

## Study of Water-Soluble Ions in Ambient Air Suspended Particles in Taichung, Taiwan

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This experiment was conducted during November in 2000 and February in 2001 in Taichung, central Taiwan. The study used a TE-PUF sampler, Universal air sampler and MOUDI (Micro-Orifice Uniform Deposit Impactor) sampler to collect the ambient air suspended particles and analyzed their water-soluble ions ( $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$ ). Analytical results indicate that the mean mass concentration for  $\text{PM}_{2.5}$ ,  $\text{PM}_{2.5-10}$  and TSP measured at the CCRT (traffic) sampling site are 54.5, 27.8 and 158  $\mu\text{g m}^{-3}$ , respectively. Additionally, the mean mass concentration for  $\text{PM}_{2.5}$ ,  $\text{PM}_{2.5-10}$  and TSP measured at the THUC sampling site are 23.6, 16.3 and 85.6  $\mu\text{g m}^{-3}$ , respectively. Furthermore, the major water-soluble ions of  $\text{PM}_{2.5}$  measured in this sampling region are  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ , while those of  $\text{PM}_{2.5-10}$  are  $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ , respectively. At the CCRT (traffic) sampling site, the mean size distributions of mass,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  exhibit a bimodal distribution whereas those of  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  have a bimodal distribution. At the THUC (campus) sampling site, the mean size distributions of mass,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  are bimodal whereas those of  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$  and  $\text{Mg}^{2+}$  have display a unimodal distribution.

**Keywords:** Suspended particles; Anion; Cation; Size distribution; relationship

### 1. Introduction

Atmospheric particle size distribution and the composition and relationship among chemical species for continental, marine background and urban influenced aerosols have been reported for various sites around the world (Zhuang *et al.*, 1999; Zheng and Fang, 2000; Parmar *et al.*,

2001). Some of these works have shown the major components of the  $\text{PM}_{10}$  mass in most urban areas and have found that the main components include 1) geological material (oxides of aluminum, silicon, calcium, titanium, and iron); 2) OC (comprising of hundreds of compounds); 3) EC; 4)  $\text{SO}_4^{2-}$ ; 5)  $\text{NO}_3^-$ ; 6)  $\text{NH}_4^+$ . Furthermore, liquid water absorbed by soluble species was also a major component in the sum when the relative humidity exceeded  $\sim 70\%$ . However, much of the absorbed liquid evaporates if the filters are equilibrated prior to weighing. Chen *et al.* (1999) found that the  $\text{PM}_{2.5}$  fraction

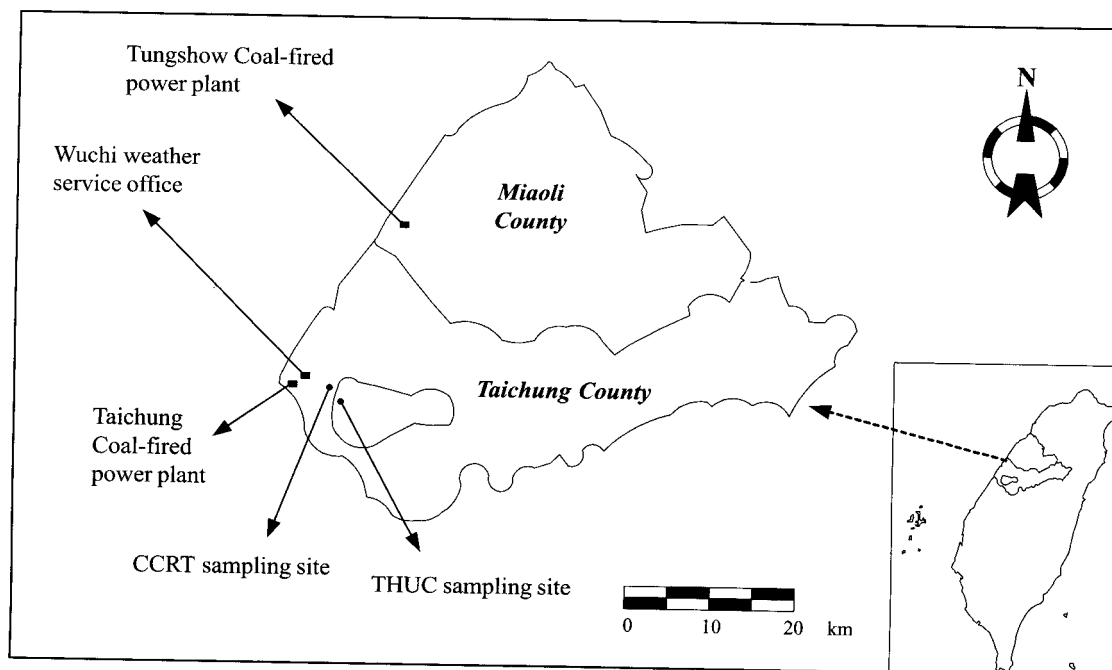
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**Fig.1.** The sampling region in this study.

comprised 61-67% of the  $PM_{10}$  mass in central and southern Taiwan, but just 54-59% in northern Taiwan. Meanwhile Parmar *et al.* (2001) confirmed that the mass and size distributions of total aerosol and the ions were bimodal for  $NH_4^+$ ,  $Cl^-$ ,  $NO_3^-$ ,  $K^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $SO_4^{2-}$  and  $Na^+$  and unimodal for  $F^-$ . Additionally,  $SO_4^{2-}$ ,  $F^-$ ,  $K^+$  and  $NH_4^+$  were dominated in the fine particulate mode, while  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Cl^-$  and  $NO_3^-$  dominated in the coarse particulate mode. Finally,  $Na^+$  was present in both the coarse and the fine modes. Regarding the relationship between the chemical species, Al-momani *et al.* (1995) showed that  $Na^+$  and  $Cl^-$  ions were strongly correlated with each other owing to their common marine source. The  $Na^+$  ion is also slightly correlated with  $K^+$ ,  $Ca^{2+}$  and  $Mg^{2+}$ , indicating that sea salt accounts for a certain fractions of these elements. Most of the crustal elements, namely  $K^+$ ,  $Ca^{2+}$  and  $Mg^{2+}$ , are also correlated with each other. Meanwhile nss- $SO_4^{2-}$  (non-sea-salt  $SO_4^{2-}$ ),  $NO_3^-$  and  $NH_4^+$  are strongly correlated with each other. The  $Ca^{2+}$  ion is also correlated with the above ions, but more weakly. Analytical results from Chongju,

South Korea found that high correlations among  $SO_4^{2-}$ ,  $NO_3^-$  and  $NH_4^+$  were observed in  $PM_{2.5}$  (Lee and Kang, 2001).

