



Water-soluble Ions in PM_{2.5} on the Qianhu Campus of Nanchang University, Nanchang City: Indoor-Outdoor Distribution and Source Implications

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ABSTRACT

Ambient PM_{2.5} was sampled in three indoor environments (a common office, a photocopy room and a student dormitory) and one outdoor environment (a rooftop) on a campus of Nanchang University in Nanchang city, China, on June 5–20, 2009. Analysis by ion chromatograph showed the indoor-outdoor differences and relationships of water-soluble ions. The indoor-outdoor distributions indicated that chemicals and household garbage contributed most to the generation of water-soluble ions indoors. The indoor-outdoor relationships of water-soluble ions in PM_{2.5} in the common office and student dormitory were unrelated or weak, which indicated that the ions were produced indoors rather than carried in from outdoors. Correlations between various water-soluble ions in indoor and outdoor PM_{2.5} are discussed here, and it is suggested that the photocopier machine contributed little to the water-soluble ions indoors. Ion balance calculations indicated that the anions and cations in the photocopy room and outdoor environment shared an origin, but part of the anions and cations in the common office room and student dormitory originated indoors. The linear regression slopes (anion/cation) are all lower than 1, with the anion deficits probably affected by the absence of data on F⁻, PO₄³⁻, NO₂⁻, CO₃²⁻ and organic acid salt.

Keywords: Indoor air; Aerosols; Water-soluble ions; Nanchang.

INTRODUCTION

Aerosol particles have many disadvantageous impacts on the global climate, atmospheric environment and even human health (Shine *et al.*, 1999; Polichetti *et al.*, 2009; Jahn *et al.*, 2011), although there is still some uncertainty and complexity about them. Recently, fine particles (PM_{2.5}) have received much more attention than coarse particle (PM₁₀) (Yang *et al.*, 2005; Feng *et al.*, 2007; Li *et al.*, 2008; Li *et al.*, 2009), as fine particles can travel more deeply into the human lungs, reaching the alveolar region of the respiratory system. They also take along higher contributions of toxic species harming health, higher contributions of light absorption species and light scattering species that affect visibility and the climate more than coarse particles (Kok *et al.*, 2006; Polichetti *et al.*, 2009). From this point of view, it is still crucial to know the concentrations, distributions and sources of fine particles to develop effective strategies

for the control of fine aerosol pollution and the removal of associated problems.

Fine particles are generated by many different outdoor sources, such as industrial production processes, biomass burning, automobile exhaust, secondary conversion from gaseous pollutants (Cao *et al.*, 2003), and also indoor sources such as smoking, cooking (Lai *et al.*, 2010), photocopying and other indoor activities (Huang *et al.*, 2011). It has been found that concentrations of indoor air particulates are sometimes much higher than those outdoors (Jones *et al.*, 2000; Huang *et al.*, 2007a). Generally, people spend most of their time indoors, including about 8 hours in indoor working environments, but the majority of particulate concentration data is based on measurements made outdoors (Chan *et al.*, 2003; Huang *et al.*, 2007b). Studies on indoor particle characteristics are urgently needed.

Adverse health and the environmental and climate effects of aerosol particles are derived from the chemical components and properties of those particles (Hueglin *et al.*, 2005; Aldabe *et al.*, 2011). Water-soluble ions are major components of atmospheric aerosols and can comprise up to 60–70% of particulate mass. Major inorganic water-soluble ions are associated with the degradation atmospheric visibility (Cheng *et al.*, 2011), adverse human health effects (Karthikeyan *et*

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et al., 2006) and acidity of precipitation (Kerminen *et al.*, 2001; Soo *et al.*, 2011). Inorganic ions can arise from both natural and anthropogenic sources such as oceans, dust storms, biomass combustion, construction, road traffic, industrial activities or even indoor activities and have been commonly used for the identification of PM sources (Hu *et al.*, 2008; Khan *et al.*, 2010; Deshmukh *et al.*, 2011; Galindo *et al.*, 2011; Zhang *et al.*, 2011).

Many studies on PM_{2.5} have been conducted around the world (Kabindra *et al.*, 2010; Dhananjay *et al.*, 2011). Some have been conducted in China on water-soluble ions in PM_{2.5}, but there have been few in indoor environments, with most focused on ambient air aerosols and insufficient coverage of cities.

The main objective of this study is to provide quantitative information on the indoor-outdoor differences of water-soluble ions in PM_{2.5}, track their sources, and explore relative mechanisms and implications.

