Ekoloji 28(107): 505-515 (2019)

Characteristics of $PM_{2.5}$ Water-soluble Ions and Source Identification during Springtime 2017 in Wuhan

Lei Ding [1*], Xuejuan Fang [2], Hongan Dong [1], Weituo Zhao [3]

[1] School of International Business & Languages, Ningbo Polytechnic, Ningbo, Zhejiang 315800, CHINA

[2] Ningbo Institute of Oceanography, Ningbo 315832, CHINA

[3] The Center of Environmental Engineering and Assessment, No. 203 Research Institute of Nuclear Industry, Xianyang 712000, CHINA

[*] Corresponding author: dinglei3616028@163.com

Abstract

Daily $PM_{2.5}$ and water-soluble inorganic ions (NH_4^+, SO_4^{2-}, NO_3^-, Cl^-, Ca^{2+}, Na^+, K^+, Mg^{2+}) were collected in Hongshan Air Monitoring Station in China University of Geosciences (Wuhan) (30°31′N, 114°23′E) from March 3 to April 9 in 2017. $PM_{2.5}$ was collected using medium flow membrane filter samplers. And the ions were determined by ion chromatography. Then, the Positive Matrix Factorization method (PMF) and the HYSPLIT were selected to analyze the $PM_{2.5}$ pollution sources. The average mass concentration of $PM_{2.5}$ was found to be 101.34±31.52 $\mu g/m^3$. And the average mass concentrations of the 8 ions was 58.88 $\mu g/m^3$, with the order as $NH_4^+ > SO_4^{2-} > NO_3^- > Cl^- > Ca^{2+} > Na^+ > K^+ > Mg^{2+}$. Among them, NH_4^+, SO_4^{2-} and NO_3^- were the major components of water-soluble ions in $PM_{2.5}$ with their sum took 97% of the total ions concentrations. The PMF results showed that the following 7 $PM_{2.5}$ pollution sources decreased in order of secondary nitrate (23%) > coal combustion (18%) > biomass burning (15%) > traffic source (13%) > secondary ammonium (12%) > road dust (10%) > sea salt (9%). HYSPLIT back trajectories described the four prevailing airflow directions included E, ES, S and NE directions that affect $PM_{2.5}$ concentrations in a spatial scale during the observing period. Meanwhile, the wind from NW and N directions might facilitate the dispersion of the pollutant.

Keywords: $PM_{2.5}$, water-soluble, PMF, HYSPLIT, Wuhan

Ding L, Fang X, Dong H, Zhao W (2019) Characteristics of $PM_{2.5}$ Water-soluble Ions and Source Identification during Springtime 2017 in Wuhan. Ekoloji 28(107): 505-515.

INTRODUCTION

With the sustainable development of industrial activity, population quantity and traffic density, $PM_{2.5}$ pollution is becoming more and more serious (Yu et al. 2013). The main sources of $PM_{2.5}$ are vehicle emission, biomass burning, coal burning, road dust, sulfate, nitrate and two kind industrial emissions, etc. (Gugamsetty et al. 2012, Pandolfi et al. 2011, Zhang et al. 2015b). Of course, the source of $PM_{2.5}$ has the diversity and uncertainty in different spatial and temporal scales. It has been widely realized that the research about the material and spatial sources of $PM_{2.5}$ plays an important role in the treatment and prevention of $PM_{2.5}$ pollution. Among them, previous researches showed that the $PM_{2.5}$ composed of organic matter, water soluble ions and various heavy metal elements, etc. (Du et al. 2011, Zhang et al. 2011), and the mass of water soluble ions could get as high as above 80% in $PM_{2.5}$, which became the main element to determine the mass concentration level of $PM_{2.5}$.

In recent years, plenty of studies have been carried out on water soluble ions of $PM_{2.5}$, such as studies about the composition of water soluble ions (Liu et al. 2015), the pollution characteristics and source analysis (Huang et al. 2015, Zhang et al. 2018), and the conversion mechanism between gaseous pollutants and water soluble ions (Zhang et al. 2018), etc.. At the same time, some scholars focused on the temporal-spatial distribution characteristics and long-distance transmission characteristics of $PM_{2.5}$'s water soluble ions (Wang et al. 2005, 2015), regarded as the groundwork for understanding the pollution characteristics and sources of $PM_{2.5}$. Seen from the $PM_{2.5}$ source apportionment methods, the Source Emissions List Method, the Diffusion Model Method and the Receptor Model Method are the main analysis methods (Zhang et al. 2015b). The analytical results from the Source Emissions List Method are simple and clear, but it always contains some defects, for instance, emission factors have greater uncertainty, the open and