Atmospheric pollutant in the form of suspended particle has been an important issue for the EPA in Taiwan during the past few years. Water-soluble ions such as  $SO_4^{2-}$ ,  $NO_3^-$  and  $NH_4^+$  are the major contributors to air pollution. This study investigates the water-soluble ion composition and size distribution of atmospheric suspended particles in Taichung, central Taiwan.

## 2. Experimental methods

### 2.1 Sampling Site

As displayed in Fig. 1, this work selected two sampling sites located in the west of Taichung City. One of those sites was the THUC sampling site, set in the campus of the Tung-Hai University (THU), and located on the west edge of Taichung City. This site was in the mid-slope area of the east side of Da-Du Mountain (300 m above

**Table 1** Design of the sampling periods in this study.

Period	Time	Device
Period-I	2000.11.01-2001.01.04	TE-PUF Sampler Universal Air Sampler MOUDI Sampler
Period-II	2001.01.14-2001.02.06	TE-PUF Sampler Universal Air Sampler MOUDI Sampler

sea level). The number of students at THU is around 15,000. The other sampling site was the CCRT sampling site, set on the traffic island in front of Hung-Kuang Institute of Technology (HKIT) on Chung-Chi Road in Sha-Lu. This site was located in the mid-slope region on the west side of Da-Du Mountain ( about 300 m from sea level). Meanwhile HKIT has about 10,000 students. Additionally, the traffic density on Chung-Chi Road was around 2,500-3,000 vehicles per hour during the sampling period. The number showed that the CCRT site was the typical traffic sampling site. Further notable features of the region include the Taichung and Tungsho coal fire power plants, located to the west and north of the general sampling area. Meanwhile, the Wuchi weather service office is also located in the sampling region. The relative humidity and temperature measurements taken by the station were simultaneously duplicated at the two sampling site.

## 2.2 Sampling Program

Sampling Period I comprised ambient particle measurements taken between 2 November 2000 and 6 February 2001. Table 1 lists the design of the sampling period in this work. Period-I was from November 2 in 2000 to January 4 in 2001. The TE-PUF Sampler, Universal Air Sampler and MOUDI Sampler collected the ambient particulate matter daily for each 24 hours period at CCRT sampling site. Simultaneously the same sampler were gathered at the THUC

sampling site. Meanwhile, Period-II was ran from January 14 to February 6 in 2001. During Period-II the TE-PUF Sampler, Universal Air Sampler and MOUDI Sampler were once again used to take consecutive 24 hour samples at THUC sampling site throughout the sampling period . Simultaneously the same sampler were gathered at CCRT sampling site. Each sampling device was located 0.5 m above ground level (except for the MOUDI which was located 1 m above ground level).

## 2.3 Ambient Suspended Particle Sampler

The TE-PUF Sampler (TE-PUF Poly-Urethane Foam High Volume Air Sampler) is a stand alone system designed to simultaneously gather suspended airborne particles at the flow rates of up to 280 liters per minute. This work adjusted the flow rate of 200 liters per minute. The TE-PUF employs the latest technological advances for accurately measuring airborne particulates and vapors. (TE-PUF Poly-Urethane Foam High Volume Air Sampler Operations manual, 1998).

The Model 310 Universal Air Sampler<sup>TM</sup> (UAS<sup>TM</sup>) is a general-purpose air sampler for atmospheric aerosol sampling and for mass concentration, and for organic or inorganic analysis. The sampler has a design inlet sampling flow rate of 300 lpm. Furthermore the sampler includes two virtual impactors for size fractionation of airborne particles. Finally, the sampler can operate as a high volume dichotomous sampler for size fractionation of airborne particles in the aerodynamic size ranges of 0-2.5  $\mu\text{m}$  and 2.5-10  $\mu\text{m}$ , respectively (Model 310 Universal Air Sampler<sup>TM</sup> Instruction Manual, 1996).

The MOUDI (Micro-Orifice Uniform Deposit Impactor, Model 100, MSP) Sampler is a cascade

**Table 2** The chemical species for PM<sub>2.5</sub>, PM<sub>2.5-10</sub> and TSP measured in this study. (Unit:  $\mu\text{g m}^{-3}$ )

Species	CCRT			THUC				
	PM <sub>2.5</sub> (N=12)	PM <sub>2.5-10</sub> (N=12)	TSP (N=23)	PM <sub>2.5</sub> / PM <sub>2.5-10</sub>	PM <sub>2.5</sub> (N=11)	PM <sub>2.5-10</sub> (N=11)	TSP (N=23)	PM <sub>2.5</sub> / PM <sub>2.5-10</sub>
Mass	54.5	27.8	158	1.96	23.6	16.3	85.5	1.45
Cl <sup>-</sup>	1.71	2.27	4.25	0.76	0.62	1.38	1.68	0.45
NO <sub>3</sub> <sup>-</sup>	2.01	2.19	5.23	0.92	1.18	1.72	2.81	0.69
SO <sub>4</sub> <sup>2-</sup>	9.47	2.36	12.8	4.02	5.26	1.26	6.82	4.19
Na <sup>+</sup>	1.52	1.79	3.26	0.85	0.33	0.87	1.15	0.38
NH <sub>4</sub> <sup>+</sup>	4.60	1.11	6.67	4.16	3.51	0.86	4.90	4.07
K <sup>+</sup>	0.59	0.23	1.24	2.59	0.58	0.13	0.77	4.36
Mg <sup>2+</sup>	0.32	0.62	0.74	0.52	0.32	0.61	0.58	0.53
Ca <sup>2+</sup>	0.63	1.49	2.08	0.42	0.51	0.73	1.19	0.70

impactor intended for general-purpose aerosol sampling. The MOUDI Sampler is an 8-stage cascade impactor and comprises five basic assemblies, rotator impactor, magnetohelic gage, rotator unit, shelter and blower motor. The flow rate is 30 liter per minute and the cut size diameter ranges from 18  $\mu\text{m}$  in the first stage to 0.056  $\mu\text{m}$  in the final stage (Model 100 Micro-Orifice Uniform Deposit Impactor, MOUDI<sup>TM</sup>, Instruction Manual, 1989).

