

Characteristics of Water-soluble Inorganic Ions in Aerosol and Precipitation and their Scavenging Ratios in an Urban Environment in Southwest China

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ABSTRACT

Daily fine particulate matter (PM_{2.5}) and precipitation samples were collected simultaneously at an urban site in southwest China in four segregated months in 2015 for measuring major water-soluble inorganic ions (WSIIs). Online hourly concentrations of PM₁₀ and PM_{2.5} were also monitored, which showed annual mean concentrations of 67.8 and 41.6 $\mu\text{g m}^{-3}$, respectively. PM_{2.5} showed the highest concentration in winter and lowest in summer. The annual mean concentration of the total WSIIs was 20.3 $\mu\text{g m}^{-3}$, accounting for about 48.7% of PM_{2.5}. Among the total WSIIs in ambient PM_{2.5}, SO₄²⁻ was the predominant component (49.7%), followed by NH₄⁺ (24.1%) and NO₃⁻ (21.4%). NH₄⁺ and SO₄²⁻ were the two most abundant ions in precipitation, followed by Ca²⁺ and NO₃⁻. Seasonal patterns of the major inorganic ions in precipitation were similar to those in PM_{2.5}, with the highest concentration in winter and lowest in summer. The mean scavenging ratios were 454, 445, 364, 456, and 394 for SO₄²⁻, NO₃⁻, NH₄⁺, Cl⁻, and K⁺, and 116, 353, and 18 for gas SO₂, HNO₃, and NH₃, respectively. The higher scavenging ratios of particulate ions than their gaseous precursors suggest the higher contributions of particles than gases to the total wet deposition.

Keywords: Wet scavenging, Water-soluble inorganic ions, PM_{2.5}, Precipitation chemistry

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
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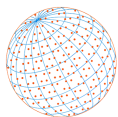
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1 INTRODUCTION

Wet scavenging efficiently removes particulate matter and their gaseous precursors from the atmosphere (Wang *et al.*, 2018a). In-cloud (rainout) and below-cloud (washout) scavenging are the two components of wet scavenging processes for air pollutants. For particulate matter, rainout and washout mechanisms are different, with rainout involving mostly nucleation of aerosols (acting as cloud condensation nuclei) and to a much less extent impaction scavenging, whereas washout involving mainly impaction scavenging (collected by falling raindrops) (Zhang and Vet, 2006). For gases, dissolution in liquid water droplets is the mechanism for both rainout and washout processes. The relative contributions of rainout and washout to the total wet deposition could be estimated based on the sequential precipitation sampling measurements (Aikawa and Hiraki, 2009). For example, using this method, Xu *et al.* (2017) estimated that washout contributed about 56%, 61%, and 47% to total wet scavenging of SO₄²⁻, NO₃⁻, and NH₄⁺ respectively, in Beijing, and Ge *et al.* (2016) obtained 88%, 92%, and 84% in Dalian and 56%, 50%, and 46% in Dandong in North China. Furthermore, washout accounted for about two-thirds of total wet scavenging for NO₃⁻ and half for SO₄²⁻ in Japan using the same method (Aikawa *et al.*, 2014; Kajino and Aikawa, 2015).



Scavenging ratio (W), defined as the ratio of a pollutant concentration in precipitation to that in air, can be a useful indicator for wet scavenging efficiency (Engelmann, 1971). This parameter depends on the physical and chemical characteristics of both particles and precipitation, and can vary by up to two orders of magnitude with different chemical species. Budhavant *et al.* (2020) estimated the scavenging ratios of ten major chemical species in PM_{10} over the Northern Indian Ocean, and obtained values of less than 50 for black carbon and up to 3000 for Cl^- and NO_3^- . Cheng and Zhang (2017) investigated the scavenging ratios of eight inorganic ions at 13 monitoring sites across Canada and obtained values varying by a factor of up to 6, with larger values for chemical species with higher fractions in coarse particles, e.g., Ca^{2+} and NO_3^- . The important role that below-cloud scavenging plays on ambient aerosol concentration under different rain conditions has also been demonstrated by field experiments and numerical studies (Zhang *et al.*, 2004; Gao *et al.*, 2019; Lu *et al.*, 2019; Luan *et al.*, 2019).

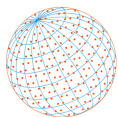
Due to the strong dependence of scavenging ratio on the characteristics of particles and precipitation, existing database are subject of large uncertainties. Scavenging ratio obtained from parallel measurements of ambient particulate matter and precipitation chemistry over long sampling periods may reduce such uncertainties. Furthermore, it is even more complicated for wet-scavenged chemical species involving both particulate- and gaseous-phase pollutants in air, such as SO_4^{2-} , NO_3^- , and NH_4^+ . Kasper-Giebl *et al.* (1999) estimated that 89–96%, 4–12%, and 49–79% of SO_4^{2-} , NO_3^- , and NH_4^+ in precipitation were from the scavenging of their ambient particulate-phase ions, respectively, with the rest from the gaseous pollutants (SO_2 , HNO_3 , and NH_3). Similarly, Cheng and Zhang (2017) suggested that HNO_3 dominated the particulate NO_3^- in wet scavenging with a percentage contribution of about 72%, whereas the contributions from particulate SO_4^{2-} and NH_4^+ were greater than the corresponding gas SO_2 and NH_3 , with an average percentage contribution of 63% and 70%, respectively. Comprehensive investigation on the relative contributions from gases and particles to the total wet deposition may improve the wet scavenging models of SO_4^{2-} , NO_3^- , and NH_4^+ , and consequently reduce the uncertainties in prediction of particle concentrations during precipitation periods.

Wanzhou is a small urban city, situated in southwest China. The climate in Wanzhou is characterized by high relative humidity, extremely low wind speeds all year round, and abundant rainfall with annual precipitation of about 1200 mm. Thus, wet scavenging is expected to play an important role in removing pollutants from the atmosphere. Previous studies have focused on the dry and wet deposition fluxes of nitrogen or other pollutants in southwest China (Wang *et al.*, 2016; Wang *et al.*, 2018a), but provided no information on the wet scavenging processes or scavenging ratios of water-soluble inorganic ions. The present study aims to fill this knowledge gap by simultaneously measuring $PM_{2.5}$ and precipitation chemistry in different seasons of a year. The main objectives are to (1) investigate the dynamic characteristics of $PM_{2.5}$ and PM_{10} during rainfall processes, (2) characterize major water-soluble inorganic ions in $PM_{2.5}$ and precipitation, and (3) estimate the scavenging ratios of major inorganic ions and gaseous precursors as well as their relative contributions.

