CHEMICAL COMPOSITION OF AEROSOLS IN THE WEST COAST OF TAIWAN STRAIT, CHINA

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ABSTRACT:

The chemical composition of aerosols was investigated using regular environmental air quality observation, a single particle aerosol mass spectrometer (SPAMS 0515) and an ambient ion monitor (URG 9000D) in Xiamen in 2018. The results showed that the annual average mass concentrations of $PM_{2.5}$ was 22 μg/m^3, and concentrations of water-soluble inorganic ions was 9.94 μg/m^3 which accounted for 45.2% of $PM_{2.5}$. SO_4^{2-}, NO_3^- and NH_4^+ were main components of secondary reactions which contributed more than 77 percent of water-soluble inorganic ion concentration. As a coastal city, Cl^- and Na^+ contributed 13.9 percent of water-soluble inorganic ion concentration. Based on single particle aerosol mass spectrometer analysing, mobile sources emission was the most important sources of particle matter which contributed over 30%.

1. INTRODUCTION

In recent decades, rapid economic development and continuous progress of industrialization and urbanization in China have led to a rapid increase in the emission of air pollutants. Artificial factors, such as energy consumption, industrial production and urban construction, combined with natural factors, such as dust storms and climate change, have led to frequent air pollution events (Yin et al., 2014). With the public increasingly concerned about air pollution, the government has vigorously promoted air pollution prevention and control since 2013. Urban air quality has improved significantly since the implementation of the air pollution prevention and control action plan. Under this background, it is urgent to manage the emission sources of air pollution scientifically and pertinently. Chemical component analysis of particulate matter and pollution source tracing can provide strong technical support for the requirements of the management department.

Water-soluble inorganic ions are important chemical components of $PM_{2.5}$, and a large number of studies have been carried out around the world (Deng et al., 2016; Huang et al., 2016; Liang et al., 2018; Luo et al., 2018; Masiol et al., 2015; Meng et al., 2016; Ming et al., 2017; Niu et al., 2016; Saxena et al., 2017; Szigeti et al., 2015; Tolis et al., 2015; Zhang et al., 2019). However, due to differences in geographical location, meteorological conditions, energy structure and industrial layout, results of these studies varied greatly. An et al., (2018) found that sulfate, nitrate and ammonium were the major components of secondary particles in the atmosphere and made important contributions to the aerosol extinction coefficient. Wang et al. (2016) found that different water-soluble inorganic ions had distinct diurnal variations.

In this study, the concentrations of water-soluble inorganic ions in Xiamen were obtained using Thermo Fisher Scientific URG 9000D in 2018. The concentrations and seasonal variations of water-soluble inorganic ions are showed in this paper. The results can improve our understanding of $PM_{2.5}$ pollution in Xiamen and provide reference for improving environmental air quality in Xiamen.

2. METHODOLOGY

2.1 Site Description and Sampling

Continuous particulate mass concentrations and chemical compositions were measured at one location in Xiamen, China. Xiamen, located on the west coast of the Taiwan Strait, has a population of 4.11 million and an area of 1699.39 km^2. The sampling site is set on the rooftops of the Xiamen Atmospheric Environment Monitoring Superstation (XAEMS; 118.1402°E, 24.4386°N). XAEMS, in a residential area, is located in the south of Xiamen Island, about 700 m from the coastline (Figure 1).

An ambient particulate matter monitor (Thermo Fisher Scientific TEOM Model 1405-DF), a single particle aerosol mass spectrometer (SPAMS 0515) and an ambient ion monitor (Thermo Fisher Scientific URG 9000D) were used to continuously monitor the mass concentrations of PM_{10} and $PM_{2.5}$, mass concentrations of nine water-soluble inorganic ions (Na^+, Mg^{2+}, K^+, Ca^{2+}, NH_4^+, F-, Cl^-, NO_3^- and SO_4^{2-}), and single particle aerosol chemical composition. The data were collected hourly by the data acquisition system.