 Received: 12 Feb 2018 / Accepted: 4 Oct 2018

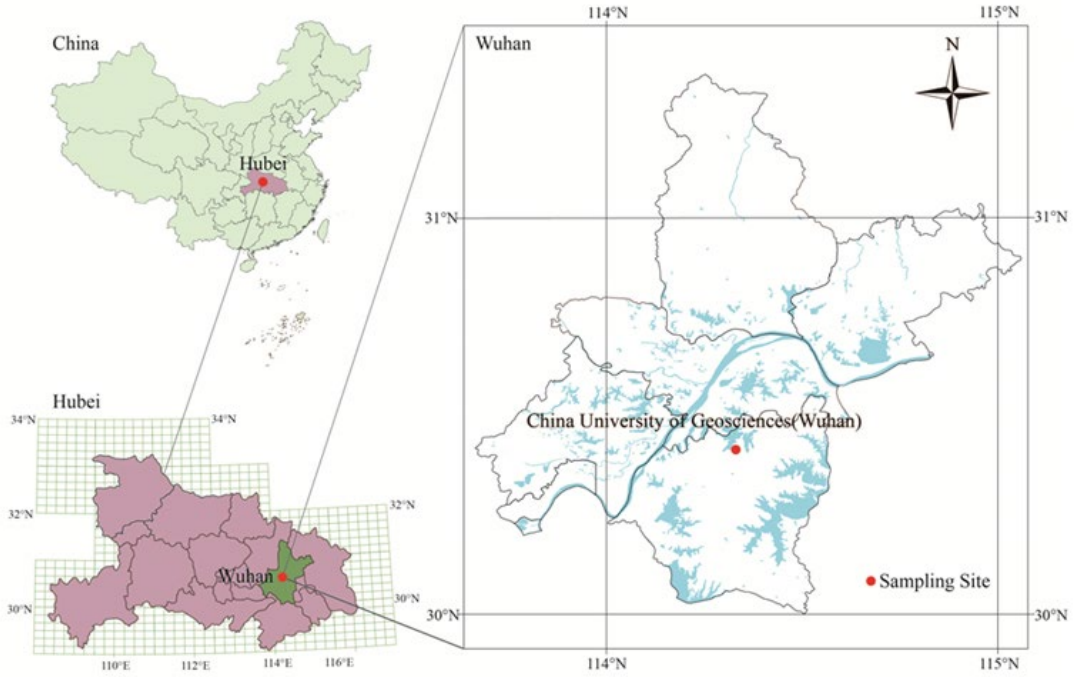

Fig. 1. The location map of the sampling site

natural sources emissions statistics is difficult to obtain. Similarly, the Diffusion Model Method also exist the uncertainty caused by the source list, the boundary layer meteorological processes and the complex atmospheric chemistry processes, whereas it is less adopted on the studies of particle source apportionment. The Receptor Model Method mainly contains the positive definite matrix factorization method (PMF) (Zhao et al. 2011), principal component analysis (PCA) (Tanet al. 2014), multivariate linear model (ME2) (Wang et al. 2013a) and UNMIX (Crawford et al. 2015), etc. In addition, the Isotope Method (Vedantham et al. 2014) and Back Trajectory Tracking Method (Wei et al. 2015) have laid the foundation of the identification of the long-distance transport of pollution sources. Based on above mentioned, this paper uses the PMF method and the Backward Trajectory Tracking Method to study the source of $PM_{2.5}$ in Wuhan during the observation period, in order to provide a reference for the particle pollution joint prevention and control in Wuhan and its surrounding areas.

MATERIALS AND METHODS

The Sampling Site and the Acquisition Process

The sampling site lies on the roof of institute of atmospheric environment in China University of Geosciences (Wuhan) (30°31′N, 114°23′E), Hongshan district of Wuhan (**Fig. 1**). The sampling elevation is 38 m, while is about 8 meters away from the ground. The sampling area is near the Lumo Road, one of the main roads in Wuhan, with large traffic flow, densely populated around, and no obvious fixed emissions sources peripherally.

We utilized medium flow membrane filter samplers to collect $PM_{2.5}$ samples (TH-150F type, Wuhan Tianhong Company, China), the flow rate was 100 L/min. Furthermore, quartz fiber membrane filter was selected to collect sample filter membrane (QFF, Φ90 mm, Whatman Company, UK). Sampling time was from March 3 to April 9 in 2017, each time maintained 24 hours, and 33 effective samples were obtained finally. Each sample was encapsulated by aluminum foil packaging in low temperature and light avoiding environment. We cut up a quarter samples' filter membranes, put them into polypropylene centrifugal pipe, then added 30ml ultrapure water, stewed them after 30min constant temperature ultrasonic extraction, and filtered them with 0.45um diameter's micro porous membranes. ICS-1100 ion chromatograph and Inductively Coupled Plasma Optical Emission Spectrometer (ICAP6300, Thermo Fisher Scientific, America) were selected to determine the mass concentration of each anion and cation. Sample analysis process was carried out with strict quality control.

The Analytical Method of $PM_{2.5}$ Source

The Positive Matrix Factorization (PMF)

The Positive Matrix Factorization (PMF) is a kind of multivariable factor analysis tool, which decomposes one special sample data matrix into two matrices: factor

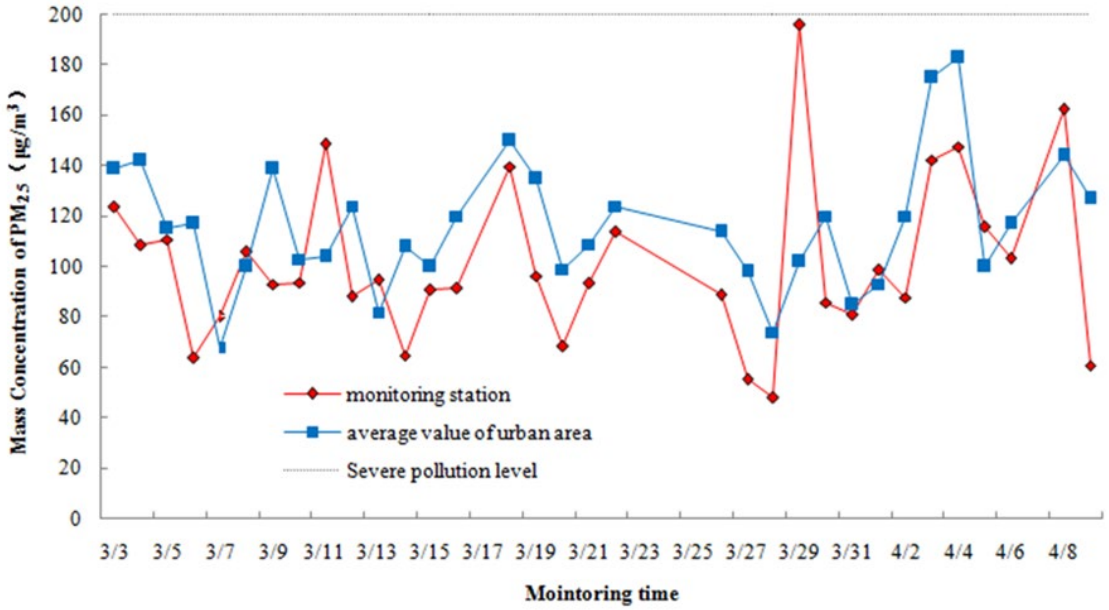

Fig. 2. The mass concentration of $PM_{2.5}$ in monitoring station and average value of urban area in Wuhan, 2017

contributions (G) and factor profiles (F), then identify the source type that contributes to the sample through the analysis of the factors (Yerramilli et al. 2012). Based on the mass balance principle, PMF method integrates in the data error estimation to solve a restricted weighted least-squares linear model (Fan et al. 2014). Its advantage is, when analyzing the source of atmosphere, we only need to enter the particulate concentration of each chemical composition and the standard deviation of the measurement, while don't need to enter the information of the component's input sources. It could not only deal with complex ingredient datum, but also provide better uncertainty evaluation result. This study specifically adopted EPA official $PMF_{5.0}$ model.

HYSPLIT back trajectory analysis

HYSPLIT back trajectory tracking simulation is exploited by Draxler et al. (1998) from the National Oceanic and Atmospheric Administration (NOAA), which is a comprehensive model system developed for diffusion and sedimentation analysis of particle trajectory and could quickly get the source place and transmission path. This study used version HYSPLIT 4.9 (http://ready.arl.noaa.gov/HYSPLIT.php), and the meteorological data came from the GDAS Database (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdasl). The resolution was 1°+1°. The back airflow trajectory time set to 12 hours and 72 hours, the trajectory initial simulation height set to 10 m, 500 m, and 1000 m. In consideration of wind at altitude of 500m field could reflect the average boundary layer flow field characteristics more accurately, the emphasis research area was set on the trajectory of 500m height, in order to reflect the characteristics of the pneumatic conveyor (Draxler and Hess 1998).