2 METHODS

2.1 Sampling and Chemical Analysis

$PM_{2.5}$ and precipitation samples were collected in parallel in Wanzhou. The sampling site is located on the roof of a building inside the Chongqing Three Gorges University ($108^{\circ}13'E$, $30^{\circ}48'N$), about 28 m above the ground, which is influenced by local vehicular emission and point sources (Wang *et al.*, 2016). 23-h $PM_{2.5}$ samples were collected by an air pollutant sampler (URG Corp., URG-3000K, North Carolina, USA) from 11:00 a.m. to 10:00 a.m. the next day at a flow rate of $15 L min^{-1}$. Two channels of the air pollutant sampler were used to collect $PM_{2.5}$ samples in parallel. The left channel was preceded by two annual denuders coated with glycerol/ Na_2CO_3 and glycerol/citric acid solution for trapping HNO_3 and NH_3 sequentially, and then a Teflon filter was equipped for mass weighting. The right channel was loaded with quartz filter for water-soluble inorganic ions analysis. Daily precipitation samples were collected using an automatic atmospheric deposition sampler equipped with a rain gauge sensor (APS-3A, Xianglan Scientific Instruments Co., Ltd., China). Wanzhou has a subtropical monsoon climate with temperature above $0^{\circ}C$ all



year round, hence, precipitation at this location is almost always in rain. Although rainfall samples were collected on a daily basis in order to match PM_{2.5} sampling campaign, the rain gauge sensor could automatically record the detailed information of each rainfall event, including the beginning and ending time, besides the rainfall amount. Sampling campaigns were conducted in four months in 2015: from 2nd to 29th in April, 2th to 30th in July, 16th October to 13th November, 16th December to 14th January in 2016, representing spring, summer, autumn, and winter, respectively. Note that PM_{2.5} samples were not available from 2nd to 7th July due to the sampler maintenance. PM_{2.5} samples were stored in the dark and at -18°C until analysis to prevent the evaporation of volatile compounds, while rainfall samples were filtered through a 0.45 μm filter after collection and refrigerated at 4°C.

Five cations (Na⁺, NH₄⁺, K⁺, Mg²⁺, Ca²⁺) and three anions (Cl⁻, SO₄²⁻, NO₃⁻) were determined by an ion chromatography (DX-600, Dionex, Sunnyvale, CA) for both PM_{2.5} and rainfall samples. Cations were measured using a CS12A column with 20 mM methanesulfonic acid (MSA) as an eluent while anions were analyzed using an AS11-HC column with 30 mM KOH as an eluent. Ions in rainfall samples were measured directly, while PM_{2.5} samples should be extracted using ultrapure water before ion chromatography analysis. HNO₃ and NH₃ captured by denuders were extracted using ultrapure water first, and then measured by an ion chromatography. A detailed description of PM_{2.5} and rainfall sampling as well as water-soluble inorganic ions analysis were presented in Wang *et al.* (2016, 2018b). For Na⁺ in PM_{2.5}, the field blanks varied largely and might result in large uncertainties. For Mg²⁺ and Ca²⁺, most of PM_{2.5} samples were below the detection limits due to their relatively low concentrations. Hence, only five water-soluble inorganic ions in PM_{2.5} including SO₄²⁻, NO₃⁻, NH₄⁺, Cl⁻, and K⁺ were discussed in the following section.

Hourly concentrations of PM_{2.5} and PM₁₀ were measured using online PM_{2.5}/PM₁₀ analyzer (FH62C14, Thermo Fisher, U.S.). Hourly concentrations of SO₂ were not available at the sampling site and obtained from an air quality monitoring station about 6 km away.

2.2 Data Analysis

The rate of change in particle concentration (ΔC) for each rainfall event was defined as follows:

$$\Delta C(\%) = \frac{(C_b - C_d)}{C_b} \times 100\% \quad (1)$$

where C_b is the average concentration of PM_{2.5} or PM₁₀ within 3 h before the rain, C_d is the average concentration during the rain. If the value of ΔC is positive, it means that PM_{2.5} or PM₁₀ decreases when rainfall occurs, otherwise, it indicates that PM_{2.5} or PM₁₀ increases during rain. Calculation of ΔC requires that the free-rainfall period between two events is higher than 3 h, otherwise, consecutive rainfall events are combined (Aikawa *et al.*, 2014). As a consequence, a total of 7, 8, 14, and 13 rainfall events were obtained in spring, summer, autumn, and winter, respectively (Table S1).

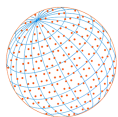
Scavenging ratio of major inorganic ions in each month was calculated based on monthly mean concentration as follows (Kasper-Giebl *et al.*, 1999; Cheng and Zhang, 2017):

$$W = \frac{C_{prec}}{C_{air}} \times \frac{\rho_a}{\rho_w} \quad (2)$$

where C_{prec} is the volume weighted mean (VWM) concentration of inorganic ions in precipitation (mg L⁻¹), C_{air} is the mean concentration of water-soluble inorganic ions in the air (μg m⁻³), and ρ_a and ρ_w are the density of air (1200 g m⁻³) and water, respectively.

SO₄²⁻, NO₃⁻, and NH₄⁺ in rainfall originated from both particles and gaseous precursors. Hence, it is assumed that the difference between the total and particulate wet scavenging was from the gaseous precursor. Scavenging ratio of gaseous precursor was calculated followed by Cheng and Zhang (2017). The procedure for estimating the scavenging ratio of HNO₃ was described in detail, and the calculation for the scavenging ratio of SO₂ and NH₃ was done similarly.

The particulate NO₃⁻ concentration in rainfall was first estimated as follows:



$$[\text{pNO}_3^-]_{\text{prec}} = W_{\text{fPM}}[\text{pNO}_3^-]_{\text{fPM}} + W_{\text{cPM}}[\text{pNO}_3^-]_{\text{cPM}} \quad (3)$$

where $[\text{pNO}_3^-]_{\text{prec}}$ is the part of NO_3^- wet scavenging from particulate NO_3^- . W_{fPM} and W_{cPM} are monthly scavenging ratio of fine and coarse particles, respectively. W_{fPM} could be calculated from the scavenging ratio of K^+ , while W_{cPM} could be calculated based on the average scavenging ratio of Ca^{2+} , Mg^{2+} , and Na^+ . In this study, the coarse particles data were not available, thus the contributions from coarse particles were neglected. $[\text{pNO}_3^-]_{\text{fPM}}$ and $[\text{pNO}_3^-]_{\text{cPM}}$ are monthly mean concentration of NO_3^- in fine and coarse particles, respectively. Note that the concentrations of major inorganic ions in coarse particles were not available, alternative data assumptions were made, which were discussed in the following section in detail. Then, the scavenging ratio of HNO_3 and its contribution were calculated as follows:

$$[\text{HNO}_3]_{\text{prec}} = [\text{total NO}_3^-]_{\text{prec}} - [\text{pNO}_3^-]_{\text{prec}} \quad (4)$$

$$W_{\text{HNO}_3} = \frac{[\text{HNO}_3]_{\text{prec}}}{[\text{HNO}_3]_{\text{air}}} \quad (5)$$

$$\% \text{HNO}_3 = \left(\frac{[\text{HNO}_3]_{\text{prec}}}{[\text{total NO}_3^-]_{\text{prec}}} \right) \times 100 \quad (6)$$

$$\% \text{pNO}_3^- = \left(\frac{[\text{pNO}_3^-]_{\text{prec}}}{[\text{total NO}_3^-]_{\text{prec}}} \right) \times 100 \quad (7)$$

where $[\text{total NO}_3^-]_{\text{prec}}$ means the monthly VWM concentrations of NO_3^- in rainfall, $[\text{HNO}_3]_{\text{prec}}$ is the concentration of NO_3^- in rainfall from gas contribution, and $[\text{HNO}_3]_{\text{air}}$ is ambient concentration of gas HNO_3 . $\% \text{HNO}_3$ and $\% \text{NO}_3^-$ are the contributions of HNO_3 and particulate NO_3^- to the total NO_3^- wet scavenging, respectively. In some cases, the calculated $[\text{HNO}_3]_{\text{prec}}$ value was negative, then we assumed that only particulate NO_3^- contributed to the wet scavenging and the contribution from gas HNO_3 was negligible.

3 RESULTS AND DISCUSSION

3.1 Variations of Hourly Particulate Matter Concentrations during Rain Events

During the observation periods, the accumulated rainfall amounts were 79.2, 152.8, 85.8, and 31.7 mm in spring, summer, autumn, and winter, respectively, while the corresponding total rainfall hours were 57.1, 61.7, 38.2, and 53.3 h. Note that the rainfall data were obtained in a typical month rather than three months in each season. In Wanzhou, rainfall was the most abundant in summer and least in winter. Although the total rainfall hours were similar in spring, summer, and winter, the accumulated amounts varied about five times due to the different rainfall intensities. Rain could be classified into three categories based on the rainfall intensity (Luan *et al.*, 2019): light rain ($0.1\text{--}2.5 \text{ mm h}^{-1}$), moderate rain ($2.6\text{--}7.6 \text{ mm h}^{-1}$), and heavy rain ($> 7.6 \text{ mm h}^{-1}$). Regarding the rain events recorded by rain gauge sensor, the predominant rain category was light rain in all seasons, in particular in spring and winter where the numbers of light rain events accounted for about 96% and 100% of the total events, respectively. Moderate and heavy rain events contributed to about 15.8% and 17.9% of the total events in summer, and 5.3% and 3.6% in autumn, respectively.

Particulate matter concentrations were affected by many factors, such as source emissions and wet scavenging effects. Fig. 1 shows the hourly concentrations of $\text{PM}_{2.5}$, PM_{10} , and rain intensities during the observation periods. For those rainfall events with relatively long durations and/or high rainfall intensities, $\text{PM}_{2.5}$ and PM_{10} concentrations decreased greatly during rain, e.g., on 19th April, 15th July, and 25th October in 2015, and 7th January 2016, indicating that wet scavenging

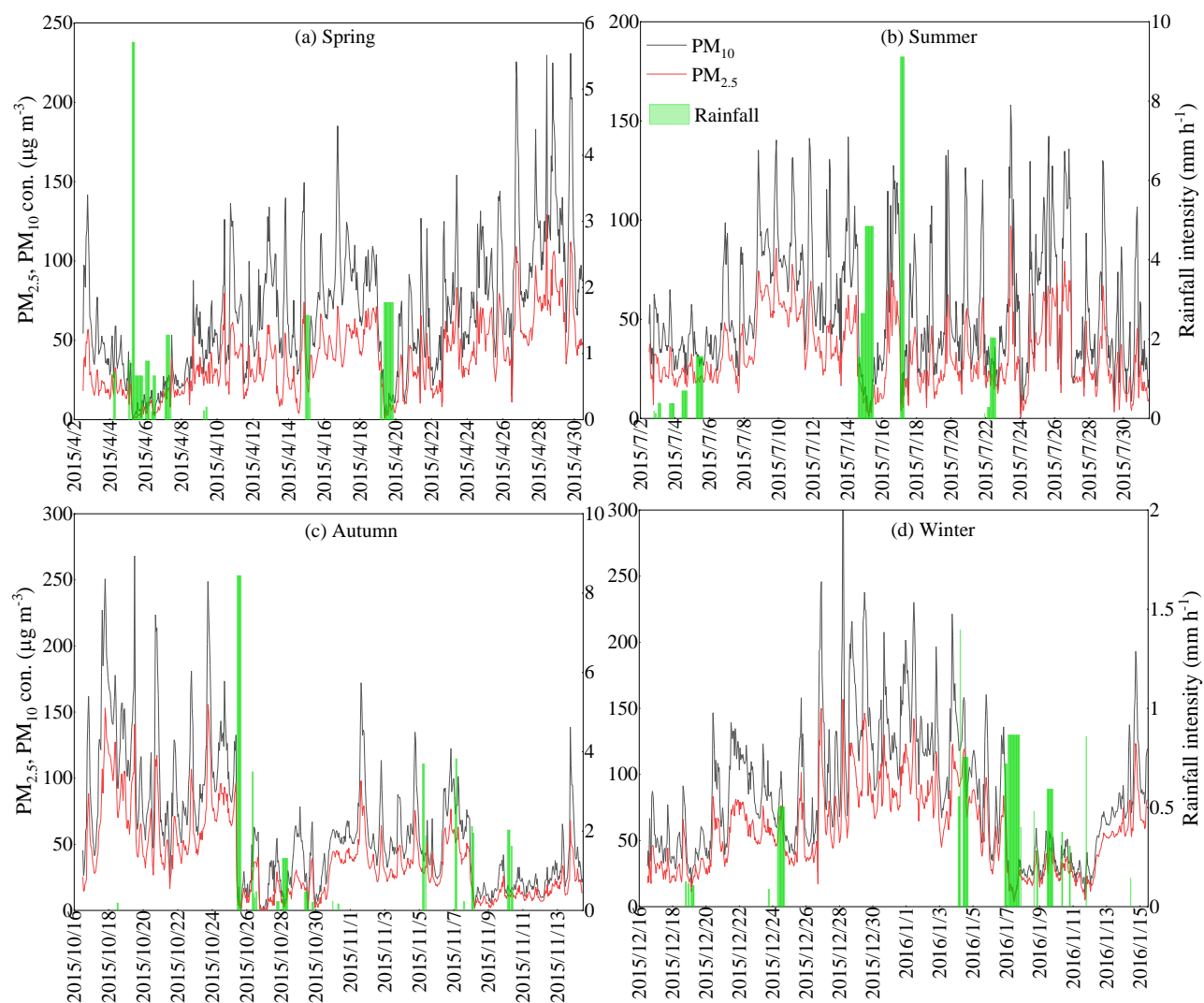
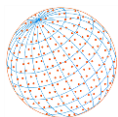
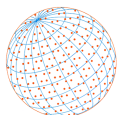