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Figure 1. Geographical location map of Xiamen city (a) and XAEMS (b)

2.2 Quality Assurance and Control

The TEOM Model 1405-DF (Thermo Fisher Scientific, Waltham, MA) ambient particulate matter monitor is equipped with a Filter Dynamic Measurement System (FDMS), which can correct for semi volatile species evaporation from mass measurement filter surface. The instruments were operated at flow rates prescribed by manufacturer: 1.67 L/min ($PM_{10-2.5}$), 3 L/min ($PM_{2.5}$), and 12 L/min (bypass). TEOM instrument maintenance was performed every 20 days and consists of changing TEOM and FDMS filters, cleaning the PM_{10} inlet, checking for seal leaks in the mass transducer, flow audit and calibration, and instrument leak check.

The URG 9000 (Thermo Fisher Scientific) and SPAMS 0515 maintenance were performed monthly and consists of cleaning the inlet and tube, instrument leak check, etc.

To assure the highest quality data were used for analysis, the status log was filled out before and after maintenance to assure that the instrument conditions did not change due to operator intervention.

3. RESULTS

3.1 $PM_{2.5}$

Daily concentration of $PM_{2.5}$ at XAEMS in 2018 were in a range of 0.003~0.082 mg/m^3, with an average and standard deviation of 0.022±0.012 mg/m^3, reaching the China National Ambient Air Quality Standards (GB3095-2012) (grade II, 0.035 mg/m^3). The maximum monthly average concentration of $PM_{2.5}$ occurred in February (0.034 mg/m^3), and the maximum daily average concentration of $PM_{2.5}$ occurred in March (0.082 mg/m^3), reaching the light pollution level of Air Quality Index (Figure 2). The seasonal concentration of $PM_{2.5}$ in an order of winter > spring > autumn > summer, which can be explained by meteorological conditions. Higher $PM_{2.5}$ concentration during winter and spring might be caused by stable atmospheric layer and lower mixing height. The stable atmosphere is conducive to the accumulation of air pollutants. In addition, heavy $PM_{2.5}$ pollution occurs frequently in eastern China during winter and spring, and regional transmission of air pollutants has a great impact on local air quality. On the contrary, the dominant wind direction in Xiamen is southerly during summer, which is conducive to the diffusion of local air pollutants.

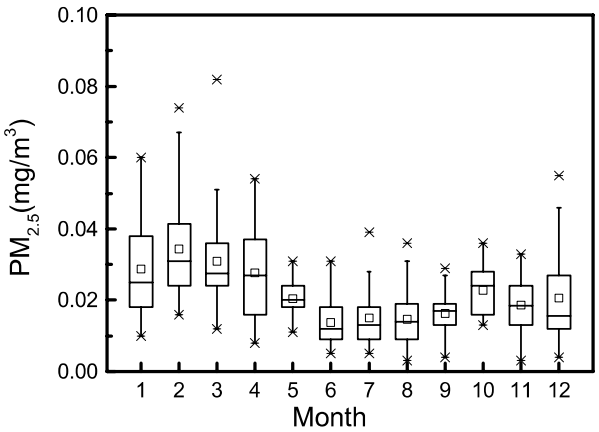

Figure 2. Monthly concentration of $PM_{2.5}$
(The upper dash indicate the maximum, the lower dash indicate the minimum, the upper "X" indicate the 99% value, the lower "X" indicate the 1% value, the small square indicate the mean value, the upper edge of the box indicate the 75% value, the lower edge of the box indicate the 25% value, the horizontal line in the box indicate the median)

To illustrate the status of $PM_{2.5}$ pollution in Xiamen, concentration of $PM_{2.5}$ were compared with other cities over China and foreign countries (Table 1). Compared with other cities in China, $PM_{2.5}$ concentration in Xiamen was lower than those values observed in inland cities and many coastal cities in China, such as Beijing (51 μg/m^3), Xi'an (63 μg/m^3), Chengdu (51 μg/m^3), Wuhan (49 μg/m^3), Dalian (30 μg/m^3), Qingdao (34 μg/m^3), Shanghai (36 μg/m^3), Ningbo (33 μg/m^3), Guangzhou (35 μg/m^3), but much higher than Haikou (18 μg/m^3). Due to similar geographical and climatic conditions, the concentration of $PM_{2.5}$ in Xiamen was closest to Shenzhen (26 μg/m^3). The annual $PM_{2.5}$ concentration in Xiamen was similar with Italy (Veneto) and Greece (Thessaloniki).