RESULTS AND DISCUSSION

The Level of $PM_{2.5}$ Quality and Comparison

The experiment results of 33 $PM_{2.5}$ samples showed that the average mass concentration of $PM_{2.5}$ during the observation period was 101.34 $\mu g/m^3$ and the variation changed from 47.99 $\mu g/m^3$ to 195.87 $\mu g/m^3$. The $PM_{2.5}$ average level was lower than the daily average level of urban area in Wuhan; while the datum in March 11 and March 29 was obviously higher than that of the urban density anomalously (**Fig. 2**). Among them, the number of samples' $PM_{2.5}$ quality concentration exceeding to the Ambient Air Quality Standards (GB3095-2012) in limit value of 75 $\mu g/m^3$ was 88%. During the period of observation, the air quality is moderate-pollutional in two days, light-pollutional in twenty-one days and well in 10 days. Compared with existing related research conclusions, $PM_{2.5}$ mass concentration levels of the functional domains in different areas exhibited obvious differences (**Table 1**). Overall, the concentration levels of $PM_{2.5}$ in the different urban zones were listed as follows: the industrial zone > the business zone > the clean control zone > the traffic area > the science and education area. The mass concentration levels of various science and education areas were close to each other.

The total mass concentration value of the 8 $PM_{2.5}$'s water-soluble ions was 58.88 $\mu g/m^3$, accounted for 60% of the whole $PM_{2.5}$ mass concentration. The mass concentration level's sequence from high to low order

Table 1. Comparing with the mass concentration level of $PM_{2.5}$ in different functional areas in Wuhan

Author	Sampling sites	Functional areas	Sampling time	$PM_{2.5}$ ($\mu g/m^3$)
Wang et al. (2013)	Wuhan Steel Group	Industrial area	2012.1	252.44
	Chinese Academy of Sciences, Wuhan botanical garden	botanical garden (background)	2012.1	120.75
	At the gate of South Lake School District, Wuhan University of Technology	Traffic area	2012.1	116.56
Zeng et al. (2013)	Optics Valley Business area	Business area	2011.3-4	121
Zhang et al. (2015)	Within Wuhan University	Science and Education area	2012.8-2013.7	106.5
This study	Within China University of Geosciences (Wuhan)	Science and Education area	2017.3-2017.4	101.34

Table 2. The descriptive statistics of $PM_{2.5}$ and 8 water-soluble ions ($\mu g/m^3$)

Items	$PM_{2.5}$	K^+	Ca^{2+}	Na^+	Mg^{2+}	NH_4^+	Cl^-	NO_3^-	SO_4^{2-}
Maximum	195.872	0.235	2.511	1.373	0.105	48.336	1.632	40.505	48.320
Minimum	47.992	0.045	0.007	0.013	0.0001	9.418	0.215	0.409	8.112
Mean	101.340	0.113	0.265	0.205	0.003	24.929	0.801	13.721	18.929
St. D	31.522	0.054	0.416	0.259	0.018	8.105	0.467	10.943	7.397
CV	0.311	0.484	1.568	1.262	5.403	0.325	0.583	0.798	0.391

was $NH^{4+}>SO_4^{2-}>NO_3^-$>Cl^->Ca^{2+}>Na^+>K^+> Mg^{2+}. The secondary ions NH_4^+, SO_4^{2-} and NO_3^- were the main water soluble ions, accounted for 97% of the total soluble water ions. It could be known from the variation coefficient in **Table 2** that the concentration volatility range of NH_4^+ and SO_4^{2-} was relatively small, which were two stable sources of pollution. Among them, the ammonium salt was mainly transforms from NH_3, which was an important component that neutralize acidic materials like SO_2 and NO_X, to form secondary particles (Zhang et al. 2015a). In addition, existing research demonstrated that NH_3 mainly came from agricultural production, industrial emissions, and other motor vehicle exhaust emission source (Battye et al. 2003).

The Relationship between SO_2, NO_2 and Sulfate, Nitrate

Sulfur oxidation rate SOR (SOR = $SO_4^{2-}/(SO_4^{2-} + SO_2)$) and nitrogen oxide rate NOR (NOR = $NO_3^{2-}/(NO_3^- + NO_2)$) could reflect the process and degree that SO_2 and NO_2 translated to sulfates and nitrates (Zhang et al. 2018). The high value of SOR and NOR illustrated that the quadratic transformation degree of the sulfur oxides and nitrogen oxides in atmospheric aerosols was high. The average concentration of SO_2 and NO_2 was 14.51 $\mu g/m^3$ and 26.17 $\mu g/m^3$ respectively, which acquired through counting every single sample's hourly average during the observation period. The average of SOR and NOR was 0.54 and 0.30. The SOR and NOR number ranged from 0.15 to 0.75 and from 0.02 to 0.64, respectively. The concentration of NO_2 was higher and the NOR was lower, which showed that the secondary conversion rate of NO_2 was lower than that of SO_2.

SO_2 in the atmosphere mainly came from combustion of fossil fuels, while NO_x in the atmosphere mainly stemmed from industrial pollution and motor vehicle exhaust (Peng et al. 2000). The ratio of NO_3^- and SO_4^{2-} quality concentration was used to measure the relative contribution of fixed source (coal) and mobile source (vehicles) to sulfur and nitrogen pollution in the atmosphere. The value of NO_3^-/SO_4^{2-} in this study fluctuated between from 0.03 to 2.71, and the average value was 0.80, which was comparable with most areas in China and slightly higher than Jinan (Wang et al. 2006), Shanghai (Cao et al. 2012). During the observing period, 67% of the samples' NO_3^-/SO_4^{2-} equivalent ratio was less than 1, which showed that the contribution rate of fixed source emissions to sulfur pollution was greater than that of mobile source to nitrogen pollution in the atmosphere.

The Main Source Analysis and Source Contribution of $PM_{2.5}$

Operation results and analysis of PMF

The operation result of PMF was evaluated by the signal-to-noise ratio, intercept, slope, r^2, etc.. Through transforming operation parameters constantly, we obtained the results (7 factors) with the intercept closing to 0, the slope closing to 1, r^2 exceeding 0.6, signal-to-noise ratio and residual meeting the requirements and Q value (393) closing to the actual point 396 (33×12/d). According to PMF analysis, this study confirmed 7 pollution sources: coal combustion, road dust, traffic source, secondary nitrate and ammonium salt, biomass combustion source, sea salt. **Fig. 3** showed the 7 Factors' source profiles.

