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STUDY ON THE CHARACTERISTICS OF WATER - SOLUBLE ION POLLUTION IN PM_{2.5} IN FULING DISTRICT

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ABSTRACT

PM_{2.5} was collected at the atmospheric observatory (located at the campus of Chongqing Yangtze Normal University in Fuling District in January and July of 2015 respectively). The variation of the mass concentration characteristics and the water-soluble ions variation of PM_{2.5} were analyzed. The pollution characteristics was found as follows: The average mass concentration of PM_{2.5} was 116.28 μg·m⁻³, 46.80 μg·m⁻³ respectively in January and July, and the mass concentration ranged from 23.21 μg·m⁻³ to 216.61 μg·m⁻³ during the sampling period. The obvious seasonal characteristics were observed. Nine water-soluble ions (F⁻, Cl⁻, NO₃⁻, SO₄²⁻, K⁺, Na⁺, NH₄⁺, Mg²⁺, Ca²⁺) were measured using the ion chromatography (ICS-600). The concentration of water-soluble ions ranged from 6.37 μg·m⁻³ to 86.30 μg·m⁻³, accounted for 42.28% in PM_{2.5} samples. Average mass concentration of SO₄²⁻, NO₃⁻ and NH₄⁺ was 30.53 μg·m⁻³, accounted for 87.99% of the total soluble ions. There existed a good correlation among SO₄²⁻, NO₃⁻ and NH₄⁺ distribution and variation. The analysis of equilibrium between cation and anion indicated a good correlation ($R^2=0.95$) with weak alkaline in winter and weak acidic in summer.

KEYWORDS:

PM_{2.5}, water-soluble ions, seasonal variation

INTRODUCTION

The research on atmospheric particles of Chongqing was mainly concentrated in the main city and Wanzhou District. The main contents mainly concentrated on concentration level, pollution characteristics, source analysis and chemical composition of total suspended particulate (TSP), PM₁₀ and PM_{2.5} [1-4]. However, study of particulate pollution in Fuling District of Chongqing is still blank. Chongqing Fuling is located in the hinterland of Three Gorges reservoir, and the Interchange of the Yangtze

River and Wujiang, which is the central city of Chongqing. Systematic research on Water soluble components of particulate matter in Fuling, which is important to study the characteristics of air pollution in Fuling area, and to provide scientific basis for the improvement of atmospheric visibility and the control of acid rain.

Water-soluble ionic component is one of the most important chemical constituents in airborne particles, and it is also one of the most studied components. There are more and more researches on water-soluble ions at home and abroad. Water-soluble components mainly include sulfate, nitrate, ammonium salt and sodium chloride, etc., generally accounted for 30% to 50% of PM_{2.5} mass [5]. SO₄²⁻, NO₃⁻ and NH₄⁺ are mainly composed of secondary particles, which are formed by physical condensation of nuclei in the atmosphere. The concentrations of SO₄²⁻, NO₃⁻ and NH₄⁺ are related to the concentrations and Conversion rate of SO₂, NO_x and NH₃ in the atmosphere, while being affected by temperature and humidity. SO₄²⁻ is mainly from fossil fuel combustion, SO₂ through oxidation reaction generated SO₄²⁻ and H₂SO₄ (including gas phase oxidation and liquid oxidation, ozone and peroxide as catalyst) [6]. NO₃⁻ is mainly from nitrogen oxides generated by oxidation reaction and motor vehicle exhaust emissions [7]. Gaseous precursor of NH₄⁺ is gaseous ammonia, mainly from plant and soil microbial release and use of farm chemical fertilizers. The Cl⁻, K⁺, Na⁺, Mg²⁺, Ca²⁺ in water-soluble ions come from sea salt and coal combustion [8], biomass combustion, marine source, soil dust, building dust and soil [9].

EXPERIMENTAL

Sample collection. The sampling point of this study is in the administrative building roof of Changjiang Normal University, Fuling District of Chongqing (E 106 ° 56'-107 ° 43 ', N 29 ° 21'-30 ° 01'). Changjiang Normal University is located in the northwest direction of Fuling District, surrounded by

the high-speed of Change Fu and South Fu, mixed with cultural and residential areas. The sampling point is about 20m above the ground, the surrounding terrain is flat, there is no obvious pollution source, the environment quality is good, can objectively represent the Fuling District atmospheric environmental pollution level and change regularity.