Study area	Sampling period	$PM_{2.5}$ (μg/m^3)	Reference
Xiamen, China	2018	22	This study

Xiamen, China	2018	25	(Xiamen Municipal Ecological Environment Bureau, 2019)
Beijing, China	2018	51	(Beijing Municipal Ecology and Environment Bureau, 2019)
Xi'an, China	2018	63	(Xi'an Ecology and Environment Bureau, 2019)
Chengdu, China	2018	51	(Chengdu Municipal Bureau of Ecology and Environment, 2019)
Wuhan, China	2018	49	(Wuhan Municipal Bureau of Ecology and Environment, 2019)
Dalian, China	2018	30	(Dalian Municipal Bureau of Ecology and Environment, 2019)
Qingdao, China	2018	34	(Qingdao Municipal Bureau of Ecology and Environment, 2019)
Shanghai, China	2018	36	(Shanghai Municipal Bureau of Ecology and Environment, 2019)
Ninbo, China	2018	33	(Ningbo Municipal Bureau of Ecology and Environment, 2019)
Guangzhou, China	2018	35	(Guangzhou Municipal Ecological Environment Bureau, 2019)
Shenzhen, China	2018	26	(Shenzhen Municipal Bureau of Ecology and Environment, 2019)
Haikou, China	2018	18	(Haikou Municipal Bureau of Ecology and Environment, 2019)
Delhi, India	2014	190	(Saxena et al., 2017)
Budapest, Hungary	2010.6~2013.5	21.0	(Szigeti et al., 2015)
Veneto, Italy	2012.4~2013.3	25.0	(Masiol et al., 2015)
Thessaloniki, Greece	2011.6~2012.5	27.7	(Tolis et al., 2015)

Table 1. Comparisons of $PM_{2.5}$ concentrations in other cities

3.2 Water-Soluble Inorganic Ions

As showed in Table 2, the annual mean concentrations of total analysed water-soluble inorganic ions in $PM_{2.5}$ in Xiamen was 9.94 μg/m^3, accounting for 45.2% of the $PM_{2.5}$ mass concentration. The concentrations of each water-soluble inorganic ions were observed in the order of SO_4^{2-}>NH_4^+>NO_3^->Na^+>K^+>F^->Cl^->Ca^{2+}>Mg^{2+}. Sulfate, ammonium and nitrate, as typical secondary inorganic aerosols (SIAs), were the dominant water-soluble inorganic species, with the combined proportions accounting for 77.4% of the total analysed water-soluble inorganic ions concentrations. The SIAs usually derived from the secondary transformation of direct emission pollutant, such as sulfur dioxide (SO_2), nitrogen oxides (NO_x), and ammonia (NH_3).

The average water-soluble inorganic ions concentrations were higher during winter and spring and lower during summer and autumn (Table 2), it is inconsistent with the seasonal variation characteristics in Nanjing (Zhang et al., 2019). The contributions of water-soluble inorganic ions in $PM_{2.5}$ ranked in an order of summer (52.0%) > spring (49.0%) > autumn (42.1%) = winter (42.1%). The proportions of the concentrations of SIAs to the total analysed water-soluble inorganic ions mass concentrations were ranked in the order of winter (81.6%) > autumn (77.5%) > spring (75.6%) > summer (72.4%). The lowest proportion of the concentrations of SIAs to the total analyzed water-soluble inorganic ions and highest contribution of water-soluble inorganic ions in $PM_{2.5}$ in summer might because of the influence of the summer monsoon, which mainly come from the ocean, leading to a greater contribution of sea salt to water-soluble ions. The highest proportion of the concentrations of SIAs to the total analyzed water-soluble inorganic ions might cause by the surrounding and long-distance transmission of air pollutants. The reason for the lowest contributions of water-soluble inorganic ions in $PM_{2.5}$ during winter and autumn is that there is less precipitation and dust has a greater impact on $PM_{2.5}$. As a coastal city, Cl^- and Na^+ contributed 13.9% of water-soluble inorganic ion concentration.

