times, indicating that the effect of fireworks on OC was much higher than that on EC. A similar study reported that OC and EC concentrations increased by 7 and 2.9 times, respectively, in Bhilai, India (Pervez et al., 2016). However, another study reported that OC and EC concentrations increased by 1.7 and 3.1 times, respectively, in Milan, Italy (Vecchi et al., 2008). The OC/EC ratio on February 15 was 4.9, which is significantly higher than that on a normal day (during the BNY or ANY period), indicating that fireworks are a specific air pollution source in contrast to conventional air pollution sources, such as coal-fired and mobile sources.

Trace elements

In this study, the concentrations of TEs during the BNY period, which can be regarded as the background concentrations of TEs in rural areas in Wenzhou, are listed in Table 2. Na, Ca, K, and Mg were found to be the most abundant elements among the 15 tested TEs, and their proportions were 49%, 14%, 12%, and 9%, respectively. The concentrations of Na, Ca, and Mg in Wenzhou exceeds those in inland cities such as Jinan, Beijing, and Zhengzhou (Yang et al., 2012; Geng et al., 2013; Yu, 2013). The mean concentration of the 15 measured TEs during the three periods is shown in Fig. 8. The total mass concentration of TEs in the CNY period was three times higher than that in the BNY period, suggesting that fireworks had a considerable effect on TEs. The concentrations of Al, Mg, K, Fe, Ni, Cr, Mn, Cu, Zn, and Pb increased sharply during the CNY period and then decreased rapidly, indicating that these elements were directly associated with fireworks emissions. K, Na, and Cu produce purple, yellow, and green flames, respectively. Cu, K, and Cr can also emit silvery and glittery colors. Cr in the form of $CuCr_2O_4$ acts as a catalyst for propellants. Magnesium–aluminum alloys have been reported to be used as reducing agents and illuminants in fireworks (Vecchi et al., 2008). Among the 15 TEs, the concentration of K showed the highest increase. K accumulation on $PM_{2.5}$ particles was 25 times greater during the CNY period than during the pre-fireworks period; thus, K can be used as a tracer for fireworks in the future.

Table 2 Average concentrations of TEs measured during the BNY period in Wenzhou and in Jinan, Beijing, and Zhengzhou in winter (μg/m^3) (*, ng/m^3).

	Wenzhou	Jinan	Beijing	Zhengzhou
Fe	0.38±0.11	2.02	1.051	1.551
Zn	0.59±0.25	0.99	0.332	0.537
K	1.33±0.05	4.58	2.76	1.53
Mg	1.04±0.10	0.18	0.096	0.384
Ca	1.56±0.07	1.09	0.703	1.039
Al	0.50±0.15	0.78	0.412	0.682
Na	5.35±0.27	0.63	-	0.139
Cu*	16.53±8.64	50	38	29
Ni*	8.11±1.56	10	28	4
Co*	5.68±0.35	20	-	1
Pb*	20.43±4.49	430	112	142
Mn*	18.53±2.50	160	74	137
Cr*	3.90±0.60	30	25	18
V*	7.96±0.24	-	18	5
Ti*	7.53±4.05	90	31	-

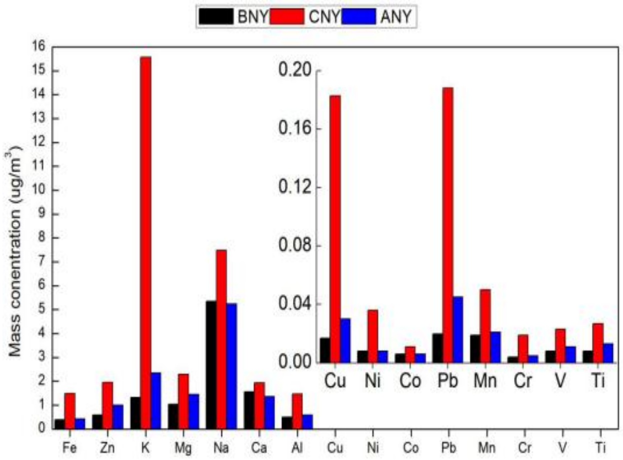

Fig. 8 Average mass concentrations of measured TEs at Wenzhou during three periods.