#### 2.4 Analysis of Water Soluble Ions

The quartz filters in the 102 mm samples (one eighths for all samples), 200 mm  $\times$  250 mm samples (one eighths for all samples) and 62 mm  $\times$  165 mm sample (one quarter for all samples) were analyzed for anion species (Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>) and cation species (Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup> and Ca<sup>2+</sup>) by Ion Chromatography (Dionex DX-100). All of the filters for each sample were individually placed in 100 ml PE bottles and 50 ml distilled-deionized water (DDW) was then added to each bottle. The bottles were sent for ultrasonic processing for 90 minutes. All the extraction solutions were filtered with a glass microfiber filter (GF/C, Whatman) with a 0.45

$\mu\text{m}$  nominal pore size diameter and stored under the refrigerator at 4 °C in PE bottles to wait for chemical analysis. The anion species were analyzed using the Ion Chromatography (Dionex DX-100) equipped with an AS4A analytical column and an AG4A guard column. Samples were injected into the sampling loop (20  $\mu\text{l}$ ) via a sterile. The eluent for these samples was 1.8 mM Na<sub>2</sub>CO<sub>3</sub>/1.7 mM NaHCO<sub>3</sub> and they had a flow rate of 2 ml min<sup>-1</sup>. The cation species were also analyzed with Ion Chromatography (Dionex DX-100) but equipped with the addition of a CS12A cation column and a CG12A guard column. Samples were injected into the sampling loop (20  $\mu\text{l}$ ) via a sterile. The eluent for these samples was 20 mM Methanesulfonic acid (MSA) solution at a flow rate of 1 ml min<sup>-1</sup>. The concentrations of the anion species (Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>) and cation species (Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup> and Ca<sup>2+</sup>) were measured by constructing calibration curves with freshly prepared solutions that met reliable commercial standards. At least 10% of the samples were analyzed by spiking with a standard anion solution (Merck INC) followed by recovery testing. The recover varied from 85% to 115%.

**Table 3** The correlation coefficient calculated at CCRT sampling site.

PM <sub>2.5</sub>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Temp.	RH
Cl <sup>-</sup>	1.00									
NO <sub>3</sub> <sup>-</sup>	0.23	1.00								
SO <sub>4</sub> <sup>2-</sup>	0.10	0.71**	1.00							
Na <sup>+</sup>	0.81**	0.22	0.13	1.00						
NH <sub>4</sub> <sup>+</sup>	0.14	0.82**	0.97**	0.18	1.00					
K <sup>+</sup>	-0.19	0.54	0.67*	-0.22	0.60*	1.00				
Mg <sup>2+</sup>	0.06	-0.66*	-0.37	0.06	-0.47	-0.10	1.00			
Ca <sup>2+</sup>	0.07	-0.79**	-0.47	0.00	-0.56	-0.50	0.56	1.00		
Temp.	0.53	0.12	0.17	0.38	0.19	-0.43	-0.36	0.00	1.00	
RH	0.29	0.22	0.52	0.36	0.43	0.12	-0.13	-0.13	0.32	1.00

PM <sub>2.5-10</sub>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Temp.	RH
Cl <sup>-</sup>	1.00									
NO <sub>3</sub> <sup>-</sup>	0.16	1.00								
SO <sub>4</sub> <sup>2-</sup>	0.51	0.57	1.00							
Na <sup>+</sup>	0.92**	0.09	0.50	1.00						
NH <sub>4</sub> <sup>+</sup>	0.46	0.52	0.81**	0.35	1.00					
K <sup>+</sup>	-0.11	0.38	0.27	-0.21	0.54	1.00				
Mg <sup>2+</sup>	0.37	0.11	0.15	0.41	-0.01	-0.38	1.00			
Ca <sup>2+</sup>	0.42	0.13	0.11	0.41	0.02	-0.13	0.86**	1.00		
Temp.	0.35	-0.34	-0.06	0.54	-0.27	-0.54	0.33	0.36	1.00	
RH	0.53	-0.04	0.38	0.43	0.24	-0.10	0.16	0.35	0.32	1.00

\*Significant level:  $p < 0.05$ \*\*Significant level:  $p < 0.01$ 

### 3. Results and Discussion

#### 3.1 Concentration of Chemical Species in the Suspended Particles

Table 2 lists the concentrations of chemical species measured in this work. The particulate mass concentration, namely mean mass concentration for PM<sub>2.5</sub>, PM<sub>2.5-10</sub> and TSP measured at the CCRT sampling site was 54.5, 27.8 and 158  $\mu\text{g m}^{-3}$ , respectively. Furthermore, the mean mass concentration for PM<sub>2.5</sub>, PM<sub>2.5-10</sub> and TSP measured at the THUC sampling site were 23.6, 16.3 and 85.6  $\mu\text{g m}^{-3}$ , respectively. The major components of PM<sub>2.5</sub> gathered at the CCRT sampling site were found to be SO<sub>4</sub><sup>2-</sup> (9.47  $\mu\text{g m}^{-3}$ ), NH<sub>4</sub><sup>+</sup> (4.60  $\mu\text{g m}^{-3}$ ) and NO<sub>3</sub><sup>-</sup> (2.01  $\mu\text{g m}^{-3}$ ). These three species are also considered to be the major secondary aerosols by anthropogenic sources. The major components of PM<sub>2.5-10</sub> at this traffic sampling site are SO<sub>4</sub><sup>2-</sup> (2.36  $\mu\text{g m}^{-3}$ ), Cl<sup>-</sup> (2.27  $\mu\text{g m}^{-3}$ ) and NO<sub>3</sub><sup>-</sup> (2.19  $\mu\text{g m}^{-3}$ ). Cl<sup>-</sup> in the coarse particles is generally

derived from the sea spray (Turnbull and Harrison, 2000). Meanwhile, the major components of TSP are SO<sub>4</sub><sup>2-</sup> (12.5  $\mu\text{g m}^{-3}$ ), NH<sub>4</sub><sup>+</sup> (5.89  $\mu\text{g m}^{-3}$ ) and NO<sub>3</sub><sup>-</sup> (5.24  $\mu\text{g m}^{-3}$ ).