Sampling of the $PM_{2.5}$ is divided into two phases, respectively, January 6, 2015 - February 5, 2015 and July 1, 2015 - July 31, 2015. Daily continuous collection of 23 hours, the sampling period for the day 11:00 am to 10:00 am the next day, an interval of 1 hour for membrane exchanging, flow monitoring and calibration or instrument cleaning and maintenance.

The sampler used to collect the samples was a multi-channel chemical species sampler manufactured by Met One Instruments of America. Equipped with $PM_{2.5}$ cutting head, and the sampler flow rate is 6.7L / min. The instrument also equipped with temperature and pressure sensor. The sampling volume can be converted into the standard volume according to the real-time temperature and pressure. And the instrument can set sampling time and date. Water-soluble ion analysis of atmospheric particulates is carried out using quartz membranes. In this study, atmospheric fine particles were collected by inertial collision using $\phi 47$ mm quartz film (Whatman). Teflon filter (Whatman) made of PTFE film, blank value is low, non-hygroscopic, non-adsorption of inorganic and organic gases, usually used for $PM_{2.5}$ mass concentration analysis.

Sample analysis. After sampling, open 1/2 of the filter box lid put it into the tray, and then into the balance tank, filter into the balance chamber at least 24h.

Cut 1/4 of the filter using knife, and then cut into pieces with scissors, put these pieces into 50ml centrifuge tube, add about 10ml of ultra-pure water, ultrasound 20minute in Ultrasonic oscillator, oscillation is completed, according to the number Filter with 0.45 μ m filter to 25ml volumetric flask, repeat the two ultrasonic oscillation and filtration, constant volume to the scale line, placed in 4 °C under refrigeration, to be measured.

The cationic column is CS12A column and the anion column is AS11 (superscript -) column. The cationic column is composed of two kinds of ions, -HC and AG11-HC guard columns and ASRS suppressors. The anion and cation eluents were analyzed for 30 mm KOH and 20 mm methanesulfonic acid (MSA), respectively.

RESULTS AND DISCUSSION

Characteristics of $PM_{2.5}$ mass concentration.

$PM_{2.5}$ daily average concentration (24h average) changes, as shown in Figure 1.

The peak of $PM_{2.5}$ in winter was on January 15, January 19 and January 25, the concentration values were 204.35 μ g \cdot m⁻³, 190.92 μ g \cdot m⁻³ and 216.61 μ g \cdot m⁻³, respectively. It appears valley in January 28, and the value was 27.40 μ g \cdot m⁻³. The concentration of $PM_{2.5}$ in summer was on July 10, July 19 and July 25, the concentration values were 83.33 μ g \cdot m⁻³, 66.47 μ g \cdot m⁻³ and 62.07 μ g \cdot m⁻³, respectively. It appears valley in July 3, and the value was 23.21 μ g \cdot m⁻³.

The concentration of $PM_{2.5}$ in winter ranges from 27.40 μ g \cdot m⁻³ to 216.61 μ g \cdot m⁻³ with an average value of 116.28 μ g \cdot m⁻³. The concentration of $PM_{2.5}$ in summer ranges from 23.21 to 83.33 μ g \cdot m⁻³ with an average value of 46.80 μ g \cdot m⁻³. Winter $PM_{2.5}$ concentration was significantly higher than the summer, about 2.5 times than summer. As the Fuling is located in the mountain city of Chongqing, the static wind day is more, followed by winter fog weather is more frequent, is not conducive to the proliferation of pollutants, and more rain in summer, is conducive to the removal of particulate matter, resulting in $PM_{2.5}$ concentration in winter higher than summer.

According to Chinese current Ambient Air Quality Standard (GB3095-2012) [10], the average mass concentration of $PM_{2.5}$ in January and July 2015 is 81.54 μ g \cdot m⁻³, which is 2.3 times of the average annual upper limit. In the year of 2015, 62 samples of effective samples were collected, among which 21 exceeded the standard of $PM_{2.5}$ in the winter and exceeded rate 67.7% and 1.08 to 2.89 times exceeded. In summer the number of the upper limit of the secondary standard about the day average concentration is 2. Thus, winter atmospheric fine particulate pollution is much higher than in summer.

