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Characteristics of $PM_{2.5}$ in rural areas of southern Jiangsu Province, China

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Abstract: To understand pollution level and possible sources of atmospheric fine particulates in rural areas of southern Jiangsu Province of China, samples of $PM_{2.5}$ were collected and analyzed in Xueyan Town and Taihu Lake Station over three seasons from July 2002 to January 2003. The mass concentrations of $PM_{2.5}$ and 14 principal component elements were obtained. The results showed that pollution of $PM_{2.5}$ was serious and the concentration levels of S, Zn, Pb and As were similar to city. There are different seasonal distribution laws of pollutant elements in $PM_{2.5}$ between two sampling sites, probably due to contribution of local sources, medium or long distance transportation of fine particulates and complicated meteorological conditions. The enrichment levels of S, Zn, Pb, As, K were high, reflecting the influence of anthropogenic activities. Particularly enrichment level of S was much higher in summer, which was probably related to meteorological condition. The result of principal components analysis showed major sources of $PM_{2.5}$ included crustal re-suspension, coal burning, metal processing industry or waste incineration, vehicular emission, which suggests anthropogenic activities is of important influence on $PM_{2.5}$ in rural areas of southern Jiangsu Province.

Keywords: rural areas; $PM_{2.5}$; sources; enrichment factor; principal components analysis

Introduction

A number of studies abroad have demonstrated that atmospheric fine particulate matter($PM_{2.5}$, $d_p < 2.5$ μm) is not only associated with atmospheric visibility reduction but also has a clear correlation with the number of daily deaths and hospitalizations as a consequence of pulmonary disease (Chan, 1997; Christoforous, 2000; Schwartz, 1996; Wilson, 1997). On 18 July, 1997, the U.S. Environmental Protection Agency (USEPA) promulgated new and severe standards for fine particulates ($PM_{2.5}$) with the size below 2.5 μm (15 $\mu g/m^3$ as annual average, 65 $\mu g/m^3$ as 24 h average). In a long time, the study of atmospheric particulates is concentrated on total suspended matter(TSP) and inhalable particulates (PM_{10}, $d_p < 10$ μm) in China. Relatively limited data are available on concentration and composition of fine particulates. Until recent several years the study on $PM_{2.5}$ was carried out in a few cities. For instance, a joint study sponsored by the USEPA determined the concentration of PM_{10} and $PM_{2.5}$ in Lanzhou, Chongqing, Wuhan, Guangzhou(Wei, 1999). The common finding from these studies was that the ambient aerosol concentrations in these cities were generally in the 100—500 $\mu g/m^3$ ranges with the highest concentration in the winter months. A study in Nanjing showed that the concentrations of PM_{10} and $PM_{2.5}$ were of great difference in different environmental function areas of city(Wang, 2002). However, there is some lack of knowledge on $PM_{2.5}$ in rural areas with a relative small quantity of pollution sources. The purpose of this study was to find $PM_{2.5}$ pollution status and composition characteristics of elements in rural areas of southern Jiangsu Province, China. This information is helpful to determine the possible sources of $PM_{2.5}$. The paper analysed $PM_{2.5}$ samplers in Xueyan Town and Taihu Lake Station by X-ray fluorescence spectrometry (XRF), and discussed characteristics of principal component elements in $PM_{2.5}$ and possible sources of $PM_{2.5}$.

1 Materials and methods

1.1 Study area and sampling sites

The area of southern Jiangsu Province, China is located on lower reaches of Yangtze River, between 30°45′—32°18′N latitude and 118°24′—112°20′E longitude. It belongs climate of subtropical monsoon, and it is of rich rainwater. In rural areas of southern Jiangsu Province, industry and agriculture develop quickly, which causes village- and town-owned enterprises being the principal part of rural economy. Sampling was conducted at two sites in rural areas of southern Jiangsu Province (Fig. 1). One is located on the campus of an elementary school near center of Xueyan Town, another is located on the Taihu Lake Observation Station of Chinese Academy of Sciences. The sampling site at Xueyan Town is near a major road. A big chemical plant and a few small plants lie in the southeast of this sampling site. The sampling site at Taihu Lake Station is located in a remote rural area and is approximately 30 km away from center of city. A few residents and factories are scattered around this sampling site.