Fig. 1. Hourly concentrations of $PM_{2.5}$, PM_{10} , and rainfall intensity in spring, summer, autumn, and winter.

played an important role on the decreases of the $PM_{2.5}$ and PM_{10} concentrations. However, for those rainfall events with short durations or low intensities, the scavenging effects exhibited large variations, implying that other factors would primarily contribute to the variations of particle concentrations. For example, on 21st July with the rain duration of 0.8 h and rain intensity of 0.13 mm h⁻¹, $PM_{2.5}$ and PM_{10} concentrations decreased by 39% and 48% during rain, respectively. We found that this phenomenon was caused by the increased $PM_{2.5}$ and PM_{10} concentrations during the rush hours in the evening, and when the rush hours ended after 21:00 local time, the air pollutants concentrations began to decrease. This can be supported by the increased NO_2 concentrations during the rush hours and the decreased concentrations one or two hours before the rain. That means the decreases of $PM_{2.5}$ and PM_{10} concentrations on this day were ascribed to the emission reduction after rush hours rather than the wet scavenging effects. On 24th December, $PM_{2.5}$ and PM_{10} concentrations increased by about 27% during rain period although it lasted about 8 hours from 7:00 to 17:00 local time. The reason for this phenomenon was similar to the rain event on 21st July, which was caused by the high emissions during the rush hours when rain occurred, resulting in relatively high average $PM_{2.5}$ and PM_{10} concentrations. Furthermore, no evident variations of $PM_{2.5}$ and PM_{10} concentrations were observed before and during rain on 7th April, in which the duration was about 5 h and rainfall intensity was greater than 1.0 mm h⁻¹. In total, those distinct scavenging effects for different rainfall events indicated that other factors also affected the overall scavenging effects besides rainfall duration and intensities.



The variations of PM_{2.5} and PM₁₀ concentrations were not uniform for each event, showing increasing or decreasing trends when rainfall occurred (Fig. 2). For PM₁₀, almost all events showed decreasing trends in spring. In other seasons, the number of events with decreasing trends accounted for about 62.5–71.4% of the total rain events. For PM_{2.5}, the number of events presenting decreasing trends accounted for 50–64.3% of the total rainfall events, implying the important role of wet scavenging effects on decreasing particle concentrations.

Generally, the decreasing rates of PM₁₀ by rainfall were higher than PM_{2.5} for every event (Fig. 2), which can be explained by the size-dependent below-cloud impaction scavenging efficiency (Wang *et al.*, 2010). Besides particle size distribution, removal rates are also related to the concentrations of particles before rain. The rates of change in particle concentrations were grouped with PM₁₀ or PM_{2.5} concentrations before rain at 20 µg m⁻³ interval bins. As shown in Fig. 3(a), ΔC typically increased with the increases of PM₁₀ and PM_{2.5} concentrations, and the highest change rate in PM₁₀ and PM_{2.5} concentrations appeared in the range of 80–100 µg m⁻³ and 60–80 µg m⁻³, respectively. This trend was also observed by Olszowski (2016), in which the scavenging effect was well correlated with higher particle concentrations before rain. Note that ΔC showed decreasing trends when PM₁₀ and PM_{2.5} concentrations were higher than 100 µg m⁻³. This phenomenon indicated that other meteorological conditions might govern the severe PM₁₀ and PM_{2.5} pollution and then the wet scavenging effect was overwhelmed.

It has been reported that the rainfall intensity also influenced the variations of particle concentrations (He and Balasubramanian, 2008; Luan *et al.*, 2019). As shown in Fig. 3(b), ΔC was highest (higher than 50% for both PM₁₀ and PM_{2.5}) when rainfall intensities were higher than 2 mm h⁻¹, indicating that moderate or heavy rain exhibited relatively high scavenging efficiency on particulate matter. This can also be explained by the precipitation-intensity dependent below-cloud scavenging efficiency (Wang *et al.*, 2010). Scavenging effects were positive for each moderate or heavy rainfall event with the value of ΔC in the range of 8.3–90.9% for PM₁₀ and 2.6–93.5% for PM_{2.5}, respectively. However, no obvious trends of ΔC were observed for light rains, likely because other factors overwhelmed the below-cloud scavenging effect.

3.2 Gaseous Precursors and Water-soluble Inorganic Ions in PM_{2.5} and Rainfall

Among the three gaseous precursors discussed here, the annual mean concentrations were the lowest for HNO₃, followed by SO₂, and highest for NH₃ (Table 1). From a seasonal perspective, SO₂ showed the highest concentration in spring and lowest in summer. SO₂ is primarily emitted from coal combustion for residential heating, power plants and industrial use in China. Wanzhou is located in southwest China with a temperature of above 0°C all year round, and residential heating using coal may not be extensive in this region. Thus, the seasonal patterns of SO₂ were