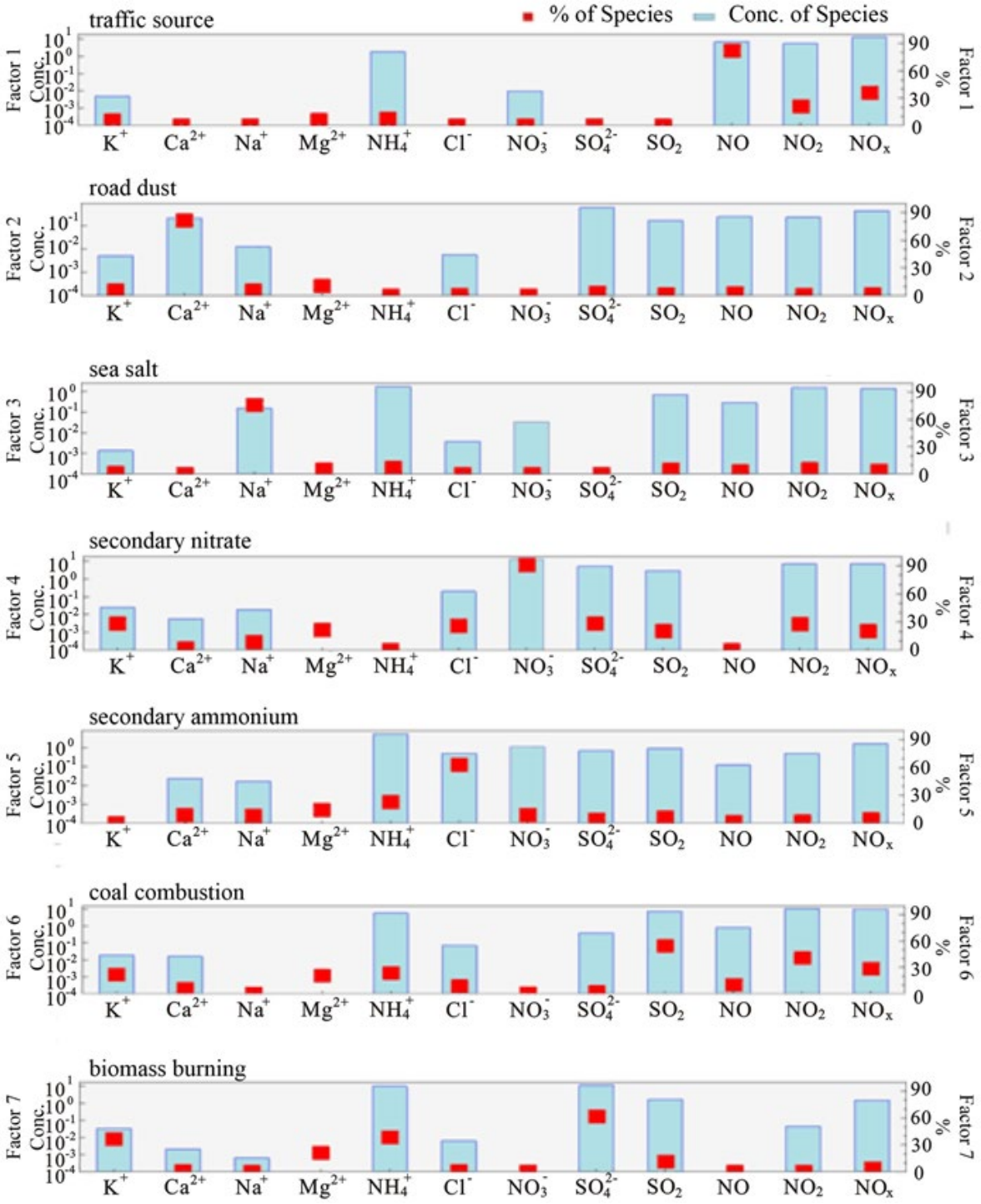

Fig. 3. Source profile resolved from PMF for $PM_{2.5}$ in Wuhan during springtime in 2017

Traffic source mainly referred to the automobile exhaust, ingredients including CO and nitrogen oxides, while nitrogen oxides maybe composed of 95% NO and the rest was NO_2. The concentration and percentage of NO, NO_X and NO_2 were larger, and the content of NO was more than 80% in Factor 1, thus the Factor 1 was judged to the source of traffic. Road dust contained a large number of the earth's crust characteristic elements (Ca, Na, Mg and K), high concentration of Ca in $PM_{2.5}$ mainly came from farmland and bare surface (Yao et al. 2002). The concentration and percentage of Ca^{2+} and Mg^{2+} in Factor 2 were larger, thus it was distinguished for the road dust source. The percentage of Na^+ reached to 76% in Factor 3, high concentration of Na could be used as a sea salt ion iconic (Koçak et al. 2015, Lough et al. 2005), thus Factor 3 was regarded as the sea salt source. Secondary aerosols were mainly composed of sulfate, nitrate and ammonium salt, and they were more

quadratic transformed by precursor gaseous NH_3, SO_2 and NO_x. The density and percentage of NO_3^- and NH_4^+ in Factor 4 and Factor 5 were respectively larger, thus the Factor 4 and Factor 5 were judged to secondary nitrate and secondary ammonium pollution. Fossil fuels contained S, N elements, they existed as SO_2 and NO_2 after burning. Therefore, SO_2 and NO_2 are regarded as the symbol pollution from fossil fuels (coal etc.). The density and percentage of NO_2 and SO_2 in Factors 6 was larger, thus Factor 6 was distinguished for coal source. Biological plastid contained large amount of elements, such as K, N, S. It existed with the form of K^+, SO_4^{2-}, SO_2, etc. after burning in atmospheric aerosol. As a result, high concentration of K^+ and SO_4^{2-} could be used as the characteristics of the ions in biomass burn, wood burn and crop burn (Ovadnevaite et al. 2012). The density and percentage of K^+ and SO_4^{2-} in the Factor 7 was higher, thus Factor 7 was discriminated as biomass burning.

The contribution of each pollution source to $PM_{2.5}$ existed difference. The contribution of secondary aerosols (nitrate and ammonium salt) and coal source was respectively 35% and 18%, bigger than other sources. The resident and industrial coal behavior made contribution to SO_2 and NO_x in the atmosphere in Wuhan and its surrounding areas. They transferred to the urban zone through the atmospheric turbulence of migration movement and happened quadratic transformation during the migration process. The contribution rate of biomass combustion source was 15%, the spring village burned large areas of hay in forest planting, and the air pollution was transferred with atmospheric flow to urban area, which affected the urban air quality.