METHODOLOGY

Sampling Sites

Nanchang city (115°27'–116°35'E, 28°10'–28°11'N) is in Jiangxi province, China. The area of the city zone is 302 square kilometers with about 2,400,000 residents and 440,000 motor vehicles. There are 60,000 students on the Qianhu Campus of Nanchang University. Samples were taken from four sites on the campus (Fig. 1): one outdoors (Site O1) and other three indoors (Sites I1, I2 and I3). Site O1 was located on the rooftop (about 15 m above the ground) of the Environment Building, with a highway running 100 m from the west of the site, and roads running alongside the campus at distances of 200 m (east) and 500 m (south) from the site. The site thus represented urban residential-commercial-traffic mixed communities in Nanchang city. Site I1 was a common office room for laboratory assistants on the second floor of the Environment Building south-corridor (about 3 m above the ground), with labs as neighboring and opposite rooms. Site I2 was a photocopy room, located in

the north-corridor of the second floor of the Environment Building (about 3 m above the ground). Site I3 was a student dormitory in the Student Apartment Building (about 6 m above the ground). Huang *et al.* (2011) provide further details of the sampling sites.

Sample Collection

Indoor-outdoor PM_{2.5} was measured from June 5–20, 2009 (summer). The samples were collected simultaneously using mainly identical samplers. The samples from Site O1 (outdoors) and Site I1 (indoors) were collected simultaneously using identical samplers from July 5 to 12, 2009, and the samples from Site O1 (outdoors) and Site I3 (indoors) were collected simultaneously using identical samplers from July 13 to 19. A total of 33 PM_{2.5} samples were collected at the outdoor and indoor sites. Mini-volume samplers (Airmetrics, Eugene, USA) with $\Phi 47$ mm quartz microfibre filters (QM/A) (Whatman, Maidstone, UK) were used to collect the samples. The samplers were operated at a flow rate of 5 L/min for 24 hours. The devices were inspected and calibrated at the Institute of Earth Environment, Chinese Academy of Sciences (CAS) before sampling. The indoor sampling height was about 1.3 m above ground to simulate the breathing zone and avoid potential interference from the excessive re-suspension of particles. The filter preparation method, filter weighting method and quality control were the same as those of Huang *et al.* (2011).

Analysis of Water-soluble Ions and Quality Control

A Dionex-600 Ion Chromatograph (Dionex Inc., Sunnyvale, CA, USA) was used for the determination of both cations (Na⁺, NH₄⁺, K⁺, Mg²⁺ and Ca²⁺) and anions (Cl⁻, NO₃⁻ and SO₄²⁻) in the aqueous extracts of the sample filters. After weighting, a quarter of the loaded $\Phi 47$ mm filters were placed in separate 15 mL vials containing 10 mL of distilled-deionized water (18.2 M Ω resistivity) to extract the water-soluble ions from the quartz filters. The vials were placed in an ultrasonic water bath and shaken with a mechanical shaker for 1 hr to extract the ions. The extracts