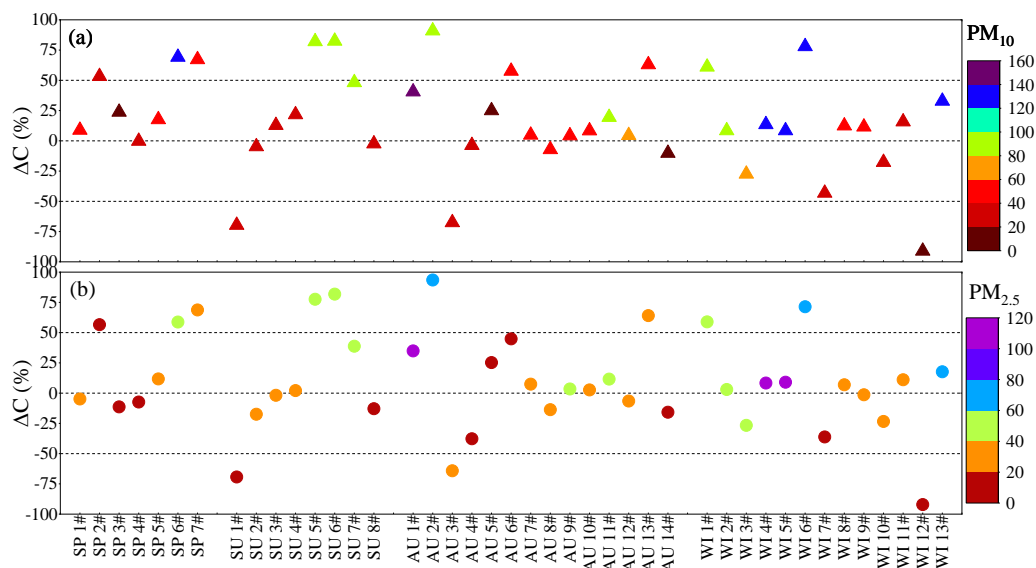


Fig. 2. The variations of PM₁₀ (a) and PM_{2.5} (b) concentrations for each rainfall event.

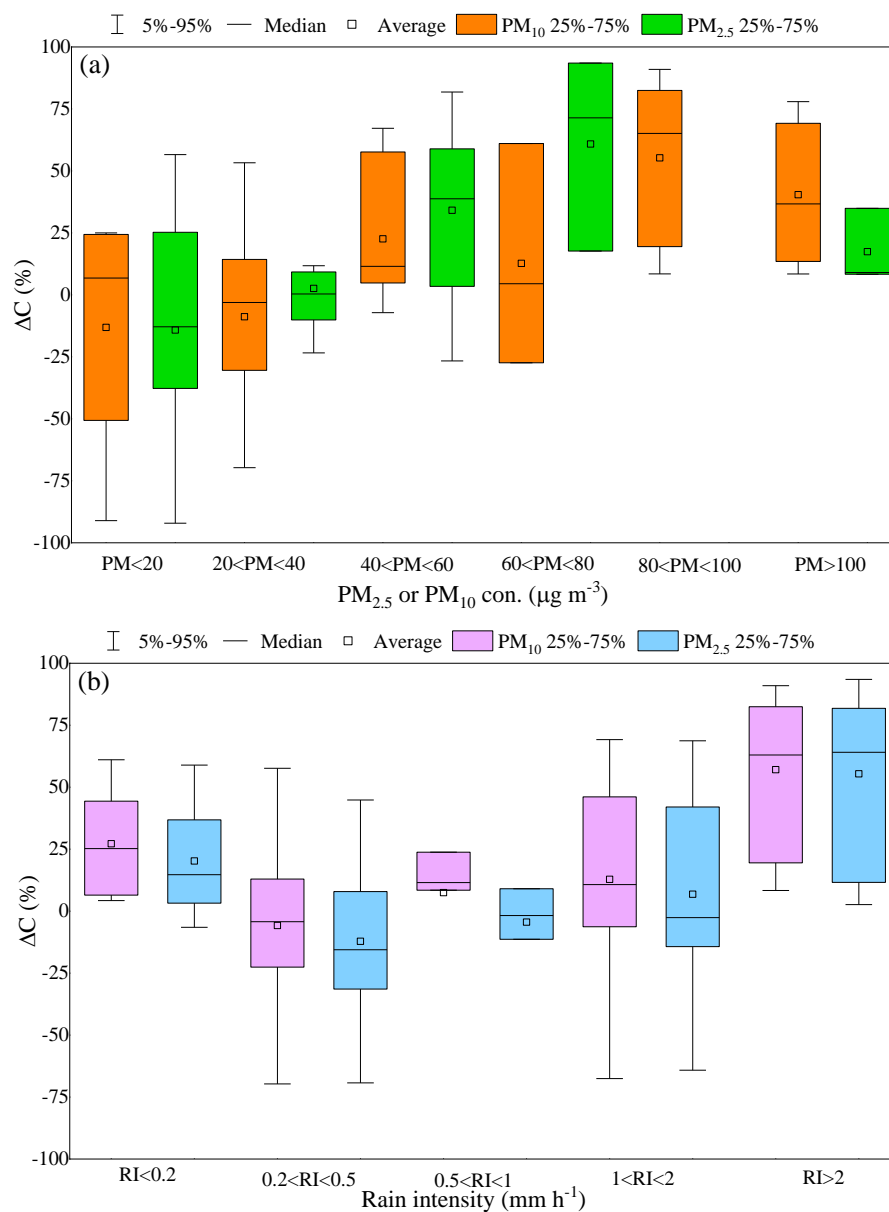
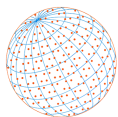
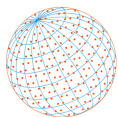


Fig. 3. The variations of PM_{10} and $PM_{2.5}$ grouped by particle concentrations before rain (a), and rain intensity bins (b).

Table 1. Seasonal and annual concentrations of gaseous precursors and major inorganic ions in $PM_{2.5}$ and precipitation.

	In $PM_{2.5}$ ($\mu\text{g m}^{-3}$)					In precipitation ($\mu\text{eq L}^{-1}$)							
	Cl^-	SO_4^{2-}	NO_3^-	NH_4^+	K^+	Cl^-	SO_4^{2-}	NO_3^-	NH_4^+	K^+	Ca^{2+}	Na^+	Mg^{2+}
Spring	0.27	8.1	2.9	4.1	0.35	4.8	92.5	30.4	78.6	4.6	74.4	3.1	10.5
Summer	0.05	8.9	1.3	3.4	0.23	4.2	50.3	19.1	70.1	1.7	29.3	2.9	4.3
Autumn	0.59	7.5	3.7	3.9	0.41	6.7	116.6	33.8	104.6	4.9	104.9	54.7	8.8
Winter	1.3	15.4	8.6	7.7	0.58	23.0	146.0	51.3	103.6	6.1	76.3	12.1	15.1
Annual	0.59	10.1	4.3	4.9	0.40	6.3	79.4	25.9	82.2	3.4	54.6	13.6	7.1
	SO_2 ($\mu\text{g m}^{-3}$)				HNO_3 ($\mu\text{g m}^{-3}$)			NH_3 ($\mu\text{g m}^{-3}$)					
Spring	14.8				1.4			14.2					
Summer	9.3				2.8			16.3					
Autumn	11.5				1.0			14.1					
Winter	12.3				1.4			8.9					
Annual	12.0				1.6			13.1					



mainly influenced by the meteorological conditions and chemical transformations rather than source emissions. High temperature and strong solar radiation in summer could enhance photochemical reactions, leading to the decrease of gaseous precursor concentration such as SO_2 and the increase of the secondary aerosol such as SO_4^{2-} . In addition, the high planetary boundary layer height in summer favors the pollutants dispersion and then leads to the low SO_2 concentration. Contrary to the case of SO_2 , HNO_3 and NH_3 exhibited the maximum concentrations in summer and lowest values in winter and autumn, which were likely associated more with the source emission and thermodynamic behavior of NH_4NO_3 than other conditions (Wang *et al.*, 2018b).