Table 2 lists the concentrations of chemical aerosols measured at the THUC sampling site. The major component for PM<sub>2.5</sub> are SO<sub>4</sub><sup>2-</sup> (5.26  $\mu\text{g m}^{-3}$ ), NH<sub>4</sub><sup>+</sup> (3.51  $\mu\text{g m}^{-3}$ ) and NO<sub>3</sub><sup>-</sup> (1.18  $\mu\text{g m}^{-3}$ ). The concentrations of these three anion species measured at the THUC sampling site are all lower than those measured at the CCRT sampling site. As for the chemical species of PM<sub>2.5-10</sub>, the major components are NO<sub>3</sub><sup>-</sup> (1.719  $\mu\text{g m}^{-3}$ ), Cl<sup>-</sup> (1.379  $\mu\text{g m}^{-3}$ ) and SO<sub>4</sub><sup>2-</sup> (1.256  $\mu\text{g m}^{-3}$ ). Meanwhile, the other components are all below 1  $\mu\text{g m}^{-3}$ . The major components of TSP are SO<sub>4</sub><sup>2-</sup> (7.380  $\mu\text{g m}^{-3}$ ), NH<sub>4</sub><sup>+</sup> (5.619  $\mu\text{g m}^{-3}$ ) and NO<sub>3</sub><sup>-</sup> (3.284  $\mu\text{g m}^{-3}$ ). As noted above, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> are also the major components of TSP at the THUC sampling site during the same sampling period. In total, the values of PM<sub>2.5</sub>/PM<sub>2.5-10</sub> for mass at these two sampling sites both exceed 1. Consequently, fine particles

**Table 4** The correlation coefficient calculated at THUC sampling site.

PM <sub>2.5</sub>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Temp.	RH
	Cl <sup>-</sup>	1.00								
NO <sub>3</sub> <sup>-</sup>	-0.52	1.00								
SO <sub>4</sub> <sup>2-</sup>	-0.20	0.64*	1.00							
Na <sup>+</sup>	0.41	0.09	-0.06	1.00						
NH <sub>4</sub> <sup>+</sup>	0.06	0.46	0.87**	0.25	1.00					
K <sup>+</sup>	0.32	0.18	0.56	0.39	0.79**	1.00				
Mg <sup>2+</sup>	0.45	0.04	0.22	0.01	0.27	0.29	1.00			
Ca <sup>2+</sup>	0.06	0.26	0.32	0.13	0.59	0.62*	0.40	1.00		
Temp.	-0.12	-0.12	-0.26	-0.38	-0.33	-0.59	0.03	-0.14	1.00	
RH	0.09	-0.25	-0.55	-0.22	-0.64*	-0.65*	-0.07	-0.38	0.76**	1.00

PM <sub>2.5-10</sub>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Temp.	RH
	Cl <sup>-</sup>	1.00								
NO <sub>3</sub> <sup>-</sup>	0.48	1.00								
SO <sub>4</sub> <sup>2-</sup>	0.09	0.42	1.00							
Na <sup>+</sup>	0.85**	0.62*	0.37	1.00						
NH <sub>4</sub> <sup>+</sup>	0.06	0.60	0.75**	0.34	1.00					
K <sup>+</sup>	-0.15	-0.24	0.19	0.11	0.06	1.00				
Mg <sup>2+</sup>	-0.63*	-0.54	-0.07	-0.74**	0.01	0.05	1.00			
Ca <sup>2+</sup>	-0.41	0.01	0.17	-0.35	0.53	0.01	0.72*	1.00		
Temp.	0.47	-0.04	-0.63*	0.21	-0.68*	-0.01	-0.32	-0.56	1.00	
RH	0.22	-0.01	-0.58	-0.04	-0.74**	-0.19	-0.44	-0.71*	0.76**	1.00

\*Significant level:  $p < 0.05$ \*\*Significant level:  $p < 0.01$ 

are the major component of PM<sub>10</sub> at both of the sampling sites. The following section discussed the size distribution of each chemical species. SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> represent the dominant chemical components in the ambient suspended particulate matter for the sampling region. Harrison and Jones (1995) noted that secondary SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> particles reside in the atmosphere for a long time due to their physical size. These particles are known to be produced largely by gas-phase atmospheric reactions of primary combustion related SO<sub>2</sub> and NO<sub>x</sub> (QUARG, 1996). During the whole sampling period, the prevailing wind direction in the sampling region was N and NE. Additionally, the Taiwan Strait, which located in the west and north of this sampling region, are the possible emission source of SO<sub>2</sub>. The PM<sub>2.5</sub>/PM<sub>2.5-10</sub> levels measured at the CCRT sampling site (4.02) are slightly lower than at the THUC sampling site (4.19). Diesel exhaust is another possible emission source of the SO<sub>2</sub> measured at the CCRT (traffic) sampling site.