Seasonal variation of water-soluble ionic. In this study, F⁻, Cl⁻, SO₄²⁻, NO₃⁻, K⁺, Ca²⁺, Na⁺, Mg²⁺ and NH₄⁺ were analyzed by ICS-600 ion chromatograph, as shown in Table 1. In Fuling, The total concentration of water-soluble ions in $PM_{2.5}$ ranges from 6.37 to 86.30 μ g \cdot m⁻³ with an average of 34.69 μ g \cdot m⁻³. Water-soluble ionic components accounted for 42.28% of the total mass of $PM_{2.5}$ sample, showing that the water-soluble ionic component is an important component of $PM_{2.5}$.

From Table 1, the average values of water-soluble ions in Fuling area in January and July are: SO₄²⁻ > NO₃⁻ > NH₄⁺ > Cl⁻ > Na⁺ > K⁺ > Ca²⁺ > F⁻ > Mg²⁺, the concentration values are 14.48, 8.27, 8.00, 1.63, 1.28, 0.76, 0.41, 0.24 and 0.07 μ g \cdot m⁻³, Water-soluble ions accounted for 41.43%, 23.66%,

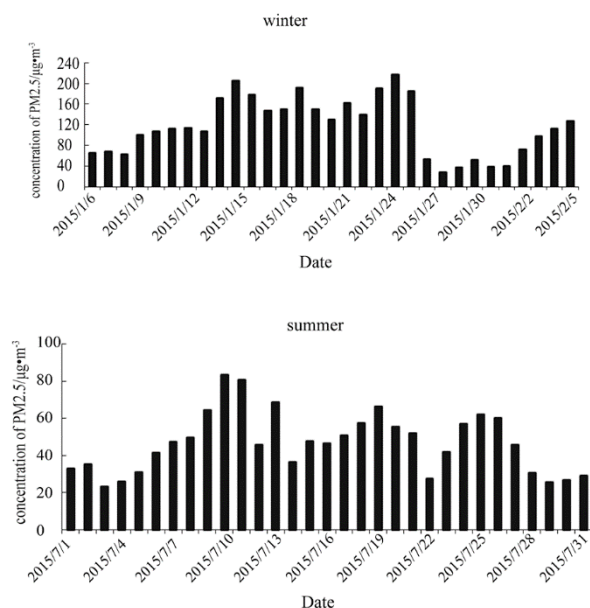


FIGURE 1
Mean daily variation of PM_{2.5} concentration in urban area of Fuling in 2015

TABLE 1
Concentration levels of water-soluble ions in PM_{2.5} (Unit: µg·m⁻³)

Analyzed series	winter			summer		
	maximum	minimum	average	maximum	minimum	average
Na ⁺	4.45	0.68	1.91	1.59	0.02	0.41
NH ₄ ⁺	20.27	2.30	12.00	10.40	0.30	3.86
K ⁺	2.88	0.16	1.29	0.48	0.05	0.21
Mg ²⁺	0.31	0.01	0.09	0.11	0.01	0.05
Ca ²⁺	2.34	0.01	0.55	0.44	0.07	0.18
F ⁻	1.20	0.02	0.38	0.30	0.01	0.09
Cl ⁻	12.71	0.12	2.94	3.69	0.32	0.95
SO ₄ ²⁻	43.91	3.86	18.63	26.51	3.30	10.20
NO ₃ ⁻	31.06	2.46	14.01	4.55	0.52	2.33