Species	Annual	Winter	Spring	Summer	Autumn
SO_4^{2-}	4.13	4.72	4.92	3.26	3.82
NO_3^-	1.14	1.36	1.73	0.94	0.69
Cl^-	0.38	0.54	0.53	0.23	0.31
F^-	0.39	0.22	0.41	0.76	0.21
NH_4^+	2.42	3.47	3.04	1.27	1.80
Na^+	0.70	0.69	0.72	0.64	0.77
K^+	0.54	0.43	1.10	0.34	0.34
Mg^{2+}	0.05	0.06	0.10	0.03	0.04
Ca^{2+}	0.18	0.20	0.29	0.09	0.16
SUM_{SIA}	7.69	9.55	9.69	5.47	6.32
SUM_{ions}	9.94	11.70	12.83	7.55	8.15
SUM_{SIA}/SUM_{ions} (%)	77.4	81.6	75.6	72.4	77.5
$SUM_{ions}/PM_{2.5}$ (%)	45.2	42.1	49.0	52.0	42.1

SUM_{SIA}: sum of SO_4^{2-}, NO_3^- and NH_4^+; SUMions: sum of all analysed water-soluble inorganic ions.

Table 2. Annual and seasonal mean concentrations of water-soluble inorganic ions in $PM_{2.5}$ in Xiamen (μg/m^3)

The monthly and seasonal trends of water-soluble inorganic ions were obviously different (figure 3). Influenced by atmospheric oxidizing, monthly variations of SO_4^{2-} to the total

analyzed water-soluble inorganic ions exhibited the maximum in September and minimum in March. Sulfate mainly forms from the photochemical oxidation of sulfur-containing precursors, such as H_2S and SO_2 (Kleeman et al., 2000). Ozone concentration in Xiamen was relatively high in September, high concentration of ozone could aggravate photochemical activity, which in turn increase sulfate formation. The highest proportion of NH_4^+ to the total analyzed water-soluble inorganic ions was observed in January, resulting from biomass burning, microbe metabolism and agricultural activity (Yin et al., 2014).

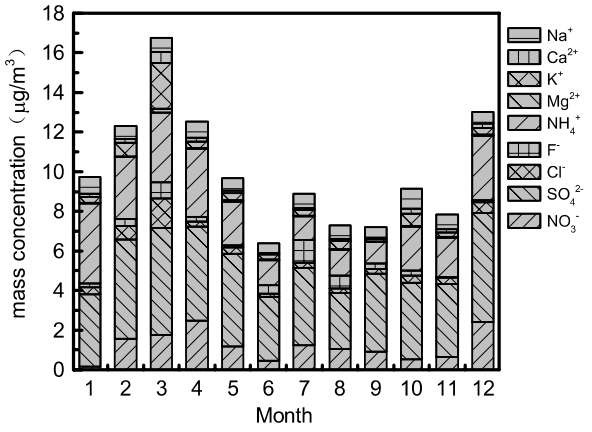

Figure 3. Monthly variations in water-soluble inorganic ions

Table 3 showed comparison between our SIAs results and previous studies. Except for Wuhan (58.0%), SIAs accounted for 31.9% ~ 47.9% of $PM_{2.5}$ concentration in Chinese cities. $PM_{2.5}$ mass concentrations in Veneto (Italy) and Thessaloniki (Greece) were similar to that in Xiamen, and SIAs accounted for about 35% of $PM_{2.5}$ mass concentration in these three cities. The combined percentages of SIAs in $PM_{2.5}$ were relatively lower in Delhi (India) and Budapest (Hungary). The differences in contribution of SIAs to $PM_{2.5}$ among cities might because of variations in energy structure, economic structure, geological condition, meteorological characteristics and population density.