The annual mean concentration of $\text{PM}_{2.5}$ was $41.6 \pm 24.5 \mu\text{g m}^{-3}$, with the highest concentration in winter and lowest in summer. The total concentration of the five major water-soluble inorganic ions (WSIIs) was $20.3 \mu\text{g m}^{-3}$, accounting for about 48.7% of $\text{PM}_{2.5}$. All five water-soluble inorganic ions displayed the highest concentrations in winter (Fig. 4 and Table 1), which were partly related to the unfavorable diffusion conditions, i.e., extremely weak wind ($< 1 \text{ m s}^{-1}$) and low precipitation amount. In addition, the relative humidity was generally higher than 80% in winter, which was conducive to secondary inorganic aerosol formation through heterogeneous reactions and thus increased the concentrations of SO_4^{2-} , NO_3^- , and NH_4^+ . Biomass burning for residential heating in winter probably contributed to the higher concentrations of Cl^- and K^+ . The lowest concentrations of the five inorganic ions appeared in summer due to the favorable diffusion conditions and abundant precipitation with the exception of SO_4^{2-} , which was in autumn. Furthermore, the pronounced high concentrations of NO_3^- in winter and extremely low concentrations in summer were governed by the thermodynamic equilibrium of NH_4NO_3 , since the low temperature and high relative humidity in winter were conducive to the formation of particulate NO_3^- , whereas high temperature in summer enhanced the volatilization of NH_4NO_3 (Wang *et al.*, 2018b). Compared with the other inorganic ions, the relatively higher concentration of SO_4^{2-} in summer was mainly ascribed to the intense photochemical reactions as mentioned above. In $\text{PM}_{2.5}$, the winter/summer ratios were 1.7, 6.7, 2.3, 25, and 2.5 for SO_4^{2-} , NO_3^- , NH_4^+ , Cl^- , and K^+ , respectively, indicating that NO_3^- and Cl^- presented the largest seasonal variations.

Among the five major water-soluble inorganic ions in $\text{PM}_{2.5}$, SO_4^{2-} is the most abundant ions, showing seasonal mean contributions of 46–64% to WSIIs during the sampling periods, implying the important contributions of coal combustion to $\text{PM}_{2.5}$ pollution. Besides SO_4^{2-} , NO_3^- and NH_4^+ were also predominant components of WSIIs. In autumn and winter, NO_3^- and NH_4^+ showed comparable contributions, each of which was about half of that of SO_4^{2-} . However, SO_4^{2-} , NO_3^- , and NH_4^+ presented distinct contributions to WSIIs in summer, where the percentage of SO_4^{2-} was about 3 times that of NH_4^+ and even 7 times higher than that of NO_3^- . The relatively high contribution of SO_4^{2-} and quite low contribution of NO_3^- were consistent with the patterns of their concentrations. The fractions of Cl^- and K^+ in WSIIs were minor, ranging from 0.37% in summer to 3.8% in winter for Cl^- and from 1.6 to 2.6% for K^+ .

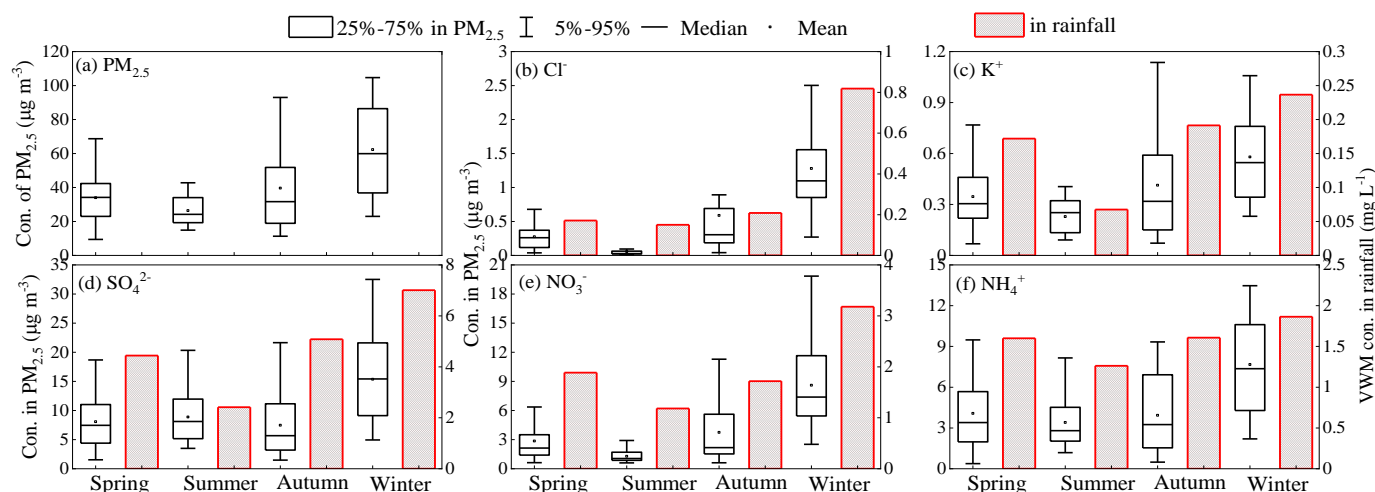
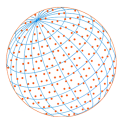


Fig. 4. Seasonal patterns of (a) $\text{PM}_{2.5}$, (b) Cl^- , (c) K^+ , (d) SO_4^{2-} , (e) NO_3^- , and (f) NH_4^+ in $\text{PM}_{2.5}$ as well as the seasonal VWM concentrations of inorganic ions in rainfall.