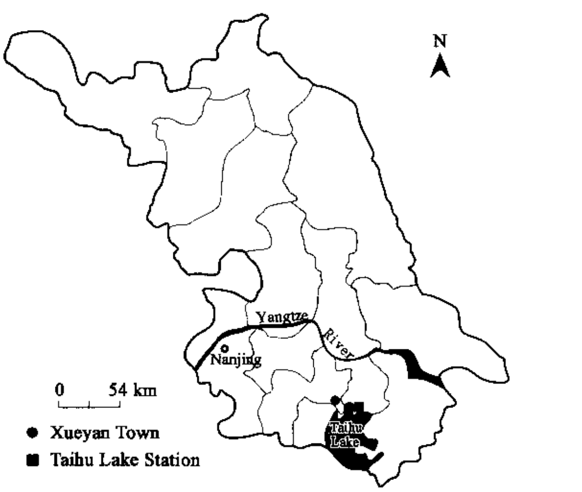

Fig. 1 Location of $PM_{2.5}$ sampling sites

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1.2 Sampling and analysis

The sampling was carried out at a constant flow rate of 28.3 L/min using an Anderson impactor pre-separator (Anderson Co. USA) that cuts off particles($d_p > 2.5$ μm). The data set was collected on 37 different days with a 12 h sampling period(from 7:00 a.m. to 7:00 p.m.) over three seasons from 15 July 2002 to 14 January 2003. The over-chlorine ethane filter(80 mm diameter) was used to collect $PM_{2.5}$ samplers in this study. In order to evaluate the blanks of filters, series of filter were analyzed in the same way as samplers to check the trace elements impurity. Except element K, the concentrations of other elements are below detection limits(Table 1), which shows it is feasible to use over-chlorine ethane filter to collect and analyze $PM_{2.5}$ samplers.

Table 1 Average element concentration of blank filter(μg/filter)

	Na	Mg	Al	Si	P	K	Ca	Ti	Mn	Fe	S	Zn	Pb	As
Blank filter	–	–	–	–	–	0.08	–	–	–	–	–	–	–	–

Sampler was placed on the roofs of 5 m tall buildings, resulting in an effective inlet height above the ground of about 6 m. There are no large buildings around the sampling sites to disrupt wind flow patterns, and the sampling was selected in fine days to avoid the influence of rainy days and snowy days.

The filters were weighted on an analytical balance (Sartorius 2004MP) with a reading precision of 10 μg after humidity equilibrium (> 24 h) in the desiccator, then exposed to the atmosphere and recorded the sampling time. At the end of sampling, the filters were taken back to the laboratory. After humidity equilibrium(> 24 h), the filters were reweighted on the same balance to calculate the mass concentration of $PM_{2.5}$. Elements concentrations were determined by X-ray fluorescence(XRF, Shimadzu VF320, Japan) using standard methods(Chow, 1994).

2 Results and discussion

2.1 $PM_{2.5}$ mass concentration

Average concentration of $PM_{2.5}$ in three seasons at two different sites is given in Table 2. The maximal value (221.10 μg/m^3) appeared in winter at Xueyan Town, and the minimal value(53.66 μg/m^3) appeared in fall at Taihu Lake Station. Among the two sampling sites, the $PM_{2.5}$ mass concentration at Xueyan Town is always higher than that at Taihu Lake Station, and the difference is obvious in fall, which is caused by influence of violent anthropogenic activities. In this study, the mass concentrations of $PM_{2.5}$ samples are above the 24 h $PM_{2.5}$ U.S. National Ambient Air Quality Standard(65 μg/m^3) at 92%, and the average $PM_{2.5}$ concentration in the sampling period is well above the annual U.S. National Ambient Air Quality Standard(15 μg/m^3). At two sampling sites, concentration was higher during the winter, decreases through the summer, and tends to be the lowest during fall. The $PM_{2.5}$ mass concentration peak in winter is most likely due to unfavorable meteorological conditions. The results show that $PM_{2.5}$ pollution level in rural areas of Southern Jiangsu Province is relatively high when compared with other studies in China(Wei, 1999; Wang, 2002a).

Table 2 $PM_{2.5}$ mass concentration and element concentration (μg/m^3)