Study area	Sampling period	$PM_{2.5}$ (μg/m³)	SUM_{SIA} (μg/m³)	$SUM_{SIA}/PM_{2.5}$ (%)	Reference
Xiamen, China	2018	22	7.69	35.0	this study
Zibo, China	2006.3 ~2007.2	164.4	59.17	36.0	(Luo et al., 2018)
Handan, China	2013 ~2014	139.4	58.8	42.2	(Meng et al., 2016)
Baoji, China	2012.3 ~2013.3	132	42.1	31.9	(Niu et al., 2016)
Shanghai, China	2013.9 ~2014.8	94.6	40.63	42.9	(Ming et al., 2017)
Nanjing, China	2014.12 ~2015.11	94.4	33.45	35.4	(Zhang et al., 2019)
Hefei, China	2012.9 ~2013.8	86.29	38.52	44.6	(Deng et al., 2016)
Wuhan, China	2013.1 ~2013.12	65	37.73	58.0	(Huang et al., 2016)
Zhuhai, China	2015.1 ~2016.1	36.4	17.42	47.9	(Liang et al., 2018)
Delhi, India	2014	190	45.53	24.0	(Saxena et al., 2017)
Veneto, Italy	2012.4 ~2013.3	25	8.1	32.4	(Masiol et al., 2015)
Budapest, Hungary	2010.6 ~2013.5	21	6.2	29.5	(Szigeti et al., 2015)
Thessaloniki, Greece	2011.6 ~2012.5	27.7	10.2	36.8	(Tolis et al., 2015)

SUM_{SIA}: sum of SO_4^{2-}, NO_3^- and NH_4^+.

Table 3. Comparisons of $PM_{2.5}$, SUM_{SIA} concentrations in other cities

3.3 Source of Particulate Matter

Based on the single particle mass spectrometry detection results and the characteristic source spectra of local air pollution sources, the source analysis of local particles can be carried out. Mobile sources emission was the most important sources of particle matter which contributed over 30% (Figure 4). Coal combustion was the second largest source, accounting for about 13%. Biomass combustion accounted for about 7%, and the percentages of dust and industrial process were around 5%. The proportion of sea salt was the least, but the seasonal variation of sea salt proportion is the most significant, with proportion changing from 1.6% in winter to 6.6% in summer.

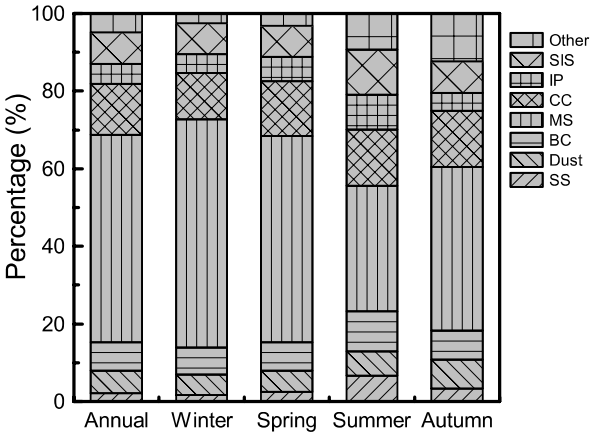

Figure 4. Seasonal variations in the source of particulate matter (SS: sea salt, BC: biomass combustion, MS: mobile source, CC: coal combustion, IP: industrial process, SIS: secondary inorganic source)

4. CONCLUSIONS

The chemical composition of aerosols in 2018 was discussed. The results showed that the annual average mass concentrations of $PM_{2.5}$ was 22 μg/m³. This value was lower than those values observed in inland cities and many coastal cities in China, suggesting that the fine particle pollution was relatively light in Xiamen. The concentration of water-soluble inorganic ions was 9.94 μg/m³ which accounted for 45.2% of $PM_{2.5}$. The average water-soluble inorganic ions concentrations were higher during winter and spring and lower during summer and autumn. SO_4^{2-}, NO_3^- and NH_4^+ were main components of secondary reactions which contributed more than 77 percent of water-soluble inorganic ion concentration. As a coastal city, Cl^- and Na^+ contributed 13.9 percent of water-soluble inorganic ion concentration. Based on single particle aerosol mass spectrometer analysing, mobile sources emission was the most important sources of particle matter which contributed over 30%. The proportion of sea salt was the least, but the seasonal variation of sea salt proportion is the most significant.

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