In addition, it's worth noting that the contribution rate of traffic pollution was 13%, with the arising number of vehicles in Wuhan city, motor vehicle exhaust emissions also increased gradually. The traffic pollution contribution to $PM_{2.5}$ was gradually obvious in Wuhan now. The contribution rate of road dust is 10%, of course, the road dust existed certain differences in season and space, and was strongly influenced by human activities. The road dust contribution in urban area was more related to human activities, a new large area of urban construction was happened in recent years in Wuhan, such as estate development, metro construction, road construction, building demolition reconstruction, etc., and the producing dust migrated with the atmospheric turbulence, which affects the urban air quality. The contribution rate of sea salt was 9%, sea salt moves to inland areas with atmospheric air movement, and affects the quality of $PM_{2.5}$ concentrations to a certain extent, in spite of with little impact in springtime.

The influenced analysis of water-soluble inorganic ions by regional transportation

For the sake of study transmission path for the air pollution in Wuhan city, using HYSPLIT (the back trajectory tracking model) to analysis air trajectory of extreme concentration of $PM_{2.5}$ with 12 h and 72 h back trace simulation during the observation time. The 12 h back trajectory tracking simulation selected the wave on March 3, 11, 18, 22, 29, April 4 and 8, seven time sections; 72 h back trajectory tracking simulation choose March 3, 9, 18 and 21, four time sections. Among them, the March 9, 21 were trough time sections, the air quality grade was fine, in addition to April 4 was the air quality for moderate pollution, other time were all light pollution.

As shown in **Fig. 4**, the 12 h back trajectory tracking was mainly from the direction of E, SE and NE direction, the direction of E on the transmission path were Jiujiang-Ezhou-Huangshi-Huanggang cities, and the 12 h back trajectory tracking observation point on March 3 in 500 m altitude was derived from the Huangshi region. Huangshi and Ezhou were given priority with heavy industry, for metallurgy, energy, cement, building materials industry as the dominant industry, and the industrial pollution was relatively serious, industrial waste gas transferred with atmospheric gas and affected the air quality of Wuhan. Meanwhile, there were a large number of sulfur oxides and nitrogen oxides transforming to secondary aerosol particulate matter during this period, NOR and SOR values are around 0.6, and NO_3^-/SO_4^{2-} equivalent ratio was greater than 1, the quality of the NO_3^- concentration appeared anomalies within the point on time, it showed that there had the extraneous source of contribution. Typically, the industrial waste gas pollution sources in Huangshi had a greater influence on Wuhan $PM_{2.5}$ concentrations in spring.

The transmission of SE direction path was mainly the Xianning- Huanggang- Huangshi cities, etc., such as the airflow path transmission path on March 11, 18, 22 and April 9. The 12 h back trajectory tracking was followed up on March 11 and 18 at 500 m in Xianning. By contrast, industrial pollution was lighter in Xianning, with a perennial good air quality, and the two long distance air mass migrations produced tiny effects. Meanwhile, the mass concentration of NH_4^+ was abnormal On March 18, which showed that pollution is

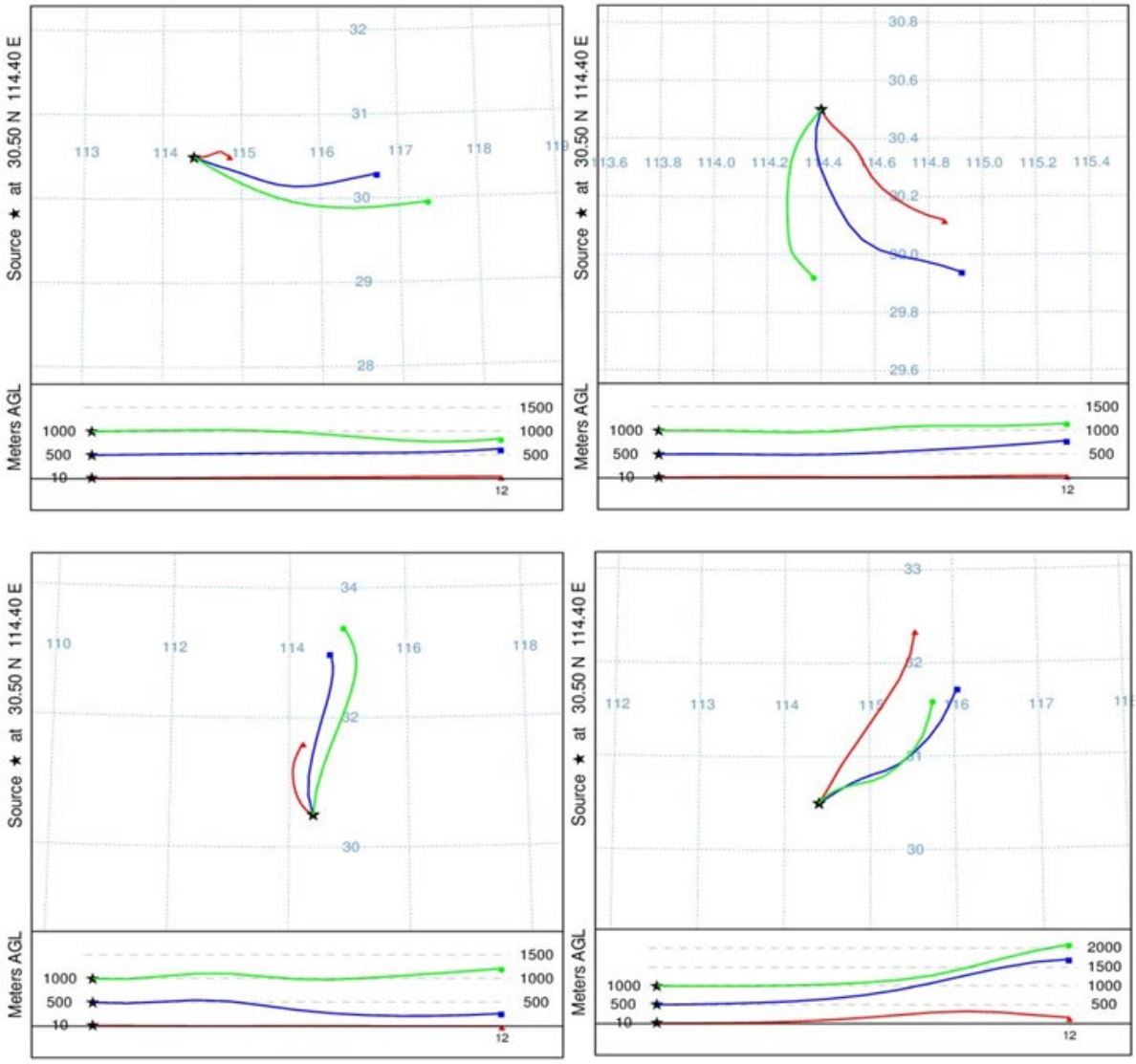

Fig. 4. The 12 h backward trajectory of airflow above observation point in slight and moderate pollution time