	Summer		Fall		Winter	
	Xueyan Town(XY)	Taihu Lake Station (TLS)	Xueyan Town (XY)	Taihu Lake Station (TLS)	Xueyan Town (XY)	Taihu Lake Station(TLS)
$PM_{2.5}$	144.38 ± 39.43	109.40 ± 53.67	119.91 ± 41.46	66.75 ± 13.09	151.68 ± 69.42	140.16 ± 72.14
Na	0.91 ± 0.17	0.75 ± 0.10	0.78 ± 0.16	0.92 ± 0.16	1.24 ± 0.34	1.28 ± 0.18
Mg	0.17 ± 0.04	0.10 ± 0.05	0.36 ± 0.14	0.18 ± 0.01	0.32 ± 0.12	0.42 ± 0.12
Al	1.10 ± 0.26	1.18 ± 0.32	2.04 ± 0.634	0.95 ± 0.25	2.56 ± 0.72	2.11 ± 1.04
Si	2.33 ± 0.50	2.72 ± 0.68	4.89 ± 1.55	2.05 ± 0.50	5.15 ± 1.43	5.88 ± 2.52
P	0.071 ± 0.043	0.049 ± 0.019	0.072 ± 0.012	0.049 ± 0.028	0.13 ± 0.03	0.21 ± 0.17
K	3.06 ± 2.65	4.10 ± 2.00	5.37 ± 2.14	2.30 ± 1.26	5.90 ± 2.47	4.29 ± 1.51
Ca	0.85 ± 0.44	0.77 ± 0.45	1.12 ± 0.48	0.48 ± 0.22	1.80 ± 0.87	2.40 ± 2.05
Ti	0.060 ± 0.029	0.046 ± 0.016	0.074 ± 0.020	0.032 ± 0.008	0.14 ± 0.06	0.12 ± 0.07
Mn	0.056 ± 0.032	0.054 ± 0.022	0.02 ± 0.006	0.063 ± 0.059	0.056 ± 0.037	0.10 ± 0.065
Fe	0.92 ± 0.63	0.71 ± 0.25	0.88 ± 0.27	0.83 ± 0.61	1.58 ± 0.58	1.39 ± 0.75
S	10.58 ± 6.35	10.58 ± 3.67	8.46 ± 3.31	5.79 ± 0.99	12.07 ± 3.42	8.16 ± 2.10
Zn	0.57 ± 0.36	0.60 ± 0.25	0.48 ± 0.41	0.52 ± 0.43	0.50 ± 0.24	0.53 ± 0.24
Pb	0.19 ± 0.10	0.38 ± 0.10	0.44 ± 0.30	0.17 ± 0.12	0.26 ± 0.08	0.30 ± 0.17
As	0.049 ± 0.028	0.040 ± 0.009	0.033 ± 0.005	0.057 ± 0.030	0.14 ± 0.03	0.08 ± 0.019

2.2 Element concentration in $PM_{2.5}$

Average concentration of fourteen elements in $PM_{2.5}$ in three seasons at two sampling sites is shown in Table 2. Element composition in $PM_{2.5}$ is similar at two sampling sites, and the main elements contain S, Si, K, Ca, Al, Na and Fe. Among the fourteen elements, the concentration of S is obviously higher than other elements. The sum of S, Si, K, Ca, Al, Na and Fe accounts for 93%—95% of total mass of fourteen determined elements at two sites and the concentration of other elements is relatively low. These trace elements mainly contain Zn, Pb, Mg and As. Similar results on $PM_{2.5}$ were also found from other studies in Beijing(Wang, 2002b; Yang, 2002), which indicates that high concentration of S in $PM_{2.5}$ is a typical characteristic both in north cities and in rural areas of southern Jiangsu Province in China. When compared with other study in four cities in China(Wei, 1999), the concentrations of S, Zn, Pb and As are close to them, even in summer and winter the concentrations are slightly higher than them. This result shows that $PM_{2.5}$ pollution in rural areas of southern Jiangsu Province is relatively serious.

As shown in Fig. 2, annual concentrations of most elements in $PM_{2.5}$ at Xueyan Town are slightly higher than those at Taihu Lake Station, which is likely due to influence of local sources. The sampling site at Xueyan Town is located near a major road, and surrounded by a few village- and town-owned enterprises. Local sources nearly exists at Taihu Lake Station, so $PM_{2.5}$ is likely associated with contribution of non-local sources.

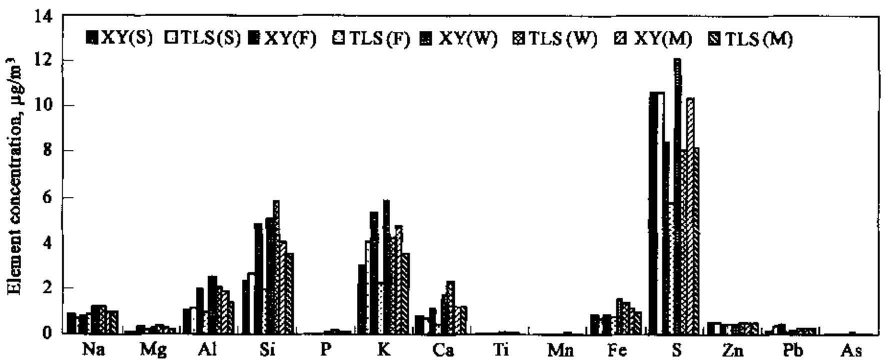

Fig.2 Seasonal variations of element concentration in $PM_{2.5}$ at two sampling sites: (1) Xueyan Town(XY), (2) Taihu Lake Station(TLS)
S. summer; F. fall; W. winter; M. annual