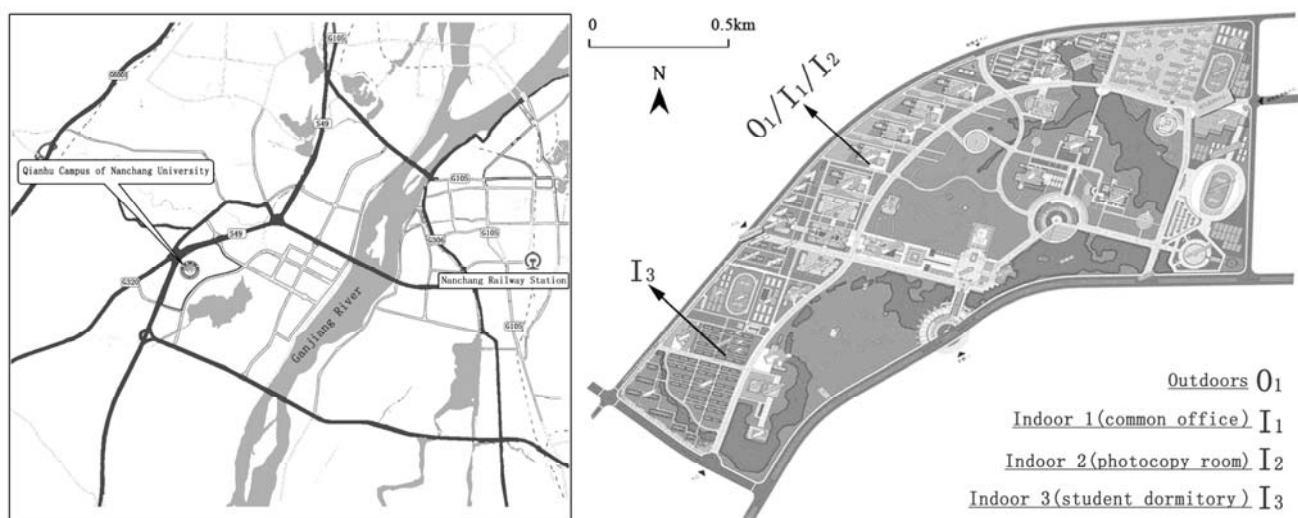


Fig. 1. Sampling sites.

were filtered through 0.45 μm pore size microporous membranes and the filtrates were stored at 4°C in clean tubes before instrumental analysis. For the cation analyses, the instrument was equipped with an IonPacCS12A column (20 mM methanesulfonic acid as the eluent), while for anions an IonPac AS14A column (8 mM Na₂CO₃/1 mM NaHCO₃ as the eluent) was used. The detection limits (MDLs) were as follows: 4.6 mg/L for Na⁺, 4.0 mg/L for NH₄⁺, 10.0 mg/L for K⁺, Mg²⁺ and Ca²⁺, 0.5 mg/L for Cl⁻, 15 mg/L for NO₃⁻ and 20 mg/L for SO₄²⁻. Standard reference materials produced by the National Research Center for Certified Reference Materials (Beijing, China) were analyzed for quality control and assurance.

RESULTS AND DISCUSSION

Levels of Water-soluble Ions in Outdoor PM_{2.5}

The outdoor water-soluble ion concentrations in PM_{2.5} are listed and compared with studies from other cities in Table 1. The concentrations in Nanchang are distinctly higher than those from cities in Japan, Singapore, Spain and Switzerland, and even higher than those in Chinese cities such as Beijing, Shanghai and Guangzhou. Yet in comparison to Xi'an, ammonium (NH₄⁺), chloride (Cl⁻) and nitrate (NO₃⁻) were lower. This indicates the pollution severity of water-soluble ion species in fine particles in the Nanchang urban air environment. However, the ratio of nitrate (NO₃⁻) to sulfate (SO₄²⁻) in Nanchang (0.10) was lower than that in all other cities listed in Table 1. This was the case because the contribution of motor vehicle emissions to PM_{2.5} in Nanchang was lower than that in the other cities.

Indoor-Outdoor Distributions of Water-soluble Ions in PM_{2.5}

Table 2 shows the indoor-outdoor ratios of PM_{2.5} mass and water-soluble ions in PM_{2.5} in the three indoor sites. The indoor mass concentrations were higher than those measured outdoors, and most of the ion species concentrations indoors were higher than outdoors, except for ammonium and potassium at Sites I1 and I2, sulfate at Site I2 and calcium at Site I3. These higher ion concentrations at the indoor sites than at the outdoor site might be partly due to the low air exchange rate in indoor environments. During sampling, the three indoor sites were subjected to natural ventilation. The windows and doors at Sites I1 and I3 were occasionally open in the daytime during occupation, but were closed at night when unoccupied. The windows at Site I2 were always closed, and the door was open in the daytime but closed at night. Although the natural ventilation rates in the three indoor sites were not measured, the air circulation rate at Site I2 was thought to be lower than at the other two indoor sites, as the windows at Site I2 were always closed. Hence, it seemed reasonable that there were existing indoor water-soluble ion sources. Site I1 was a common office in which laboratory assistants implemented paperwork on the second floor of the Environment Building south-corridor, with the neighboring and opposite rooms being labs for teaching and research experiments. During the sampling, a variety of experiments were performed in the rooms, with many

Table 1. Water-soluble ions in ambient PM_{2.5} in Nanchang and other studies (μg/m³).