## 3.2 Relationship Between Chemical Species and Meteorological Factors

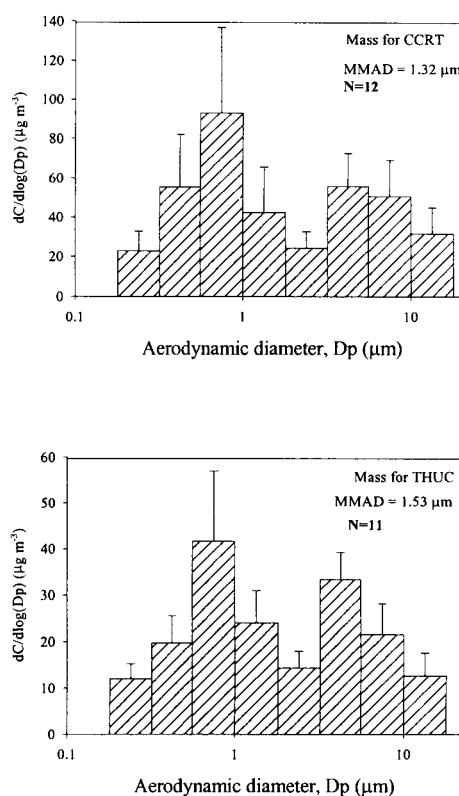
### 3.2.1 Pearson Correlation Coefficient Matrix for the CCRT Sampling Site

The relationship among the chemical species was estimated based on the Pearson Correlation Coefficient Matrix (PCCM). Table 3 lists the results for the CCRT sampling site. In the fine particles, a high correlation was observed among SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> suggesting that these species are associated in ambient air. (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub> may be the possible form of these particles. This relationship is similar to that obtained by Tsai and Cheng (1999) who investigated a coastal area in Central Taiwan located 8 km from the two sampling sites examined here. Tsai and Cheng found the SO<sub>4</sub><sup>2-</sup> concentrations displayed moderate correlation with NH<sub>4</sub><sup>+</sup> ( $r = 0.78$ ) and NO<sub>3</sub><sup>-</sup> ( $r = 0.63$ ) in PM<sub>10</sub>. Meanwhile, Kim *et al.* (1998) obtained a similar result at Kosan on Cheju Island, Korea (another

sampling region located close to the sea), and showed a correlation between  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  ( $r = 0.79$ ) and between  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  ( $r = 0.50$ ) for the fine particles. Furthermore, a significant correlation between  $\text{NH}_4^+$  and  $\text{K}^+$  was found at Kosan ( $r = 0.66$ ), similar to that found in this study ( $r = 0.60$ ). As for the coarse particles, a strong correlation coefficient between  $\text{Na}^+$  and  $\text{Cl}^-$  is found in the Table 3 ( $r = 0.92$ ), meaning that these two chemical species share the same sources at the CCRT sampling site. Tasi and Cheng (1999) found a significant correlation between  $\text{Na}^+$  and  $\text{Cl}^-$  in  $\text{PM}_{10}$  ( $r = 0.61$ ), but the correlation was lower than that found in this study. The source of these two species appears to be marine aerosol that is transported from the Taiwan Strait in the west side of the sampling region (<10 km). A good correlation ( $r = 0.81$ ) is also found in the coarse particle for  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ . Zhuang *et al.* (1999) noted that both  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$  would occur in the coarse mode when agglomeration was the major process.

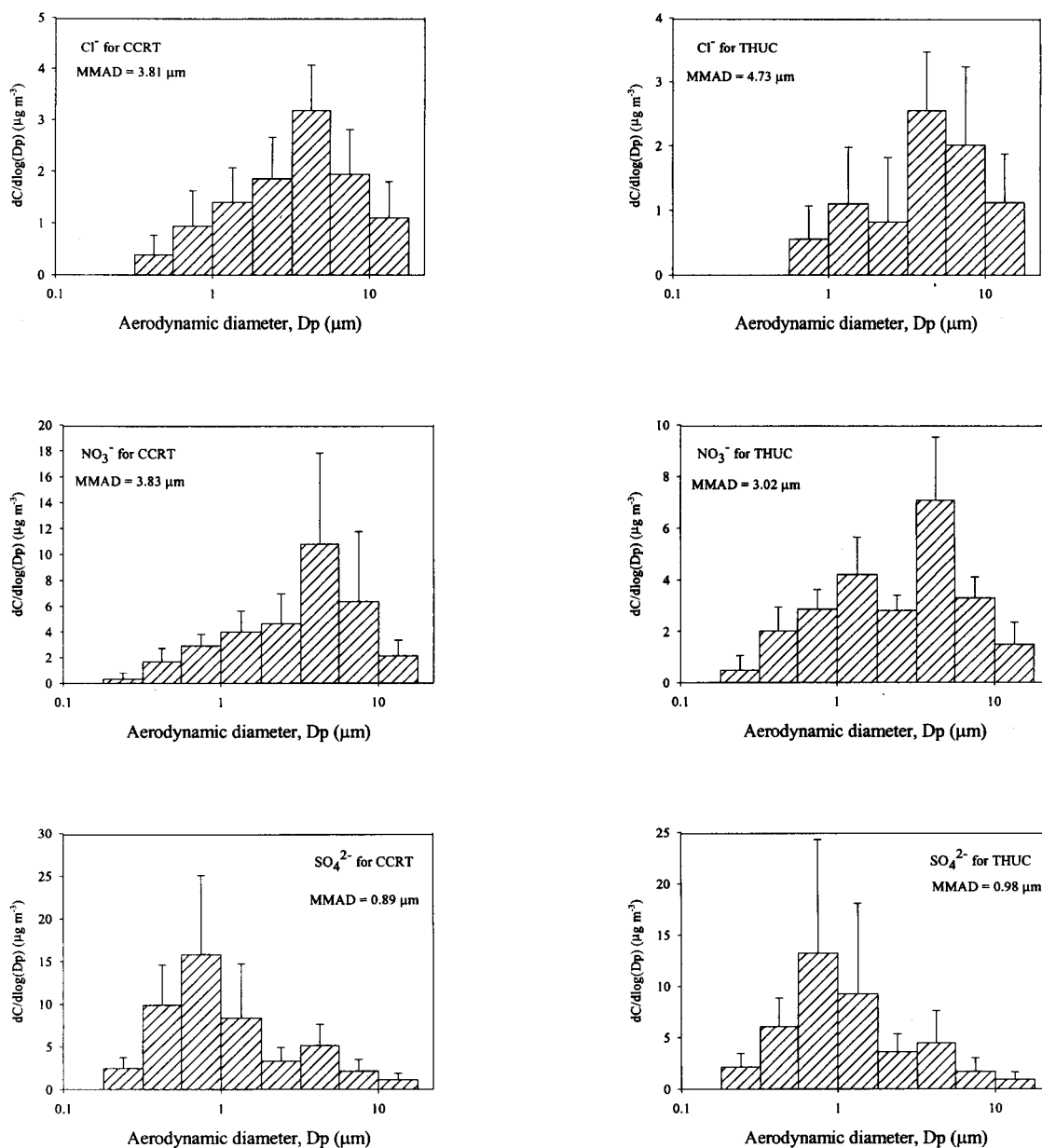
### 3.2.2 Pearson Correlation Coefficient Matrix for the THUC Sampling Site