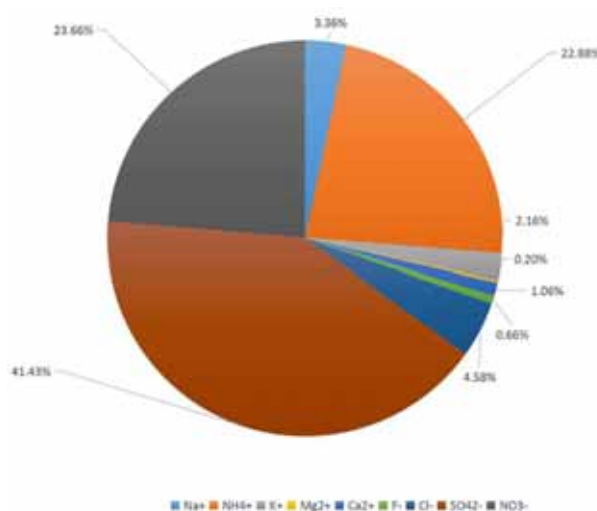


FIGURE 2
Percentage of water-soluble ions in total ions

22.88%, 4.58%, 3.36%, 2.16%, 1.06%, 0.66%, 0.20% of total water-soluble ions, respectively. Figure 2 shows the percentages of water-soluble ions in the total ions. The total mass concentration of SO_4^{2-} , NO_3^- and NH_4^+ was $30.53 \mu\text{g} \cdot \text{m}^{-3}$, accounting for 87.99% of total water-soluble ions and 35.97% of $\text{PM}_{2.5}$, indicating that SO_4^{2-} , NO_3^- and NH_4^+ were the most important water-soluble ingredients in $\text{PM}_{2.5}$, but also that more serious secondary pollution.

The concentrations of SO_4^{2-} , NO_3^- and NH_4^+ were the highest in winter, which appeared on Feb. 5, Jan. 16 and Jan. 25, whose value were 43.91, 31.06 and $20.27 \mu\text{g} \cdot \text{m}^{-3}$, respectively. Mass concentrations of $\text{PM}_{2.5}$ were 126.87, 178.18 and $216.61 \mu\text{g} \cdot \text{m}^{-3}$, respectively. The formation of SO_4^{2-} and NO_3^- is mainly due to the photo-chemical oxidation of SO_x and NO_x , which is susceptible to temperature, humidity and light intensity. Therefore, in contrast to the coastal city of Tianjin [11], (the secondary aerosol composition is summer > winter), while Fuling is winter > summer. In winter, the atmosphere structure is stable, the mixing layer height is relatively low, the cumulative effect is obvious, the pollutants accumulate in the atmosphere and spread slowly. Pollutant retention time is longer, but also makes primary pollutants prone to secondary transformation [12]. Fuling is located in the edge of Sichuan Basin, mostly mountainous, less rainfall and relatively high temperature in winter, is not conducive to the proliferation of pollutants, conducive to the formation of SNA.

Similar to water-soluble ion concentrations of $\text{PM}_{2.5}$ in Wanzhou, the seasonal distributions of water-soluble ionic components in $\text{PM}_{2.5}$ were all in winter > summer, and the average concentrations of SO_4^{2-} , NO_3^- and NH_4^+ are higher, about 1.12, 2.32, 1.30 times of Wanzhou, respectively [13]. This shows that pollution is more serious than Fuling, on the one hand is due to its serious pollution, on the other hand because of the main city of Fuling close to Chongqing, more vulnerable to the impact of the main city of pollutants.

The concentrations of Cl^- , Na^+ and Ca^{2+} in the primary aerosols were less than 2.5% of $\text{PM}_{2.5}$, which could reflect the characteristics of the source emission, and less rainfall in winter, the construction dust and ground dust caused by human activities, and long-distance dust lead to the three ions in winter are greater than the summer. The change of K^+ in $\text{PM}_{2.5}$ is mainly biomass burning, in winter ($1.29 \mu\text{g} \cdot \text{m}^{-3}$) which is about 6.14 times of that in summer ($0.212 \mu\text{g} \cdot \text{m}^{-3}$), which is related to the agricultural production in Fuling area. The maximum value of K^+ is $2.88 \mu\text{g} \cdot \text{m}^{-3}$ on January 26, the day Cl^- , Na^+ also has a higher value, most likely there is a straw burning activity. The total concentration of water-soluble ions in winter ranges from 10.82 to $86.30 \mu\text{g} \cdot \text{m}^{-3}$ with an average value of $51.08 \mu\text{g} \cdot \text{m}^{-3}$. Figure 3 shows the

daily variation of water-soluble ion concentration about $\text{PM}_{2.5}$ in winter.

The average daily average concentration of water-soluble ions in $\text{PM}_{2.5}$ was $85.66 \mu\text{g} \cdot \text{m}^{-3}$ on January 16 and $86.30 \mu\text{g} \cdot \text{m}^{-3}$ on January 25, and the $\text{PM}_{2.5}$ concentration was $178.18 \mu\text{g} \cdot \text{m}^{-3}$ and $216.61 \mu\text{g} \cdot \text{m}^{-3}$ respectively, which accounted for 48.08 and 39.84% of the $\text{PM}_{2.5}$ concentration on the same day. The minimum value was $10.82 \mu\text{g} \cdot \text{m}^{-3}$ on January 28, accounting for 39.49% of the $\text{PM}_{2.5}$ concentration. The concentration of $\text{PM}_{2.5}$ on the day of sampling was $27.40 \mu\text{g} \cdot \text{m}^{-3}$. Since February 6, no sampling, it cannot determine the February 5 water-soluble ions are peak, but the day the proportion of water-soluble ions accounted for the largest concentration of $\text{PM}_{2.5}$, about 62.18%, concentration of $\text{PM}_{2.5}$ on the day was $126.87 \mu\text{g} \cdot \text{m}^{-3}$.