Nation/ Region	City/Region	Site	Site type	Time	n	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	NO ₃ ⁻ / SO ₄ ²⁻	Reference
	Nanchang	qianhu	mixing	2009.6	8	6.92	4.85	1.18	0.35	2.57	1.62	2.75	28.91	0.10	This study
	Guangzhou	xinken	suburb	2004.10–2004.11	-	-	9.2	-	-	-	2.4	7.2	24.1	0.30	Hu et al., 2008
China	Shanghai	Tongji	Mixing	2000	-	0.7	6.2	1.9	0.3	0.3	1.7	6.5	15.2	0.43	Yao et al., 2002
	Beijing	Tsighua	Mixing		-	0.7	6.5	2.2	0.4	0.8	1.8	9.9	16.9	0.59	
	Xi'an	AEE	Mixing	2006.6–2006.8	-	-	10.5	-	-	-	-	11.8	38.9	0.30	Zhang et al., 2011
Taiwan	Kaohsiung	H-kang	Mixing	1998.11–1999.4	7	2.79	9.54	-	0.09	0.03	3.07	12.78	17.82	0.72	Lin et al., 2002
Japan	Yokohama	YNU	mixing	2007.9–2008.8	-	0.25	2.27	0.13	0.05	0.2	0.21	0.96	3.8	0.57	Khan et al., 2010
Singapore		NUSC	mixing	2000.1–2000.12	-	0.6	1.62	0.53	0.07	0.29	0.6	0.9	5	0.36	Karthikeyan et al., 2006
Spain	Alicante	UA	mixing	2005.9–2006.8	-	0.34	1.52	0.28	0.05	0.75	0.52	1.53	4.2	0.10	Galindo et al., 2011
Switzerland	Bern	City centre,	mixing	1998.4–1999.3	79	0.094	1.6	0.18	0.009	0.132	0.102	3.0	2.8	1.07	Hueglin et al., 2005

Table 2. Ratios of indoor to outdoor concentrations of water-soluble ions in PM_{2.5}.

	Item	PM _{2.5} mass	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
I1/O1 ratio	Common office/Outdoor	1.11	1.32	0.97	0.96	2.20	1.74	1.09	1.55	1.08
I2/O1 ratio	Photocopy room/Outdoor	1.25	1.14	0.81	0.86	1.26	1.09	1.10	1.01	0.73
I3/O1 ratio	Student dormitory/Outdoor	1.41	1.17	1.28	1.20	1.20	0.97	1.36	1.99	1.03

chemicals such as acid, alkali and salt used. Those chemicals could have contributed to indoor PM_{2.5} with the release of water-soluble ion species or precursors for water-soluble ions species. Site I2 was located in the north-corridor of the second floor in the Environment Building, around which lab rooms were seldom used during the sampling time. Hence, the concentrations of water-soluble ion species at the site were lower than those at Site I1. The indoor-outdoor ratios of NH₄⁺, K⁺ and SO₄²⁻ at Site I2 were lower than 1 (0.81, 0.86 and 0.73), indicating that the sources of the three ions were mainly outdoor air penetration rather than indoor photocopiers and printers. Site I3 was a student dormitory in another building, and it also had higher water-soluble ion concentrations than outdoors. The activities and objects in the room and around it in the same corridor, such as house refuse, dust brought in by students, the use of shower gel and so on, were conducive to increases in water-soluble ions in fine particles in the air.

Indoor-outdoor Relationship of Water-soluble Ions in PM_{2.5}

For the samples collected simultaneously at Site O1 (outdoors) and Site I1 (indoors), the indoor-outdoor relationship of water-soluble ions is shown in Fig. 2. The indoor-outdoor relationship of water-soluble ions between Site O1 (outdoors) and Site I3 (indoors) is shown in the Fig. 3. As indicated by Fig. 2 and linear correlation testing, there were no good indoor-outdoor correlations for water-soluble ions between Sites O1 and I1. Even the indoor-outdoor relationships of sodium, chloride and sulfate between Sites O1 and I1, and the indoor-outdoor relationships of sodium, calcium, nitrate and sulfate between Sites O1 and I3 were almost completely unrelated, with the square of the correlation coefficient lower than 0.1. These indoor-outdoor relationships indicated that the indoor and outdoor water-soluble ion components had different sources. The indoor water-soluble ion components were partially delivered by outdoor air penetration, but mainly originated from indoor sources.

Correlation between Various Water-soluble Ions

The correlation coefficient matrix for water-soluble ions in PM_{2.5} outdoors is given in Table 3. As the table shows, the concentrations of SO₄²⁻ and NH₄⁺ were significantly correlated ($R = 0.883$, $P < 0.01$). Similarly, a strong correlation ($R > 0.813$, $P < 0.05$) was observed for the concentrations of NO₃⁻ and NH₄⁺. These good correlations suggest that the species were associated with ambient air and primarily existed as ammonium sulfate ((NH₄)₂SO₄), ammonium bisulfate (NH₄HSO₄), and ammonium nitrate (NH₄NO₃) (Zhang et al., 2011). NO₃⁻ and SO₄²⁻ were highly correlated with a coefficient of 0.796 ($P < 0.05$), suggesting that the precursors of the two species were released from similar emission