Table 4 lists the calculation results for PCCM at the THUC sampling site.  $\text{SO}_4^{2-}$ , a typical anthropogenic air pollutant, displays a good correlation with  $\text{NH}_4^+$  ( $r = 0.87$ ) and  $\text{NO}_3^-$  ( $r = 0.64$ ). The correlation is slightly stronger than that found by Lee and Kang (2001) in their investigation at the roof of the Chongju National College of Science and Technology building in Chongju, South Korea. This sampling site is also a typical campus environment like the THUC sampling site in this study. Lee and Kang suggested good correlations between  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  ( $r = 0.77$ ) and between  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  ( $r = 0.49$ ). The particularly high correlation between  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  indicates that these two species generally coexist in fine particle (ammonium



**Fig. 2.** The mean mass size distribution of ambient suspended particles.

sulfate) in the atmosphere. Zhuang *et al.* (1999) found the same correlation in their study. A good correlation also exists between  $\text{K}^+$  and  $\text{NH}_4^+$  ( $r = 0.79$ ). Finally, these two species both have a negative correlation with relative humidity. Table 4 lists the calculation results for PCCM of coarse particles at the THUC sampling site. A good correlation exists between  $\text{Na}^+$  and  $\text{Cl}^-$  ( $r = 0.85$ ).  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  is also found to have a good correlation ( $r = 0.75$ ) for the coarse particles. This result resembles the relationship found at the CCRT sampling site. The correlations between  $\text{Mg}^{2+}$  and  $\text{Cl}^-$  and between  $\text{Mg}^{2+}$  and  $\text{Na}^+$  are both negative. We hypothesize that this phenomenon occurs because the concentration of  $\text{Mg}^{2+}$  measured at this sampling site is transformed directly from the local soil dust rather than being transformed from the sea salt.