As shown in Fig. 4 and Table 1, the seasonal variations of eight inorganic ions in rainfall followed the patterns of those in PM_{2.5}, showing the highest VWM concentrations in winter and lowest in summer. Besides the relatively low concentrations of pollutants in the air, dilution effect due to the abundant rainfall was also responsible for the lower concentrations of those inorganic ions in summer. Compared the winter/summer ratios of the inorganic ions in PM_{2.5} and rainfall, it could be found that the seasonal variations were more pronounced in PM_{2.5} than in rainfall except for SO₄²⁻, implying the sensitivity of the air pollutants to the meteorological conditions. Although Ca²⁺ concentrations were very low in PM_{2.5}, they were almost comparable to NH₄⁺ in rainfall in spring and autumn, highlighting a high contribution from coarse particles to rainfall. On an equivalent amount basis, rainfall was dominated by NH₄⁺ and SO₄²⁻, followed by Ca²⁺ and NO₃⁻ (Table 1). The distribution of inorganic ions abundance in rainfall at Wanzhou was similar to Hangzhou (Han *et al.*, 2019), but slightly different from the mean value across China which showed SO₄²⁻ and Ca²⁺ as the most abundant species (Li *et al.*, 2019b).

3.3 Scavenging Ratios of Major Inorganic Ions

Calculating scavenging ratios of inorganic ions and gaseous pollutants required more parameters, including the concentrations of major inorganic ions in fine particles, coarse particles and rainfall, and the concentrations of SO₂, HNO₃, and NH₃. Fine/coarse fractions of these ions were not measured in the present study, but were previously reported for urban sites in Chongqing (Li *et al.*, 2018a, b), which can be considered to be representative of the location of the present study. Briefly, Mg²⁺ and Ca²⁺ were mainly distributed in coarse particles, SO₄²⁻, NH₄⁺, and K⁺ were primarily in fine particles, and Na⁺, NO₃⁻, and Cl⁻ were distributed in both fine and coarse particles (Table S2). In this study, the concentrations of inorganic ions in coarse particles were not available. Considering the relatively high fractions of coarse particles for Mg²⁺, Ca²⁺, and Na⁺, the scavenging ratios of these ions will be highly uncertain if their concentrations in coarse particles were ignored. Meanwhile, the scavenging ratio of coarse particles (W_{CPM}) cannot be obtained due to the lack of coarse particles data, since W_{CPM} was determined by averaging scavenging ratios of Na⁺, Mg²⁺, and Ca²⁺ (Cheng and Zhang, 2017). Nevertheless, it is assumed that the influences due to the lack of coarse particles data would be minor on those inorganic ions dominated in fine particles. Thus, the scavenging ratios were only analyzed for SO₄²⁻, NO₃⁻, NH₄⁺, Cl⁻, and K⁺ using their concentrations in PM_{2.5}. Taking into account the fractions of K⁺ in fine particles around 80%, the scavenging ratio of K⁺ multiplied by 0.8 was assumed to be the scavenging ratio of fine particle (W_{fPM}).

Seasonal and annual mean scavenging ratios of five inorganic ions and three gases are summarized in Table 2. On an annual basis, SO₄²⁻, NO₃⁻, and Cl⁻ showed comparable scavenging ratios at around 450, which was about 13–25% higher than the values for NH₄⁺ and K⁺. As shown in Fig. S1, positive correlations of equivalent concentrations between [NH₄⁺] and [SO₄²⁻ + NO₃⁻] were observed in the four seasons, with the correlation coefficients higher than 0.90 and the slopes of linear regressions being around 1.0, suggesting that SO₄²⁻ and NO₃⁻ were fully neutralized by NH₄⁺, and (NH₄)₂SO₄ and NH₄NO₃ were the two major chemical forms in PM_{2.5}. Consequently, it seems that SO₄²⁻, NO₃⁻, and NH₄⁺ should have consistent scavenging ratios due to their coexistence in fine particles. However, slightly higher values of SO₄²⁻ and NO₃⁻ than NH₄⁺ were observed. As mentioned above, about 30%–40% of Cl⁻ and NO₃⁻ were found in coarse particles, thus the relatively higher scavenging ratio of NO₃⁻ was in part ascribed to the contributions from coarse particles. The slightly higher scavenging ratio of SO₄²⁻ was probably attributed to the in-cloud rainout process, since SO₄²⁻ could be formed efficiently through in-cloud oxidation of SO₂ by H₂O₂ or O₃, and then the cloud droplets including SO₄²⁻ could be removed as the raindrop falls. Based on Eqs. (2)–(5), the annual mean scavenging ratio was 116, 353, and 18 for SO₂, HNO₃, and NH₃, respectively. The different scavenging ratios of the three gases could be partly explained by their different solubilities. HNO₃ is the most soluble species, followed by NH₃, and SO₂ is the least soluble gas, with a Henry's law constant of around 2.1×10^3 , 6.1×10^{-1} , and 1.2×10^{-2} mol m⁻³ pa⁻¹, respectively (Sander, 2015). NH₃ solubility is higher than SO₂, their relative low scavenging ratio might be responsible for the short lifetime of NH₃. The air pollutants incorporated into precipitation occurs at cloud level, however, the ambient concentrations were measured at the ground. Over this vertical distance, NH₃ may transform into particulate NH₄⁺ and consequently lead to the wet scavenging mainly from particulate NH₄⁺ rather than gas NH₃.

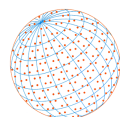


Table 2. Scavenging ratios of inorganic ions and gaseous precursors on mass basis in literatures.

Sampling site	Period	Scavenging ratio								Reference
		$W_{\text{SO}_4^{2-}}$	$W_{\text{NO}_3^-}$	$W_{\text{NH}_4^+}$	W_{Cl^-}	W_{K^+}	W_{SO_2}	W_{HNO_3}	W_{NH_3}	
Wanzhou, China (urban)	2015 spring	660	792	471	747	597	100	637	0	This study
	2015 summer	326	1100	445	3532	355	40	373	34	
	2015 autumn	815	551	491	426	556	242	387	13	
	2015 winter	547	443	292	768	490	194	326	0	
	2015 annual	454	445	364	456	394	116	353	18	
Mt. Sonnblick, Europe (mountain)	Nov. 1991–Nov. 1993	1680	3120	2160	n.a.	n.a.	n.a.	n.a.	n.a.	Kasper–Giebl <i>et al.</i> (1999) ^a
Salvatierra, Spanish (rural)	Jul. 1992–Jun. 1993	1000	3000	2000	10000	2000	n.a.	n.a.	n.a.	Encinas and Casado (1999)
Vitoria, Spanish (urban)	Jan. 1995–Dec. 2000	2830	2330	1739	3030	1151	n.a.	n.a.	n.a.	Encinas <i>et al.</i> (2004)
AIRMoN 14 sites, U.S.	A decade	672	n.a.	372	n.a.	n.a.	n.a.	588	n.a.	Hicks (2005) ^a
Singapore (urban)	Jan.–Sep. 2006	2596	2134	1660	2624	744	n.a.	n.a.	n.a.	He and Balasubramanian (2008)
CAPMON, Canada (13 sites, rural)	between 1984 and 2011	471–	583–	283–	1855–	497–	125–	749–	n.a.	Cheng and Zhang (2017)
		734	1224	848	5779	996	1339	5527		
Hyderabad, India	Aug. 2006	160	317	389	109	723	n.a.	n.a.	n.a.	Kulshrestha <i>et al.</i> (2009)
Maldives	May 2014–Apr. 2015	203–	1300–	251–	1019–	554–	n.a.	n.a.	n.a.	Budhavant <i>et al.</i> (2020) ^b
		696	3045	771	3273	1488				
Beijing, China (urban)	Jun.–Aug. 2014	312	420	168	n.a.	n.a.	n.a.	n.a.	n.a.	Xu <i>et al.</i> (2017) ^a