Figure 4 shows the ratio of water-soluble ions in $\text{PM}_{2.5}$ in winter. The concentration of water-soluble ions in winter is in the order of $\text{SO}_4^{2-} > \text{NO}_3^- > \text{NH}_4^+ > \text{Cl}^- > \text{Na}^+ > \text{K}^+ > \text{Ca}^{2+} > \text{F}^- > \text{Mg}^{2+}$, and the values are 18.63, 14.01, 12.00, 2.30, 1.91, 1.29, 0.59, 0.38, $0.09 \mu\text{g} \cdot \text{m}^{-3}$, the percentages of the ions are 36.48%, 27.44%, 23.50%, 4.36%, 3.73%, 2.52%, 1.08%, 0.73% and 0.17% respectively. So the proportion of SO_4^{2-} , NO_3^- and respectively, is 86.53%, which is the biggest contribution of water-soluble ions. Water-soluble ions on January 27, a sharp decline, mainly rainy weather on the role of pollutants removal, so that rapid reduction.

The total concentration of water-soluble ions in summer ranges from $6.37 \mu\text{g} \cdot \text{m}^{-3}$ to $43.48 \mu\text{g} \cdot \text{m}^{-3}$, with an average of $18.30 \mu\text{g} \cdot \text{m}^{-3}$. Figure 5 shows the daily variation of water-soluble ion concentration in $\text{PM}_{2.5}$ in summer.

In summer the daily maximum concentration of water-soluble ions in $\text{PM}_{2.5}$ was $43.48 \mu\text{g} \cdot \text{m}^{-3}$ on July 10, accounting for 52.18% of $\text{PM}_{2.5}$ concentration, and the $\text{PM}_{2.5}$ concentration on the day was $83.33 \mu\text{g} \cdot \text{m}^{-3}$. The lowest concentration of $\text{PM}_{2.5}$ on July 29 was $6.37 \mu\text{g} \cdot \text{m}^{-3}$. On the day of sampling, the mass concentration of $\text{PM}_{2.5}$ was $25.62 \mu\text{g} \cdot \text{m}^{-3}$, accounting for 24.82% of the mass concentration of $\text{PM}_{2.5}$ on the same day.

In the summer, the water-soluble ions were in the order of $\text{SO}_4^{2-} > \text{NH}_4^+ > \text{NO}_3^- > \text{Cl}^- > \text{Na}^+ > \text{Ca}^{2+} > \text{K}^+ > \text{F}^- > \text{Mg}^{2+}$, the average concentration values were 10.20, 3.86, 2.33, 0.95, 0.50, 0.22, 0.21, 0.09, $0.05 \mu\text{g} \cdot \text{m}^{-3}$. The percentages of water-soluble ions were 55.72%, 21.11%, 12.75%, 5.21%, 2.29%, 1.13%, 1.02%, 0.47% and 0.28% respectively. Figure 6 shows the proportion of water-soluble ions in $\text{PM}_{2.5}$ in summer. It can be seen, SO_4^{2-} , NH_4^+ and NO_3^- are still the main component of water-soluble ions. The water-soluble ions decreased sharply on July 12, mainly due to rainfall and rainfall, which was favorable for the removal of particulate pollutants.