**Fig. 3a.** The mean size distributions of anion species measured at the CCRT sampling site. (N=12)

**Fig. 3b.** The mean size distribution of anion species measured at THUC sampling site. (N=11)

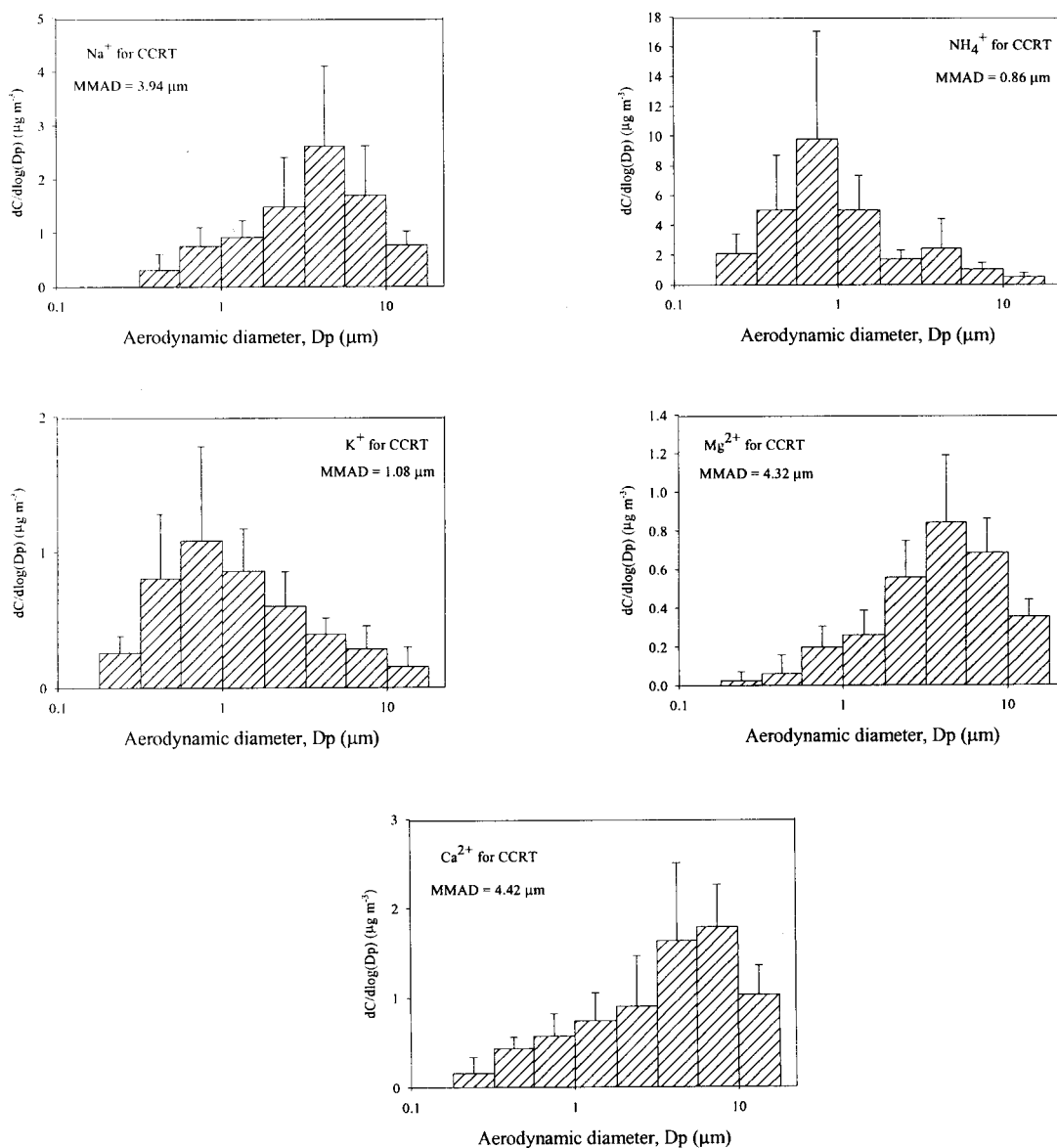
### 3.3 Size Distribution

#### 3.3.1 Mass Size Distribution

Fig. 2 displays the mean mass size distribution of indicates that the mass size distribution of ambient suspended particles exhibits two modes for both two sampling sites and the size ranges

ambient suspended particles. The figure of the particles in the two modes are both between 0.56-1  $\mu\text{m}$  and 3.2-5.6  $\mu\text{m}$ , respectively. Additionally, the particle mass is 56.9% in the fine particulate mode (0.18-1.8  $\mu\text{m}$ ) and 43.1% in the coarse particulate mode (1.8-10  $\mu\text{m}$ ) at the CCRT sampling site compared to 54.6% in the fine fraction (0.18-1.8  $\mu\text{m}$ ) and 45.4% in the coarse fraction (1.8-10  $\mu\text{m}$ ) at the THUC



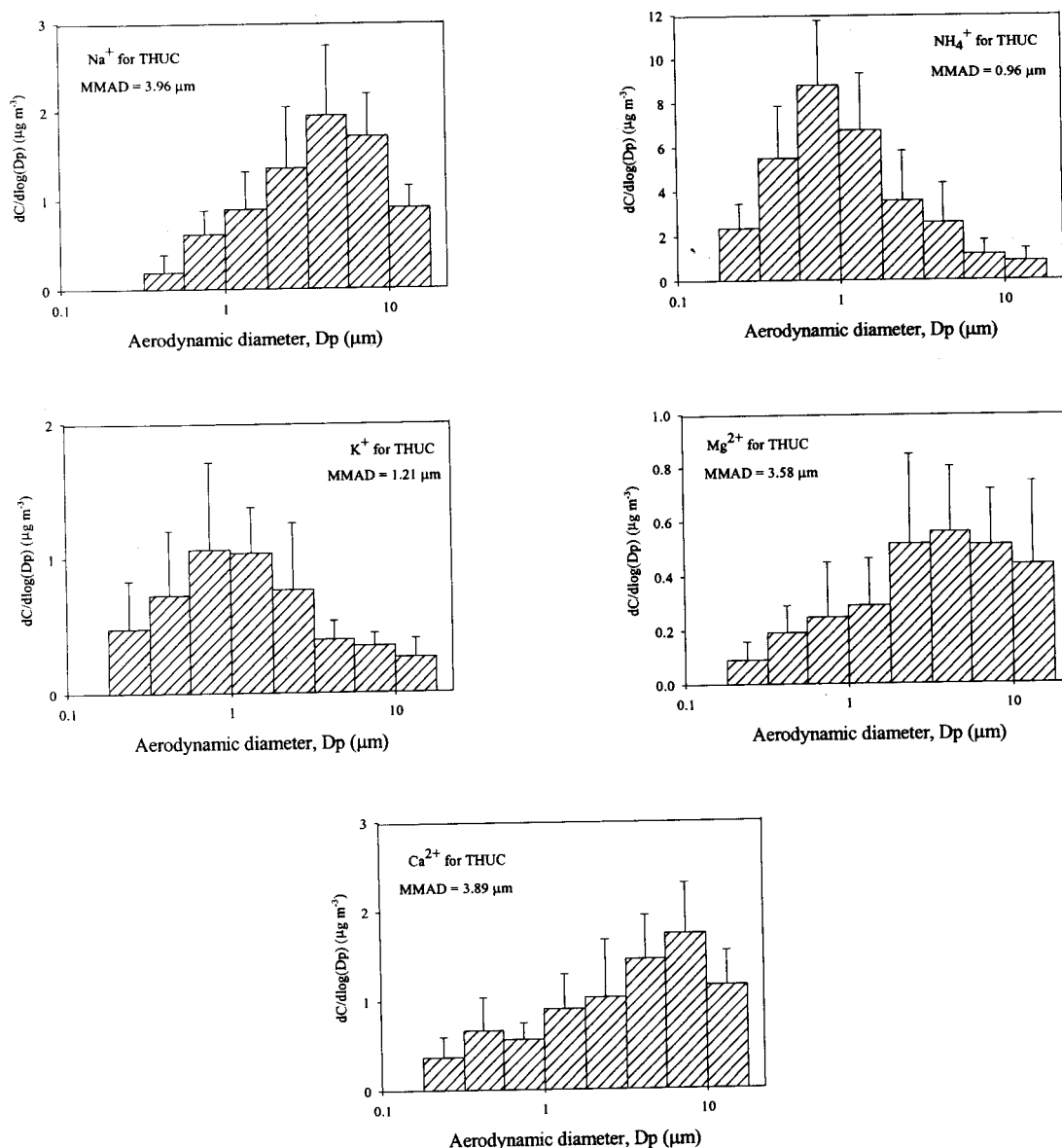


**Fig. 4a.** The mean size distribution of cation species measured at CCRT sampling site. (N=12)

sampling site. The average mass median aerodynamic diameters (MMAD) of suspended particles for the CCRT and THUC sites are 1.32  $\mu\text{m}$  and 1.53  $\mu\text{m}$ , respectively. These values are slightly lower than those found in study conducted at Agra, India by Parmar *et al.* (2001) (MMAD: 1.7  $\mu\text{m}$ ). This phenomenon indicates that the size of the suspended particulates collected in the present sampling region is smaller than that at Agra.