n.a. no available data.

^a convert to the scavenging ratio on mass basis; ^b calculated scavenging ratio based on the concentrations in aerosol and precipitation.

Different seasonal patterns of the scavenging ratios were observed for different inorganic ions and gases. Seasonal variations in scavenging ratios are caused by many factors, such as ambient concentrations of the ions, rain intensities and associated precipitation chemistry and droplet spectra. These factors would affect the dissociate rate of related pollutants between ambient air and rain droplets, below-cloud evaporation of droplets, etc., all of which would contribute to the overall calculated scavenging ratio. The maximum scavenging ratio was observed in autumn for both SO_4^{2-} and NH_4^+ , and the minimum value appeared in summer and winter, respectively. In contrast, the highest scavenging ratio was found in summer for Cl^- and NO_3^- and lowest in autumn and winter, respectively. The highest scavenging ratios of SO_2 , HNO_3 , and NH_3 were observed in autumn, spring, and summer, respectively.

The scavenging ratio values of SO_4^{2-} , NH_4^+ , and K^+ obtained in the present study generally agreed well with existing data reported in literature (Table 2); however, those of NO_3^- were slightly lower than reported in the other regions, which was probably related to the different size distributions among different regions, among other factors. Contrary to the predominant coarse mode distribution of NO_3^- influenced by natural sources or sea salt in other regions (Zhang *et al.*, 2008), about 70%–80% of NO_3^- was found in fine mode in the study region, which might result in lower scavenging ratio since rainfall scavenges coarse particles more efficiently. Additionally, the sampling site in this study is an urban site, which is severely affected by the vehicle emissions and hence results in relatively high ambient concentrations of NO_2 and NO_3^- . For the three gases, limited NH_3 scavenging ratios were reported in the previous studies. The scavenging ratio of SO_2

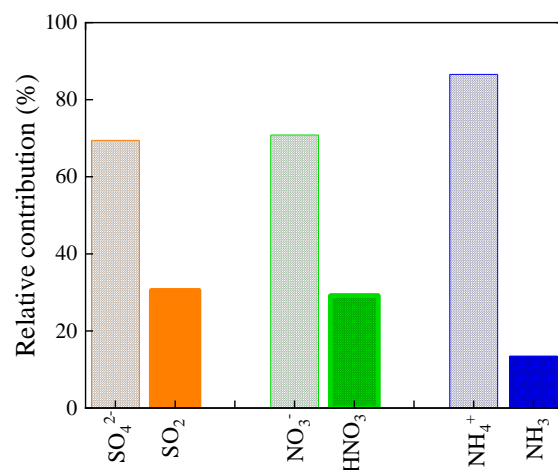
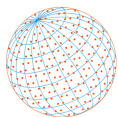


Fig. 5. Relative contributions of particulate and gas-phase to the total wet scavenging of SO₄²⁻, NO₃⁻, and NH₄⁺ during the sampling periods.

in this study was within the range of those values in the literatures, while the value for HNO₃ was somewhat lower in this study (Table 2).

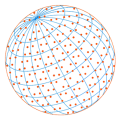
As expected, particulate SO₄²⁻ was the dominant contributor to rainfall SO₄²⁻, accounting for 69.4%, while gas SO₂ contributed about 30.6% (Fig. 5), which were consistent with those conducted at rural sites in Canada (Cheng and Zhang, 2017). Similar to SO₄²⁻, particulate NO₃⁻ accounted for about 70% of the rainfall NO₃⁻, which was much higher than those reported by Cheng and Zhang (2017) (around 28%) and Kasper-Giebl *et al.* (1999) (about 4–12%). The discrepancies between our results and other literatures were probably attributed to the different characteristic of sampling location, where the sampling site with higher ambient NO₃⁻ concentrations exhibited high relative contributions of particulate NO₃⁻. The relative contribution of particulate NH₄⁺ to total wet scavenging was 86.6%, which was higher than those in Cheng and Zhang (2017) (70%) and Kasper-Giebl *et al.* (1999) (48–79%).

4 CONCLUSIONS

Water-soluble inorganic ions in ambient fine particles and in precipitation were characterized, and relative contributions from particulate and gaseous pollutants to inorganic ions in precipitation were quantified. PM_{2.5} and PM₁₀ concentrations were generally decreased after occurrence of moderate or heavy rain event, but this was not always the case after light rain event due to too many other factors affecting the ambient concentration. The scavenging ratio values derived for the five inorganic ions that were mostly distributed in fine particles varied by 25% on average, e.g., about 450 for SO₄²⁻, NO₃⁻, and Cl⁻, 364 for NH₄⁺, and 395 for K⁺, likely due to their slightly different fine/coarse fractions. Scavenging ratios for gaseous species investigated here differed greatly, e.g., 353 for HNO₃, 116 for SO₂, and 18 for NH₃, likely related to their different solubility or lifetime in air. Besides, major sources and chemical processes might have contributed to the ambient concentrations of some chemical species and thus substantially lowered the scavenging ratio, e.g., for the case of NH₃. Seasonal variations of the scavenging ratios were a factor of 1.7–8 for different air pollutants. Wet scavenging of SO₄²⁻, NO₃⁻, and NH₄⁺ were primarily from particulate forms (> 69%), especially considering that only fine particles were considered in this estimation due to the lack of the coarse particles data. The scavenging ratio data obtained in this study may be used for improving the prediction of particle concentrations during rain periods using air quality models.

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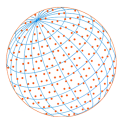
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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <https://doi.org/10.4209/aaqr.200513>

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