strongly influenced by local human activities in Wuhan city. The 12h back trajectory tracking came from Nanchang and Huangshi on March 22 and April 9 at 500 meters, respectively. In consideration of the air quality in Nanchang city reached the standard rate of 96% on March 2017, thus the air transport of observation points had little impact on air quality, local pollution sources mainly came from Wuhan. The SO_4^{2-} and NH_4^+ quality density appear anomalies, which were affected by industrial pollution in Huangshi on April 9; it suggested that the pollution mainly came from Huangshi. The observation point of NO_3^-/SO_4^{2-} equivalent ratio were far less than 1 in the four days, sources were given priority to stationary pollution.

The NE direction on the transmission path mainly stems from Anhui, Henan and other places passed the Qingshan District of Wuhan City, such as the airflow transmission path on March 29 and April 4. The 12 h back trajectory tracking was followed up on March 29 at 500 meters in Henan Zhoukou, by way of via suburbs of Caidian and Huangpi District in Wuhan. The 12 h back trajectory tracking was followed the trail on April 4 at 500 m in Anhui and Henan, go through the Qingshan District of Wuhan City.

The observation point of $PM_{2.5}$ mass concentration was much higher than average on March 29, it showed that external pollution sources from Henan coal (Wu et al. 2007) and Qingshan district industrial waste gas exacerbated the observation point of $PM_{2.5}$ pollution level, and gave priority with stationary sources.

The airflow trajectory atmosphere in Wuhan $PM_{2.5}$ was under different air quality during observation, as was shown in **Fig. 5**, weather conditions after 72 h to back airflow trajectory were respectively from the

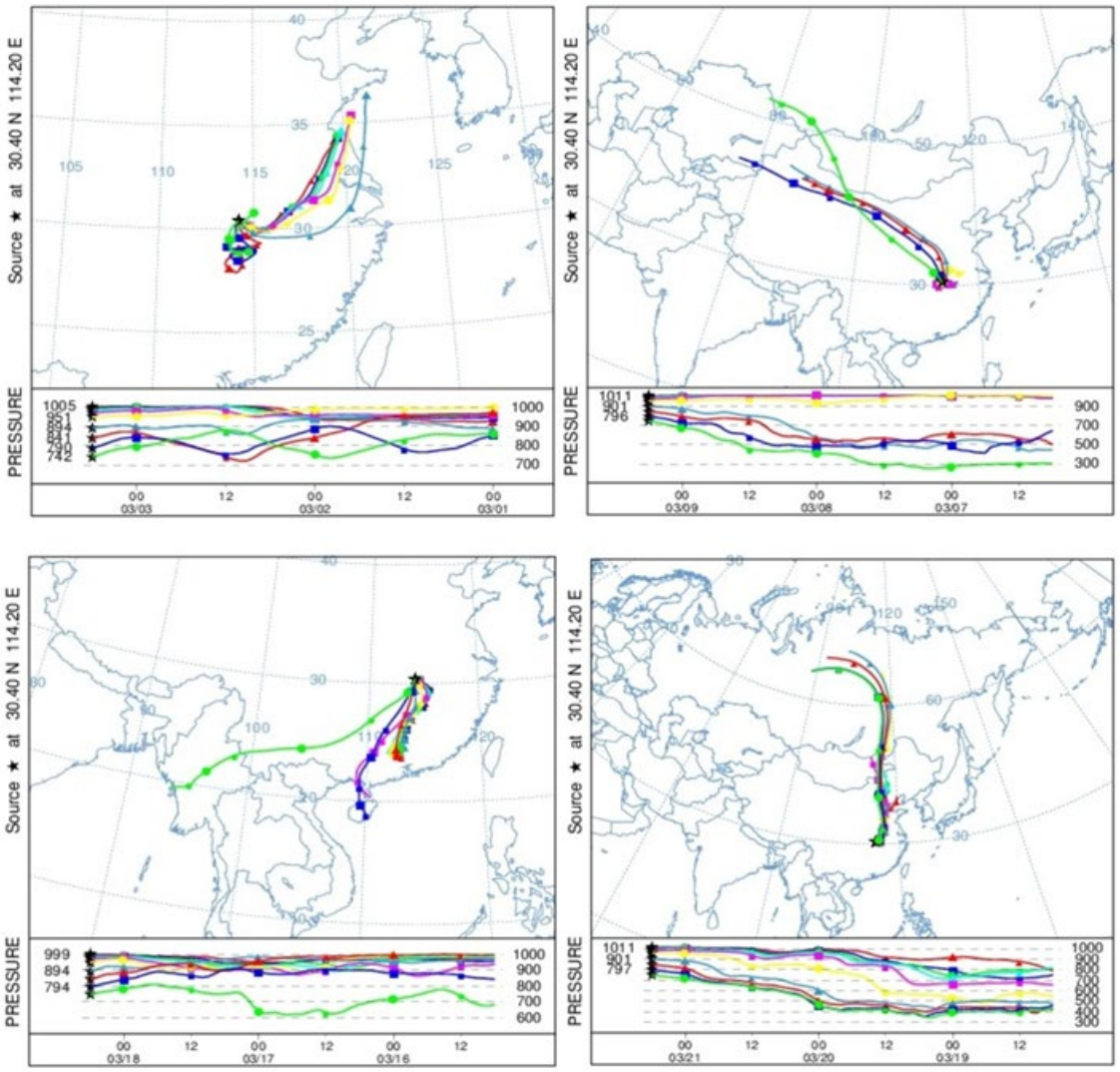

Fig. 5. The 72 h backward trajectory of airflow above observation point on March 3, 9, 18, 21 in Wuhan