sources, such as coal burning, vehicle exhaust or industry. Furthermore, there was a high correlation between Ca²⁺ and SO₄²⁻ ($P < 0.01$) and between Ca²⁺ and NO₃⁻ ($P < 0.01$), indicating there was insufficient ammonia (NH₃) to completely neutralize the sulfuric acid (H₂SO₄) formed from oxidation of SO₂. The calcium could be related to the re-suspension of road dust caused by moving vehicles and exhaust gases. The strong correlations between Mg²⁺ and Ca²⁺, Mg²⁺ and Cl⁻, and Na⁺ and Mg²⁺, indicate that they may be associated with crustal dust. There was strong correlation coefficient between NH₄⁺ and K⁺, suggesting that the two species have similar sources: biomass burning and secondary aerosols.

The correlation coefficient matrixes for water-soluble ions in PM_{2.5} in the three indoor environments are given in Tables 4, 5 and 6. The correlations between various ions at Sites I1 and I3 (Table 4 and Table 6) were not as strong as those outdoors, and there were only significant correlations between three ions. The relative lack of correlation between ion species indicated different emission sources and/or removal mechanism, and possibly different transport patterns for indoor water-soluble ions at Sites I1 and I3. This would be consistent with the weak and unrelated indoor-outdoor relationship of water-soluble ions at the two indoor sites. As Table 5 indicates, the correlation coefficient matrixes for water-soluble ions at Site I2 (photocopy room) were similar to those outdoors (Site O1), indicating that most of the water-soluble ions in the photocopy room originated outdoors.

Ion Balance Calculations

The relationships between anions and cations expressed by the sum of the equivalent concentration ($\mu\text{mol}/\text{m}^3$) are shown in Fig. 4. Cation and anion equivalents were calculated by Eqs. (1) and (2), respectively. Fig. 4 indicates that the correlation coefficients between anion and cation were 0.90 and 0.98 at Sites O1 and I2, respectively, and the correlation coefficients between anion and cation were 0.79 and 0.84 at Sites I1 and I3, respectively. This indicated that the anions and cations shared an origin in Sites O1 and I2, but they originated from different indoor sources at Sites I1 and I3, which can be deduced by the correlation between various water-soluble ions at the four sites.

$$CE(\text{Cation equivalent}) = \frac{Na^+}{23} + \frac{NH_4^+}{18} + \frac{K^+}{39} + \frac{Mg^{2+}}{12} + \frac{Ca^{2+}}{20} \quad (1)$$