Seasonal variation of elements composition in $PM_{2.5}$ at Xueyan Town is found, with the higher elemental concentration in winter and the lower in summer and fall (Fig.2). Seasonal variation of most elements at Taihu Lake Station is similar with that at Xueyan Town, but the highest concentrations of non-crustal elements S, Zn, Pb appeared in summer. Seasonal variation of elements in $PM_{2.5}$ at two sampling sites is most likely related to local sources and meteorological condition. In winter, thick thermal inversion layer forms frequently, which affects dispersion of $PM_{2.5}$ at Xueyan Town. In summer, good meteorological condition is helpful to medium or long distance transportation of $PM_{2.5}$ and photo-chemical reaction (Song, 2002), which enhances the contribution of non-local sources at Taihu Lake Station.

2.3 Enrichment factor

The crustal enrichment factor method has commonly been used as a first step in attempting to evaluate the strength of the crustal and non-crustal sources (Gao, 1992). The Enrichment factor for any element i relative to crustal material is defined by $EF = (Ci/Cr)_p/(Ci/Cr)_r$. Where EF is the enrichment factor of i; r is a reference element for crustal element and $(Ci/Cr)_p$ is the concentration ratio of i to r in the $PM_{2.5}$ sample, and $(Ci/Cr)_r$ is the average concentration ratio of i to r in the crust.

To determine the strength of crustal and non-crustal sources for elements associated with $PM_{2.5}$, the enrichment factor was calculated for each element based on samples collected at two sites (Fig.3). We take Al as the reference element in this study based on chemical composition of the earth crust, assuming minor contributions of pollutant Al. Fig.3 indicates that Enrichment factors of Na, Mg, Al, Si, P, Ca, Ti, Mn, and Fe in $PM_{2.5}$ are less than 10, suggesting that these elements are not enriched in $PM_{2.5}$ and their dominant sources are crustal sources. K, S, Zn, Pb, As obviously have high enrichment factors more than 10, even average annual values of S, Pb, As are above 1000, which suggests that these elements are likely related to anthropogenic activities. Very similar level of enrichment factors at two sites shows that spatial variation of enrichment factors is not obvious. Among the elements, S has relatively higher enrichment factor in summer than in fall and winter. In areas of southern Jiangsu Province, there is no heating period in winter and coal consumption does not increase accordingly, which does not affect enrichment level of S. Many studies indicated that the source of S in $PM_{2.5}$ is strongly associated with SO_4^{2-}, and continued high temperature weather in summer is helpful to photochemical reaction to produce SO_4^{2-} (Song, 2002). So meteorological condition is likely the main factor to cause high enrichment of S in summer.

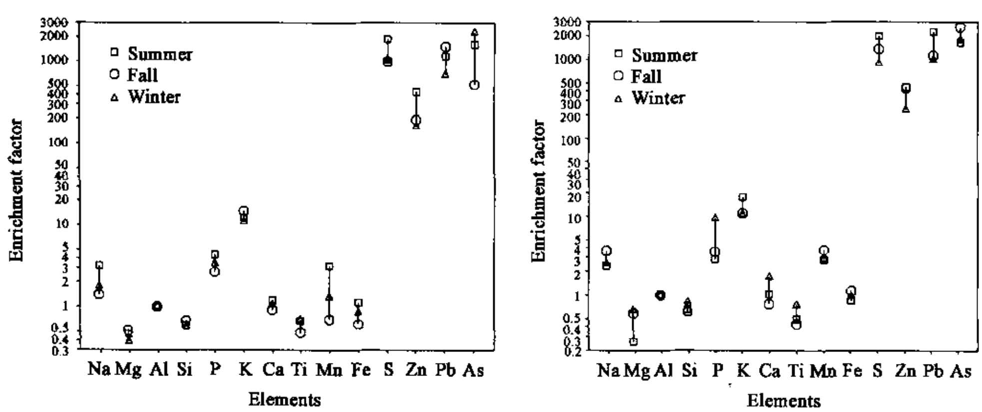

Fig.3 Comparison of enrichment of elements in $PM_{2.5}$
The left is Xueyan Town, the right is Taihu Lake Station

2.4 Principal components analysis

Principal components analysis (PCA) was used to identify the possible sources of $PM_{2.5}$. PCA was executed to total $PM_{2.5}$ samples at two sites by the Rotated Factor Matrix method in SPSS 11.0. Several combinations of variables were made to extract principal component and the best possible results were obtained after varimax rotation (Table 3). A total of four principal components were extracted, which explained 92.34% of the variance in the data set.