direction of NE, NW and S, N on March 3, 18 (light pollution), and March 9, 21 (good). 2000 m height of airflow trajectory is came from Shandong (March 1) and Jiangsu, Anhui and other places (March 2) in NE direction, the lower atmosphere air in 500 m height came from Wuhan, the upper atmosphere over the air flow stems from the Yellow Sea.1000 m above the height of airflow trajectory came from Hainan, Guangdong (March 18), Guangxi and Hunan Provinces etc. (March 21) in S direction. Air pollution matter content was less from the sea, and the pollutants which as coal of the main fuel in Guangzhou (Wu et al. 2015) were migrate along with currents move to Wuhan over. 500m height of airflow trajectory (March 7) came from Xinxiang, Gansu, Inner Mongolia (March 8) in NW direction, 2000m high atmosphere air came from Wuhan. 700m upper atmosphere over the N direction airflow (March 19) derived from Russia (19 March), Mongolia and Inner Mongolia (20 March). The atmospheric turbulence in N and NW direction via region vast, less air pollution matter, long-distance migration of the atmospheric turbulence had small effect on the air pollution of Wuhan City, and would accelerate the diffusion of pollutants in the air in Wuhan City, so as to reduce the concentration of $PM_{2.5}$ levels. Overall, long-distance transmission of atmospheric turbulence had cause certain influence to the concentration of $PM_{2.5}$ in Wuhan City, and mainly from E, ES, S and NE direction of cities, such as Huangshi, Ezhou and other regions within the scope of urban circle.

CONCLUSIONS

The average mass concentration of $PM_{2.5}$ was found to be 101.34±31.52 $\mu g/m^3$ during the March 3 to April 9 in 2017 with obvious fluctuation, the concentration ranged from 47.99$\mu g/m^3$ to 195.87$\mu g/m^3$. The $PM_{2.5}$ daily average concentration exceeding standard rate was

up to 88%. The total mass concentration of eight kinds of water-soluble inorganic ions (K^+, Ca^{2+}, Na^+, Mg^{2+}, NH_4^+, Cl^-, NO_3^-, SO_4^{2-}) was 58.88$\mu g/m^3$, accounting for 59.66% of the mass concentration of $PM_{2.5}$, with the order as $\rho NH_4^+ > \rho SO_4^{2-} > \rho NO_3^- > \rho Cl^- > \rho Ca^{2+} > \rho Na^+ > \rho K^+ > \rho Mg^{2+}$. Among them, NH_4^+, SO_4^{2-} and NO_3^- were accounted for 97% in all measured water-soluble ions, which were the main ions to control the level of $PM_{2.5}$ mass concentration.

The SOR of $PM_{2.5}$ water-soluble ion value was greater than 0.10, the NOR value was greater than 0.10, and the average value reaches 0.54 and 0.30, respectively. The conversion level of SO_2, NO_2 and NO_x was higher, while the NO_3^{2-} and SO_4^{2-} in atmospheric aerosol were mainly from the secondary transformation with SO_2 and NO_x. The range of equivalent ratio NO_3^-/SO_4^{2-} in $PM_{2.5}$ was between 0.03-2.71, with average value was 0.80, the equivalent ratio of NO_3^-/SO_4^{2-} less than 1 sample number accounted for 67% in the total number of samples. It illustrated that the source contribution proportion of $PM_{2.5}$ from fixed emitters (such as coal burning from Wuhan Iron and Steel Corporation in Qingshan Distinct) was bigger than the mobile emission source (vehicle) during springtime in Wuhan.

During the period of observation, it had large contributed of $PM_{2.5}$ pollution sources, secondary aerosols (nitrate and ammonium were 23% and 12%) > coal source (18%) > biomass combustion source (15%) >traffic pollution (13%) > road dust (10%) > sea salt (9%), the structure of each pollution source was closely related to the energy structure, traffic condition, biomass fuel burning and urban construction in Wuhan City and surrounding areas. Another important aspect, air flow in different direction also had certain influence on mass concentrations of $PM_{2.5}$ in Wuhan, and the airflow contribution to $PM_{2.5}$ was more apparent from E, ES, S and NE direction, while airflow would accelerate the diffusion of pollutants in Wuhan city in N and NW direction. Therefore, we need to strengthen the air pollution with joint prevention and control in Wuhan and the southeast of Huangshi, Ezhou, Huanggang in future, take steps on the technical transformation, the structure upgrade, energy conservation and emissions reduction and optimize, in the meantime, layout the industrial reasonable, strictly control the environment access.

ACKNOWLEDGEMENTS

This work was financially supported by the Talent Introduction Research Project of Ningbo Polytechnic (Grant No. RC201705) and the Zhejiang Philosophy and Social Science Planning Project (Grant No. 18NDJC109YB).

REFERENCES

Battye W, Aneja VP, Roelle PA (2003) Evaluation and improvement of ammonia emissions inventories. Atmos. Environ. 37: 3873-3883.

Cao JJ, Shen ZX, Chow JC, Watson JG, Lee SC, Tie XX, Ho KF, Wang GH, Han YM (2012) Winter and summer PM2.5 chemical compositions in fourteen Chinese cities. Journal of the Air & Waste Management Association 62: 1214-1226.

Crawford J, Chambers S, Cohen DD, Griffiths A, Williams A, Stelcer E (2015) Using radon-222 as an indicator of atmospheric mixing depth in ME-2 for PM2.5 source apportionment. Aerosol and Air Quality Research 15: 611-624.

Draxler RR, Hess GD (1998) An overview of the HYSPLIT_4 modeling system for trajectories, dispersion, and deposition. Australian meteorological magazine 47: 295-308.

Du HH, Kong LD, Cheng TT, Chen JM, Du JF, Li L, Xia XG, Leng CP, Huang GH (2011) Insights into summertime haze pollution events over Shanghai based on online water-soluble ionic composition of aerosols. Atmos. Environ. 45: 5131-5137.

Fan J, Yue XY, Jing Y, Chen Q, Wang SG (2014) Online monitoring of water-soluble ionic composition of PM10 during early summer over Lanzhou City. J. Environ. Sci. 26: 353-361.

Gao M, An JL, Hang YX, Li YY (2014) Comparison of PMF and PCA/APCS for VOCs source apportionment in north suburb of Nanjing. J. Meteor. Environ. 30: 43-50.

Gugamsetty BK, Wei H, Liu CN, Awasthi A, Hsu SC, Tsai CJ, Roam GD, Wu YC, Chen CF (2012) Source characterization and apportionment of PM10, PM2.5 and PM0.1 by using positive matrix factorization. Aerosol. Air. Qual. 12: 476-491.

Hagler GSW, Bergin MH, Salmon LG, Yu JZ, Wan ECH, Zheng M, Zeng LM, Kiang CS, Zhang YH, Lau AKH, Schauer JJ (2006) Source areas and chemical composition of fine particulate matter in the Pearl River Delta region of China. Atmos. Environ. 40: 3802-3815.

Huang P, Zhang JY, Tang YX, Liu L (2015) Spatial and Temporal Distribution of PM2.5 Pollution in Xi'an City, China. Int. J. Environ. Res. Public Health. 12: 6608-6625.