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

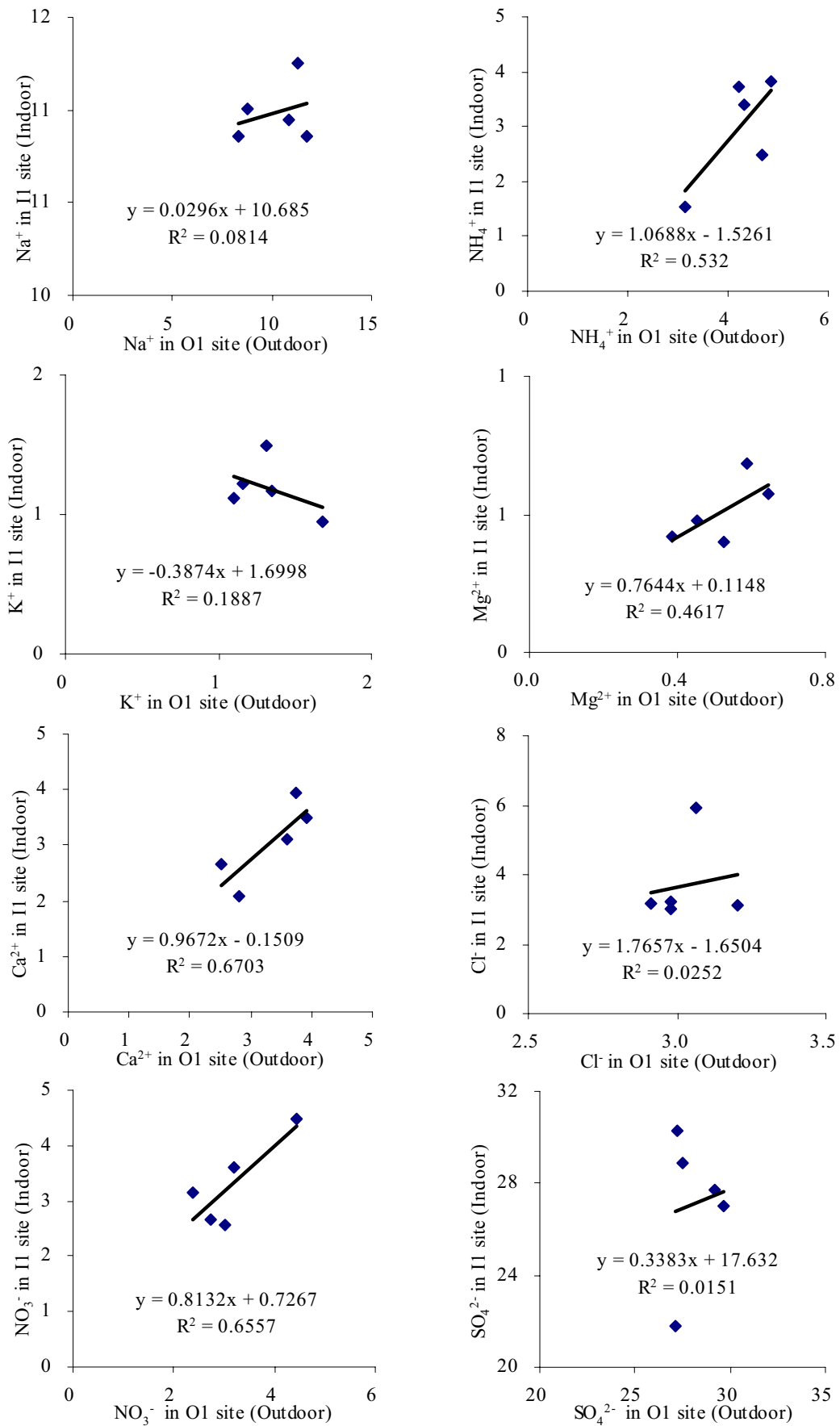


Fig. 2. Indoor-outdoor relationship of water-soluble ions between Site O1 (Outdoors) and Site I1 (Common office).