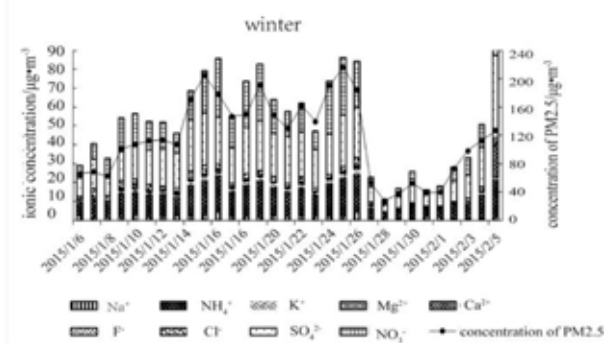


FIGURE 3
Daily variation of water-soluble ion concentration about PM_{2.5} in winter

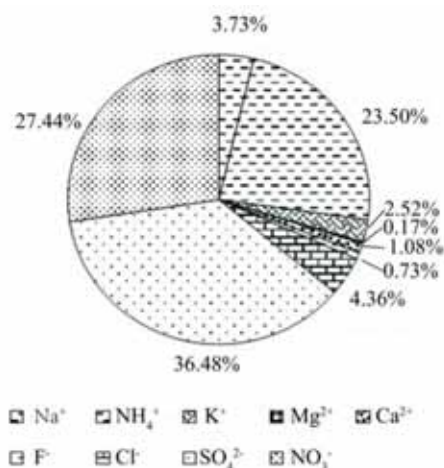


FIGURE 4
Percentage of water-soluble ions in winter

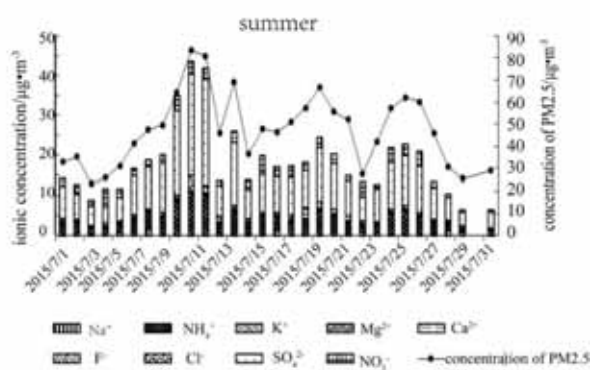


FIGURE 5
Daily average of water-soluble ions in summer

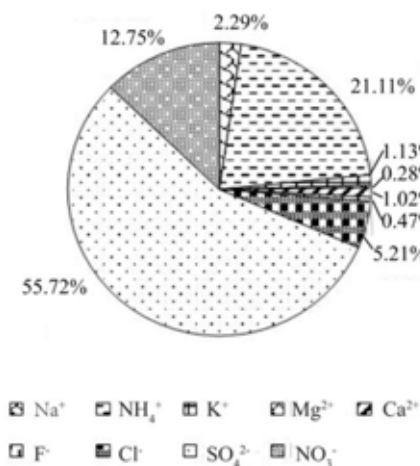


FIGURE 6
Percentage of water-soluble ions in summer

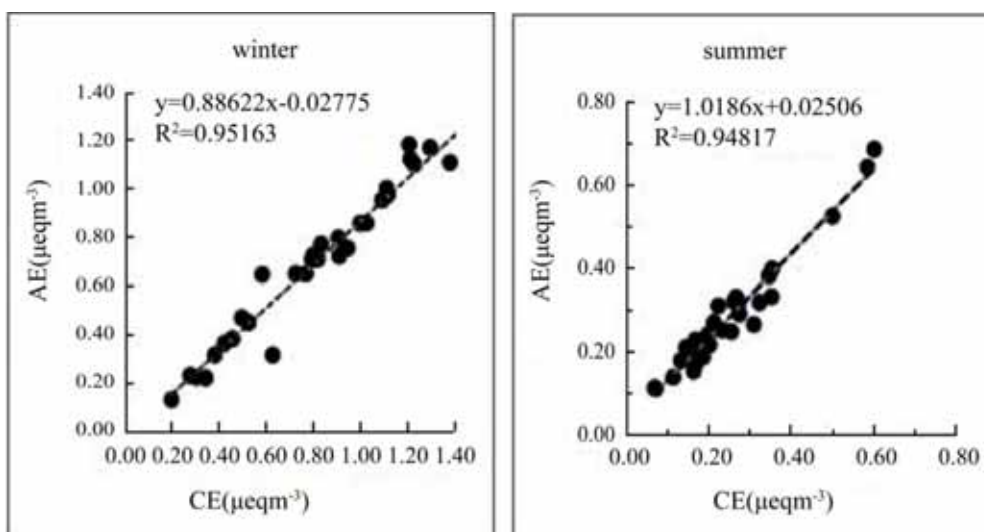


FIGURE 7
Balance of anions and cations

TABLE 2
Correlation of water-soluble ion

	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	F ⁻	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻
Na ⁺	1	-0.314	-0.301	0.174	0.399	0.051	-0.278	-0.257	-0.365
NH ₄ ⁺	0.539	1	0.840	-0.006	-0.359	-0.203	0.583	0.983	0.509
K ⁺	0.658	0.900	1	0.067	-0.393	-0.167	0.510	0.843	0.362
Mg ²⁺	0.774	0.555	0.580	1	0.317	-0.587	-0.198	-0.033	0.109
Ca ²⁺	0.473	-0.003	0.092	0.359	1	0.011	-0.450	-0.413	-0.003
F ⁻	0.178	0.320	0.147	0.131	-0.051	1	0.018	-0.173	-0.376
Cl ⁻	0.371	0.227	0.239	0.286	0.474	0.082	1	0.571	0.137
SO ₄ ²⁻	0.401	0.926	0.724	0.384	-0.076	0.442	0.180	1	0.395
NO ₃ ⁻	0.566	0.869	0.889	0.670	0.110	0.101	0.156	0.634	1

Note: the lower left corner for the winter, the upper right corner for the summer

Balance of anions and cations and acidity and alkaline of particular matter.

$$\text{Cation(CE)} = \frac{Na^+}{23} + \frac{NH_4^+}{18} + \frac{K^+}{39} + \frac{Mg^{2+}}{12} + \frac{Ca^{2+}}{20}$$

$$\text{Anion(AE)} = \frac{SO_4^{2-}}{48} + \frac{NO_3^-}{62} + \frac{Cl^-}{35.5} + \frac{F^-}{19}$$

It can be seen from Figure 7 that the correlation of anions and cations is very good, and R^2 in both winter and summer is equal to 0.95, indicating that the sample data are valid and the ions analyzed are the main components of PM_{2.5}. According to the slope of the equation, the slope is less than 1 in winter, the overall level of ions is alkaline; the slope is greater than 1 in summer, the overall is acidic. Although the average concentration of SO₄²⁻, NO₃⁻ and NH₄⁺ in winter is higher than that in summer, the dust is not conducive to the diffusion due to the less rain and windy days in winter, and the dust, soil dust and crop incineration are not conducive to diffusion and can neutralize the acid aerosol, resulting in the ions are alkaline in winter. In summer, the temperature is high and rainy, and the weather changes frequently, which is favorable for the chemical reaction of SO_x and NO_x, resulting in the ions are acidity in summer.