Koçak M, Mihalopoulos N, Tutsak E, Theodosi C, Zarmpas P, Kalegeri P (2015) PM10 and PM2.5 composition over the Central Black Sea: origin and seasonal variability. Environ. Sci. Pollut. Res.: 1-17.

Lam KS, Wang TJ, Chan LY, Wang T, Harris J (2001) Flow patterns influencing the seasonal behavior of surface ozone and carbon monoxide at a coastal site near Hong Kong. Atmos. Environ. 35: 3121-3135.

Liu BX, Yang DY, Zhang DW, Chen T, Zhao HS, Chen YY, Lu HY (2015) Chemical species of PM2.5 in the urban area of Beijing. Environmental Science. 36: 2346-2352.

Lough GC, Schauer JJ, Park JS, Shafer MM, Deminter JT, Weinstein JP (2005) Emissions of metals associated with motor vehicle roadways. Environ. Sci. Technol. 39: 826-836.

Ovadnevaite J, Ceburnis D, Canagaratna M, Berresheim H, Bialek J, Martucci G, Worsnop DR, Dowd CO (2012) On the effect of wind speed on submicron sea salt mass concentrations and source fluxes. J. Geophys. Res. Atmospheres 117(D16).

Pandolfi M, Gonzalez-Castanedo Y, Alastuey A, de la Rosa JD, Mantilla E, de la Campa AS, Querol X, Pey J, Amato F, Moreno T (2011) Source apportionment of PM10 and PM2.5 at multiple sites in the strait of Gibraltar by PMF: impact of shipping emissions. Environ. Sci. Pollut. Res. 18: 260-269.

Peng YD, Yang MZ, Shen LX (2000) Ammonia emission and its effects on the format ion of secondary particles in Beijing. Environmental Science 21: 101-103.

Tan JH, Duan JC, Chai FH, He KB, Hao JM (2014) Source apportionment of size segregated fine/ultrafine particle by PMF in Beijing. Atmos. Environ. 139: 90-100.

Vedantham R, Landis MS, Olson D, Pancras JP (2014) Source identification of PM2.5 in Steubenville, Ohio using a hybrid method for highly time-resolved data. Environ. Sci. Technol. 48: 1718-1726.

Wang J, Hu ZM, Chen YY, Chen ZL, Xu SY (2013a) Contamination characteristics and possible sources of PM10 and PM2.5 in different functional areas of Shanghai, China. Atmos. Environ. 68: 221-229.

Wang L, Zhou JB, Yuan JP, Guo HT, Xiao JH, Zhou Y, Yu WY, Liu YL (2013b) Distribution and source identification of hydrocarbons in atmospheric PM2.5 in Wuhan. Journal of Wuhan University of Technology 12: 140-145.

Wang MT, Zhu B, Wang HL, Xue GQ, He J, Xu HH, Cao JF (2015) Composition and regional characteristics of atmosphere aerosol and its water soluble ions over the Yangtze River Delta region in a winter haze period. Environmental Science 36: 2337-2345.

Wang Y, Zhuang GS, Tang AH, Yuan H, Sun YL, Chen S, Zheng AH (2005) The ion chemistry and the source of PM2.5 aerosol in Beijing. Atmos. Environ. 39: 3771-3784.

Wang Y, Zhuang GS, Zhang XY, Huang K, Xu C, Tang AH, Chen JM, An ZS (2006) The ion chemistry, seasonal cycle, and sources of PM2.5 and TSP aerosol in Shanghai. Atmos. Environ. 40: 2935-2952.

Wei Y, Guo ZB, Ge X, Zhu SN, Jiang WJ, Shi L, Chen S (2015) Tracing sources of sulfate aerosol in Nanjing northern suburb using sulfur and oxygen isotopes. Environmental Science 36: 1182-1186.

Wu CF, Larson TV, Wu SY, Williamson J, Westberg HH, Liu LJS (2007) Source apportionment of PM2.5 and selected hazardous air pollutants in Seattle. Sci. Total. Environ. 386: 42-52.

Wu FY, Wang W, Man YB, Chan CY, Liu WX, Tao S, Wong MH (2015) Levels of PM2.5/PM10 and associated metal (loid) s in rural households of Henan Province, China. Sci. Total. Environ. 512: 194-200.

Yao XH, Chan CK, Fang M, Cadle S, Chan T, Mulawa P, He K, Ye B (2002) The water-soluble ionic composition of PM2.5 in Shanghai and Beijing, China. Atmos. Environ. 36: 4223-4234.

Yerramilli A, Dodla VBR, Challa VS, Myles LT, Pendergrass WR, Vogel CA, Dasari HP, Tulri F, Baham JM (2012) An integrated WRF/HYSPLIT modeling approach for the assessment of PM2.5 source regions over the Mississippi Gulf Coast region. Air. Qual. Atmos. Health 5: 401-412.

Yu LD, Wang GF, Zhang RJ, Zhang LM, Song Y, Wu BB, Li XF, An K, Chu JH (2013) Characterization and source apportionment of PM2.5 in an urban environment in Beijing. Aerosol. Air. Qual. 13: 574-583.

Zeng F, Hu H, Gao SP, Liang SW, Wang JM (2013) Analysis of the water-soluble anion in PM10 & PM2.5 in Optics Valley, Wuhan. Environmental Science & Technology 36: 123-126.

Zhang F, Wang ZW, Cheng HR, Lv XP, Gong W, Wang XM, Zhang G (2015a) Seasonal variations and chemical characteristics of PM2.5 in Wuhan, central China. Sci. Total. Environ. 518: 97-105.

Zhang T, Cao JJ, Tie XX, Shen ZX, Liu SX, Ding H, Han YM, Wang GH, Ho KF, Qiang J, Li WT (2011) Water-soluble ions in atmospheric aerosols measured in Xi'an, China: seasonal variations and sources. Atmos. Res. 102: 110-119.

Zhang Y, Li X, Nie T, et al. (2018) Source apportionment of PM2. 5 pollution in the central six districts of Beijing, China. Journal of Cleaner Production, 174: 661-669.

Zhang YJ, Zheng M, Cai J, Yan CQ, Hu YT, Russell AG, Wang XS, Wang SX, Zhang YH (2015b) Comparison and overview of PM2.5 source apportionment methods. Chin. Sci. Bull. 60: 109-121.

Zhao PS, Zhang XL, Xu XF, Zhao XJ (2011) Long-term visibility trends and characteristics in the region of Beijing, Tianjin, and Hebei, China. Atmos. Environ. 101: 711-718.