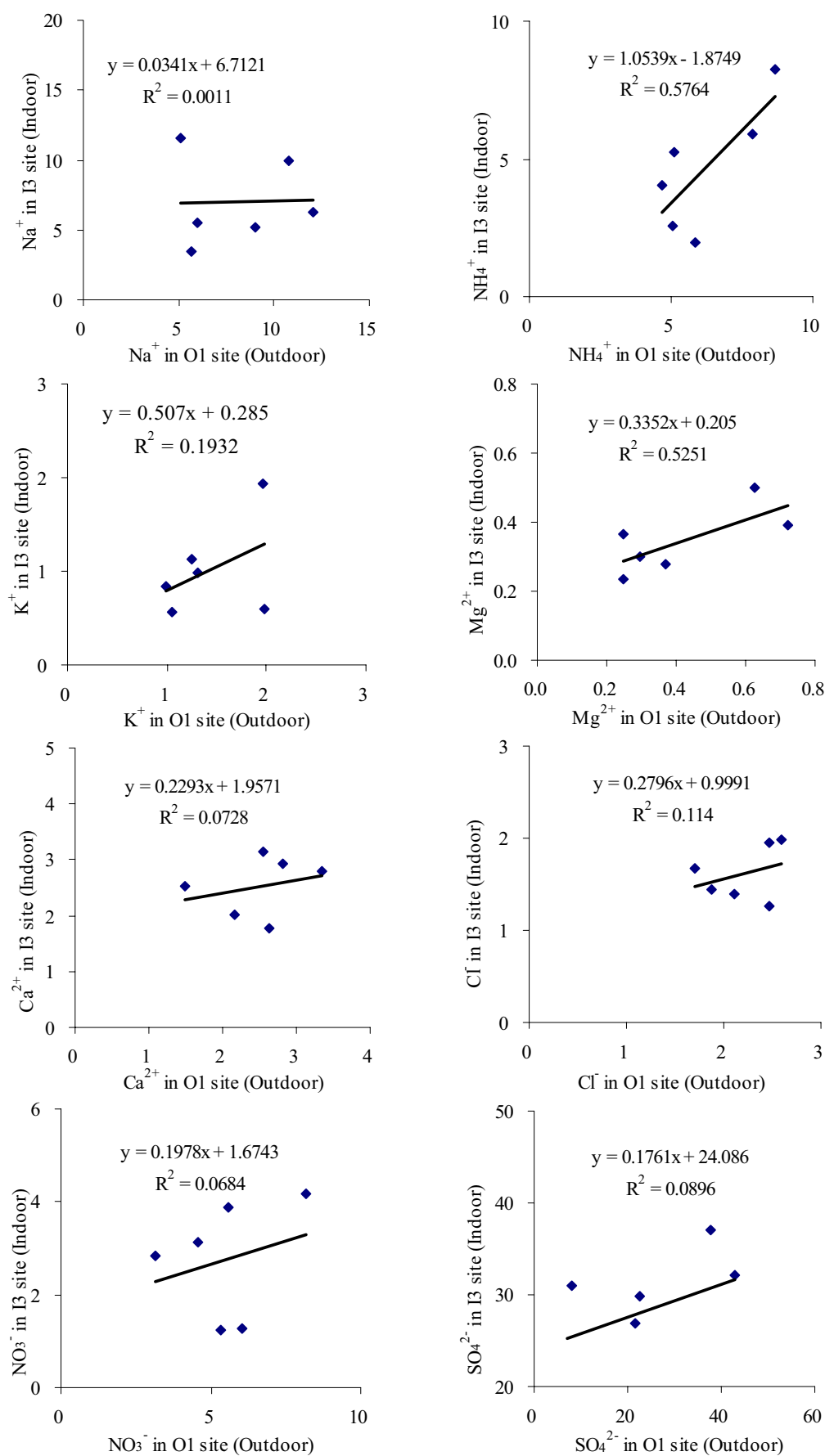


Fig. 3. Indoor-outdoor relationship of water-soluble ions between Site O1 (Outdoors) and Site I1 (Student dormitory).

Table 3. Correlation coefficient matrix for water-soluble ions in PM_{2.5} of Site O1 (Outdoors).

	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Na ⁺	1	.070	-.106	.719*	.261	.532	.519	.354
NH ₄ ⁺		1	.817*	.415	.857**	-.050	.813*	.883**
K ⁺			1	.116	.584	-.199	.535	.577
Mg ²⁺				1	.756*	.727*	.652	.592
Ca ⁺					1	.339	.742*	.886**
Cl ⁻						1	.257	.064
NO ₃ ⁻							1	.796*
SO ₄ ²⁻								1

n = 8; *Correlation is significant at the 0.05 level (2-tailed). ** Correlation is significant at the 0.01 level (2-tailed). It is same as in Table 4, 5 and 6.

Table 4. Correlation coefficient matrix for water-soluble ions in PM_{2.5} of Site I1 (Common office).

	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Na ⁺	1	-.460	.525	.851	.799	-.263	.458	.564
NH ₄ ⁺		1	.460	-.598	-.046	.889	.503	.302
K ⁺			1	.414	.552	.681	.996**	.965*
Mg ²⁺				1	.370	-.224	.383	.583
Ca ²⁺					1	-.100	.475	.426
Cl ⁻						1	.737	.626
NO ₃ ⁻							1	.969*
SO ₄ ²⁻								1

Table 5. Correlation coefficient matrix for water-soluble ions in PM_{2.5} of Site I2 (Photocopy room).

	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Na ⁺	1	.666	.812	.980**	.482	.580	.911*	.980**
NH ₄ ⁺		1	.250	.720	.866	.763	.904*	.797
K ⁺			1	.806	.286	.552	.631	.700
Mg ²⁺				1	.617	.682	.940*	.975**
Ca ²⁺					1	.914*	.772	.598
Cl ⁻						1	.796	.637
NO ₃ ⁻							1	.965**
SO ₄ ²⁻								1

Table 6. Correlation coefficient matrix for water-soluble ions in PM_{2.5} of Site I3 (Student dormitory).

	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Na ⁺	1	-.089	-.413	.883*	-.061	.589	-.047	.341
NH ₄ ⁺		1	.565	.200	.702	.582	.703	.895*
K ⁺			1	-.159	.754	.416	.566	.283
Mg ²⁺				1	.362	.810	-.026	.574
Ca ²⁺					1	.744	.406	.595
Cl ⁻						1	.454	.752
NO ₃ ⁻							1	.547
SO ₄ ²⁻								1

The slopes (anion/cation) of the linear regression are all lower than 1, and the anion deficits are thought to have been affected by the absence of data on F⁻, PO₄³⁻, NO₂⁻, CO₃²⁻, and organic acid salt.