Table 3 Results of principal component analysis with varimax rotation

Variable	Factor 1	Factor 2	Factor 3	Factor 4
Na	0.753	0.078	0.405	-0.383
Mg	0.925	0.072	-0.118	0.126
Al	0.931	0.276	0.165	0.094
Si	0.936	0.205	0.094	0.225
P	0.673	0.644	0.167	-0.144
K	0.514	0.694	0.216	0.312
Ca	0.755	0.541	0.268	0.081
Ti	0.818	0.445	0.258	-0.108
Mn	0.085	0.055	0.969	-0.065
Fe	0.648	0.400	0.597	-0.033
S	0.126	0.963	0.033	0.025
Zn	0.106	0.119	0.784	0.533
Pb	0.225	0.147	0.161	0.901
As	0.508	0.466	0.048	-0.602
Percentage of variance, %	42.02	20.43	16.91	12.98
Cumulative variance, %	42.02	62.45	79.36	92.34

Principal component 1 is strongly associated with crustal elements of Mg, Al, Si and Ti, which suggest it represents crustal re-suspension. The principal factor 2 has higher correlation with S than that with other elements, so it likely represents coal burning source. The third principal component has high loadings for Mn, Zn. As stated in above enrichment analysis, the enrichment factor of Mn is more than 1 and less than 10, which suggests that Mn has been affected by anthropogenic activities and enriches slightly in $PM_{2.5}$. This factor may be associated with metal processing industry or waste incineration. The fourth principal component is strongly associated with Pb and represents vehicular emission. Through above analysis, the possible sources of $PM_{2.5}$ in rural areas of southern Jiangsu Province include crustal re-suspension, coal burning, metal processing industry or waste incineration, vehicular emission, and contributions of four sources are 42.02%, 20.43%, 16.91%, 12.98% respectively. The result shows that $PM_{2.5}$ is not only associated with crustal re-suspension but also relates with anthropogenic activities strongly. On the other hand, the source of $PM_{2.5}$ is not only probably due to contribution of local sources, but also affected by medium or long distance transportation of fine particulates.

3 Conclusions

In rural areas of southern Jiangsu Province, the maximal value (221.10 $\mu g/m^3$) of $PM_{2.5}$ appeared in winter at Xueyan Town, and the minimal value (53.66 $\mu g/m^3$) of $PM_{2.5}$ appeared in fall at Taihu Lake Station. The $PM_{2.5}$ mass concentration at Xueyan Town was always higher than that at Taihu Lake Station. The $PM_{2.5}$ mass concentration was higher during the winter, decreases through the summer, and tends to be the lowest in the fall. The $PM_{2.5}$ mass concentration peak in winter is most likely due to unfavorable meteorological conditions. When compared with other studies in China, the result shows that $PM_{2.5}$ pollution level in rural areas of Southern Jiangsu Province is relatively high.

Element composition in $PM_{2.5}$ is similar at two sampling sites, and the main elements contain S, Si, K, Ca, Al, Na and Fe. Among the fourteen elements, the concentration of S was obviously higher than other elements. The sum of S, Si, K, Ca, Al, Na and Fe accounted for 93%—95% of total mass of fourteen determined elements at the two sites. The concentrations of elements are slightly higher than study results in four cities in China, suggesting $PM_{2.5}$ pollution in rural areas of Southern Jiangsu Province is relatively serious.

Annual concentrations of most elements in $PM_{2.5}$ at Xueyan Town are slightly higher than those at Taihu Lake Station. Seasonal variation of elements composition in $PM_{2.5}$ at Xueyan Town was found, with the higher elemental concentration in the winter and the lower in the summer and fall. The spatial and seasonal variation is likely due to contribution of local source, medium or long distance transportation of fine particulates and complicated meteorological condition.

Enrichment factors of K, S, Zn, Pb, As were obviously more than 10, even average annual values of S, Pb, As are above 1000, which suggests that these elements are likely related to anthropogenic activities. S had relatively higher enrichment factors in summer than in fall and winter, which is likely affected by meteorological condition.

Through principal components analysis, the possible sources of $PM_{2.5}$ in rural areas of southern Jiangsu Province include crustal re-suspension, coal burning, metal processing industry or waste incineration, vehicular emission. Among the four sources, crustal re-suspension accounts for 42.02%, which suggests that the source of $PM_{2.5}$ is likely related to anthropogenic activities strongly.

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