The relationship between the water-soluble ions. Table 2 for the water-soluble ion correlation coefficient matrix. The correlation between SO₄²⁻ and NO₃⁻ in winter (0.634) is better than summer (0.395) indicates that the two have good homology in winter, both from the combustion of fossil fuels and the conversion of SO_x and NO_x from motor vehicles. The correlation between NH₄⁺ and SO₄²⁻ was good in winter and summer, but it was slightly higher in summer than in winter, 0.983 in summer and 0.926 in winter. Indicating that the meteorological conditions in summer, the source of emission characteristics more conducive to their formation. The correlation between NH₄⁺ and NO₃⁻ was less obvious in winter than in winter. Thus, NH₄⁺ in the winter and easy to NO₃⁻ combination; in the summer and SO₄²⁻ priority.

The correlation between Cl⁻ and NH₄⁺ in the summer was 0.583, indicating that excess NH₄⁺ would combine with Cl⁻ to form NH₄Cl. Cl⁻ and Na⁺, SO₄²⁻, NO₃⁻ are poor, indicating that the homology is not obvious, and Cl⁻ not from coal and motor vehicle emissions, may come from biomass combustion. The correlation between Cl⁻ and K⁺ in summer is 0.51, indicating that KCl exists in summer PM_{2.5}.

CONCLUSIONS

In this paper, the data in two seasons of Chongqing Fuling in 2015 were collected to study the water-soluble ions in Fuling area. The diurnal and seasonal changes of water-soluble ions, the anion and cation balance, and correlation of water-soluble ions were analyzed. The results reflected in a certain extent that PM_{2.5} pollution characteristics. Mastering the PM_{2.5} pollution status, distribution characteristics and temporal and spatial variation of Fuling PM_{2.5} are of great significance to comprehensively understand the atmospheric pollution in Fuling area.

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The authors declare no conflict of interest.

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