CONCLUSIONS

Water-soluble ion concentrations were high in Nanchang, but the ratio of NO₃⁻ to SO₄²⁻ was low (< 0.15) given the low contribution of motor vehicle emissions to PM_{2.5}. Most of the ion species concentrations indoors were higher than outdoors, especially at Site I1 (Common office) and Site I3 (Student dormitory), indicating that the indoor ion species were affected by outdoor air penetration and indoor

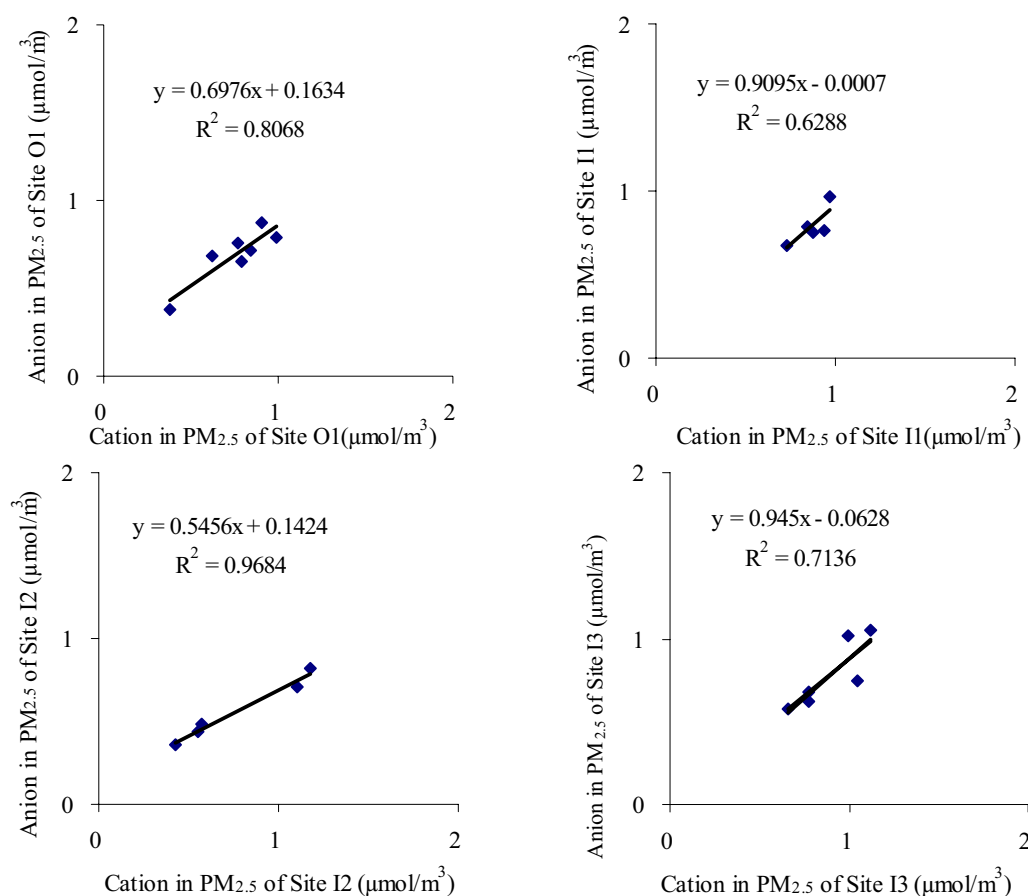


Fig. 4. Anion-cation relationships at different sites.

pollution sources. It was thought chemical experiments on a lab room and indoor household garbage contributed to water soluble ion species indoors.

Unrelated or weak indoor-outdoor relationships at Sites I1 and I3 indicated different sources of contributions to indoor and outdoor particle water-soluble ions and that the two sites had indoor water-soluble ion sources.

The correlation coefficient matrix for water-soluble ions in ambient PM_{2.5} and at Site I2 (photocopy room) suggested that the ion species primarily originated from coal burning, vehicle exhaust, industry, crustal dust, re-suspension from road dust, biomass burning and secondary aerosols. The lack of correlation between various ion species for indoor PM_{2.5} at Sites I1 and I3 indicated different production and/or removal mechanisms, and possibly different transport patterns.

The relationships between anions and cations indicated that they shared an origin at Sites O1 and I2, but they originated from different sources at Sites I1 and I3. The slopes (anion/cation) of the linear regression were all lower than 1, and the anion deficits have probably been affected by the absence of data on F^- , PO_4^{3-} , NO_2^- , CO_3^{2-} , and organic acid salt.

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