### 3.3.2 Anion Size Distribution

Figures 3a and 3b show the mean size distributions of anion species. Figure 3a displays that the size distributions of  $\text{Cl}^-$  and  $\text{NO}_3^-$  are unimodal and both with a peak between 3.2-5.6  $\mu\text{m}$  while the MMAD of  $\text{Cl}^-$  and  $\text{NO}_3^-$  is 3.81 and 3.83  $\mu\text{m}$ , respectively. Furthermore, the figure shows that the size distribution of  $\text{SO}_4^{2-}$  is bimodal with the major peaks between 0.56-1  $\mu\text{m}$ ; while the MMAD of  $\text{SO}_4^{2-}$  is 0.89  $\mu\text{m}$ . Figure 3b displays that the size distributions of  $\text{Cl}^-$ ,  $\text{NO}_3^-$



**Fig. 4b.** The mean size distribution of cation species measured at THUC sampling site. (N=11)

and  $\text{SO}_4^{2-}$  are bimodal and the major peak is between 3.2-5.6  $\mu\text{m}$  both for  $\text{Cl}^-$  and  $\text{NO}_3^-$ , and between 0.56-1  $\mu\text{m}$  for  $\text{SO}_4^{2-}$ , while the minor peak is between 1.0-1.8  $\mu\text{m}$  both for  $\text{Cl}^-$  and  $\text{NO}_3^-$ , and between 3.2-5.6  $\mu\text{m}$  for  $\text{SO}_4^{2-}$ . The unimodal mean size distributions of  $\text{Cl}^-$  and  $\text{NO}_3^-$  are inconsistent with the mean size distributions of anion species measured at CCRT. Additionally, the MMAD of  $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  is 4.73, 3.02 and 0.98  $\mu\text{m}$ , respectively, showing that the MMAD of  $\text{Cl}^-$  and  $\text{NO}_3^-$  differ slightly between

the two sampling sites but are similar to each other with the MMAD of  $\text{SO}_4^{2-}$ . In the results from Mészáros *et al.* (1997), the  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  both had MMAD of 0.8  $\mu\text{m}$ , consistent with the  $\text{SO}_4^{2-}$  but extremely different to the MMAD of  $\text{NO}_3^-$  found in this study.

### 3.3.3 Cation Size Distribution

Figures 4a and 4b display the mean size distributions of the cation species. Figure 4a

reveals that the size distributions of  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  are unimodal and peak between 3.2-5.6  $\mu\text{m}$ , 0.56-1  $\mu\text{m}$ , 3.2-5.6  $\mu\text{m}$  and 5.6-10  $\mu\text{m}$ , respectively. Meanwhile, the MMAD of  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  is 3.94, 1.08, 4.32 and 4.42  $\mu\text{m}$ , respectively. Besides, the size distribution of  $\text{NH}_4^+$  is bimodal with the major peak occurs between 0.56-1  $\mu\text{m}$ ; while the MMAD of  $\text{NH}_4^+$  is 0.86  $\mu\text{m}$ . As in Fig. 4b, it is found that the size distributions of  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$  and  $\text{Mg}^{2+}$  are unimodal and peak between 3.2-5.6  $\mu\text{m}$ , 0.56-1  $\mu\text{m}$ , 0.56-1  $\mu\text{m}$  and 3.2-5.6  $\mu\text{m}$ , respectively. The MMAD of  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$  and  $\text{Mg}^{2+}$  is 3.96, 0.96, 1.21 and 3.58  $\mu\text{m}$ , respectively. Moreover, the size distribution of  $\text{Ca}^{2+}$  is bimodal and the major peak is between 5.6-10  $\mu\text{m}$  and the minor peak is between 0.32-0.56  $\mu\text{m}$ ; meanwhile the MMAD of  $\text{Ca}^{2+}$  is 3.89  $\mu\text{m}$ . The analytical results indicate that the MMAD of the cation species of the two sampling sites are similar. Mészáros *et al.* (1997) demonstrated that in winter the MMAD of  $\text{NH}_4^+$ ,  $\text{K}^+$  and  $\text{Ca}^{2+}$  was 0.8, 1.4 and 5.6  $\mu\text{m}$ , respectively. The values for  $\text{NH}_4^+$  and  $\text{K}^+$  are mutually consistent but inconsistent with the MMAD of  $\text{Ca}^{2+}$ . Besides, the mean size distribution and the MMAD values for  $\text{Cl}^-$  and  $\text{Na}^+$  measured at the CCRT sampling site are also mutually consistent, showing that the two species share the same source and may be transported from the sea via sea breeze circulation.

#### 4. Conclusions

The main conclusions of this study are shown as follows:

(1) The mean mass concentration for  $\text{PM}_{2.5}$ ,  $\text{PM}_{2.5-10}$  and TSP measured at the CCRT (traffic) sampling site are 54.5, 27.8 and 158  $\mu\text{g m}^{-3}$ , respectively. Additionally, the mean mass concentrations for  $\text{PM}_{2.5}$ ,  $\text{PM}_{2.5-10}$  and TSP measured at the THUC (campus) sampling site are 23.6, 16.3 and 85.6  $\mu\text{g m}^{-3}$ ,

respectively.

- (2) The major water-soluble ions of  $\text{PM}_{2.5}$  are identified as  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ . Meanwhile the dominant species for  $\text{PM}_{2.5-10}$  are  $\text{Cl}^-$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  in central Taiwan, Taichung.
- (3) High correlations are noted among  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  at both the CCRT(traffic) and THUC (campus) sampling sites, suggesting that these species tended to be associated in ambient air. The particulate can take the forms  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$ .
- (4) At the CCRT (traffic) sampling site, the mean size distributions of mass,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  are bimodal while those of  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  are unimodal. Meanwhile, at the THUC (campus) sampling site, the mean size distributions of mass,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  are bimodal while those of  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$  and  $\text{Mg}^{2+}$  are unimodal.

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