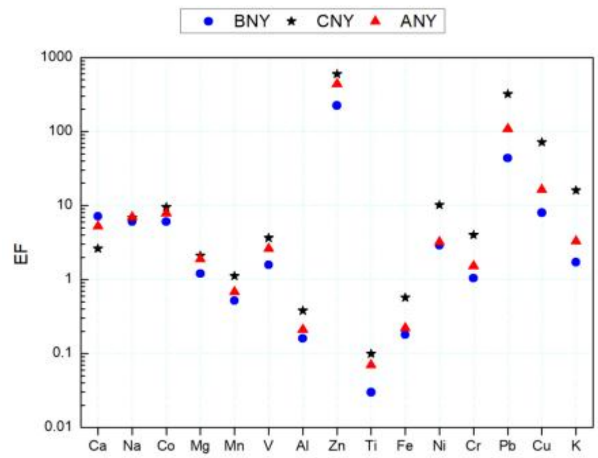

Fig. 9 Average EF value of 15 TEs during the three periods at the sample site

Enrichment factors (EFs), which indicate the degree of enrichment of elements in atmospheric particles, can be used to determine and evaluate the source (natural or artificial) of elements in particles as follows (Hieu and Lee, 2010; Rohra et al., 2018):

$$EFs = \frac{(M/R)\ \text{particulat es}}{(M/R)\ \text{crust}}$$

where (M/R) particles represent the mass concentration ratio of a target element M and a reference element R, and the (M/R) subscript represents particles in PM samples or crustal material. When EF > 10, the sample is considered have a high content of the target element compared with the reference element; this phenomenon is considered to be caused by human activities. When EF = 1, the element may have originated mainly from the Earth's crust or soil. EF < 1 indicates minuscule concentrations of elements in aerosol (Zhang et al., 2017).

Fig. 9 shows the average EFs of the 15 TEs during various sampling periods. During the BNY period, the EFs of Fe, Al, Mn, and Ti were less than 1, indicating that these four elements naturally exist in the Earth's crust or soil; thus, small quantities of these elements were observed in aerosols. The EFs of K, Ca, Na, V, Mg, Ni, Cr, Cu, and Co did not exceed 10, indicating slight contamination. The EFs of Zn and Pb exceeded 500 and 300, respectively, indicating that Zn and Pb were mainly derived from various human activities, particularly from motor vehicles (Xu et al., 2013). During the CNY period, the EFs of K, Cu, Pb, Cr and Ni increased by 9.36, 8.98, 7.40, 3.89 and 3.51 times, respectively, suggesting that fireworks largely contributed to the emission of these elements. Heavy metals in $PM_{2.5}$, namely Pb, Cr, Cu, and Ni, were directly emitted from fireworks, causing potential harm to human health.

Conclusions:-

Twelve $PM_{2.5}$ samples were collected from rural areas in Wenzhou during the Spring Festival in 2018, and all samples were categorized and analyzed for their chemical composition including WSIIs, OC, EC, and TEs.
Fireworks considerably increased the $PM_{2.5}$ concentration and were an important source of pollution resulting from human activities. K, NO_3^-, Cl^-, SO_4^{2-}, and OC were the main components loaded on fine particles during the CNY period, accounting for approximately half of the mass of $PM_{2.5}$ particles.

The decrease in the C/A ratio indicated that fireworks released a high amount of acidic substances. The concentrations of K^+, Cl^-, NO_3^-, SO_4^{2-}, and NH_4^+ increased sharply when fireworks were used and then decreased rapidly, indicating that firework explosions had a significant effect on the increase in these WSIIs. OC was significantly affected by the $PM_{2.5}$ concentration. The effect of fireworks on OC was much higher than that on EC, as indicated by the growth rate of OC being 2.0 times and that of EC being only 1.05 times. The EFs of K, Ni, Pb, Cu, and Cr increased drastically, suggesting that fireworks largely contributed to the emission of these elements and caused potential harm to human health.

Acknowledgments:-
This work was supported by the Zhejiang Provincial Natural Science Foundation Project (LY14E070002).This manuscript was edited by Wallace Academic Editing.

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