Below-Cloud Aerosol Scavenging by Different-Intensity Rains in Beijing City

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ABSTRACT

Below-cloud aerosol scavenging process by precipitation is important for cleaning the polluted aerosols in the atmosphere, and is also a main process for acid rain formation. However, the related physical mechanism has not been well documented and clarified yet. In this paper, we investigated the below-cloud $PM_{2.5}$ (particulate matter with aerodynamic diameter being 2.5 μm or less) scavenging by different-intensity rains under polluted conditions characterized by high $PM_{2.5}$ concentrations, based on in-situ measurements from March 2014 to July 2016 in Beijing city. It was found that relatively more intense rainfall events were more efficient in removing the polluted aerosols in the atmosphere. The mean $PM_{2.5}$ scavenging ratio and its standard deviation (SD) were 5.1% ± 25.7%, 38.5% ± 29.0%, and 50.6% ± 21.2% for light, moderate, and heavy rain events, respectively. We further found that the key impact factors on below-cloud $PM_{2.5}$ scavenging ratio for light rain events were rain duration and wind speed rather than raindrop size distribution. However, the impacts of rain duration and wind speed on scavenging ratio were not important for moderate and heavy rain events. To our knowledge, this is the first statistical result about the effects of rain intensity, rain duration, and raindrop size distribution on below-cloud scavenging in China.

Key words: $PM_{2.5}$, below-cloud scavenging, rain intensity, impact factors

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1. Introduction

The wet scavenging process is an important mechanism for cleaning the polluted atmosphere, which can be classified as in-cloud and below-cloud scavenging processes (Seinfeld and Pandis, 2006). The in-cloud scavenging process is that aerosol particles may serve as cloud condensation nuclei (CCN)/ice nuclei (IN), or they could be directly captured by cloud drops in a cloud (Zhang et al., 2004, 2006; Andronache et al., 2006). The below-cloud scavenging process is that particles below cloud-base are scavenged by the falling rain (Andronache, 2003; Chate et al., 2011; Zhang et al., 2013; Zhao et al., 2015). With regard to human health and air quality in the earth atmosphere, the below-cloud scavenging process may be more important than the in-cloud scavenging process, because polluted aerosol particles could be eventually removed from air and transmitted to the ground in the former process (Bae et al., 2006; Chate, 2011).

The actual below-cloud aerosol scavenging process by rain is usually determined by scavenging coefficient that is defined as the fraction of aerosol particles captured by raindrops per unit of time. The scavenging coefficient has been determined theoretically or measured by observing the changes in atmospheric aerosol concentration during rain (Davenport and Peters, 1978; Andronache, 2003, 2004; Laakso et al., 2003; Chate and Pranesha, 2004; Feng, 2007; Croft et al., 2009). Unfortunately, the scavenging coefficient derived by the two methods has great difference for aerosol particles with

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diameters ranging from 0.01 to 3 μm (Andronache et al., 2006; Wang et al., 2010, 2011).

The key limitation for theoretical approach is accurately obtaining the collection efficiency (Andronache et al., 2006). This is a poorly known parameter, although many experimental and theoretical studies have been conducted (Lai et al., 1978; Andronache, 2004; Ladino et al., 2011; Quérel et al., 2014a, b; Ardon-Dryer et al., 2015; Lemaitre et al., 2017). Several mechanisms and forces were found to influence the aerosol particles collection process by water drops, such as inertial impaction, Brownian diffusion, interception, electro-scavenging, thermophoresis, and diffusiophoresis (Wang and Pruppacher, 1977; Pruppacher and Klett, 1997; Ardon-Dryer et al., 2015).

These mechanisms have been found to depend on the collected aerosol particle size. For coarse aerosol particles (diameter > 2 μm), the inertial impaction was dominant, while for Aitken nuclei mode aerosol particles (diameter < 0.1 μm), the Brownian diffusion was dominant. The processes of thermophoresis and diffusiophoresis had important impact on the collection process of accumulation mode aerosol particles (diameter 0.1–2 μm), but the collection efficiency of these processes was minimum; this was called the "Greenfield gap" (Greenfield, 1957). Electro-scavenging also had an important contribution while the droplet and aerosol particles had opposite polarity (Tinsley et al., 2001, 2006; Tinsley, 2010). In addition, other factors such as drop size, particle density, turbulence, and relative humidity have been also found to affect collection efficiency (Byrne and Jennings, 1993; Ardon-Dryer et al., 2015). Aerosol particles in Aitken nuclei mode were found to be collected more efficiently by small-size raindrops rather than by large-size raindrops. For aerosol particles in coarse mode, there were fewer differences in collection efficiency due to the dominance of inertial term (Andronache, 2003).

The limitation for measuring the scavenging coefficient in atmosphere is that the change of aerosol concentration is assumed to be only dependent on the collection process by raindrops. The processes of advection, turbulent diffusion, coagulation, and the hygroscopic behaviour of aerosol particles could not be avoided even though the rain cases are carefully selected (Laakso et al., 2003). The field measurements for investigating the scavenging process of aerosol particles by rain have been conducted under various atmospheric conditions (Laakso et al., 2003; Chate and Pranesha, 2004; Maria and Russell, 2005; Andronache et al., 2006; Olszowski, 2016; Zikova and Zdimal, 2016). The results showed that the below-cloud scavenging process was important for cleaning aerosol particles in Aitken nuclei mode and coarse mode, and the coarse aerosol particles could be removed fast even in small rain intensity ($R \sim 1$ mm h^{-1}). Besides, the scavenging effect is more significant with larger aerosol concentrations in atmosphere before the rain (Olszowski, 2016). In the previous field studies, the observation environment was usually clean or dusty, and the researchers concerned the aerosol size-dependent scavenging coefficient calculated by the aerosol number concentration. Moreover, most of observations were based on the individual case studies.

Because of the "Greenfield gap" effect, the scavenging process of aerosol particles in accumulation mode was generally considered to be less efficient. Castro et al. (2010) pointed out that the aerosol concentrations increased during the rain event with rain intensity less than 0.6 mm h^{-1}. Andronache (2003) indicated that the below-cloud scavenging coefficient varied about two orders of magnitude for different rainfall rates within 0.01–100 mm h^{-1}, indicating that the below-cloud aerosol scavenging process is sensitive to the rain intensity. Thus, further investigation of the below-cloud scavenging process by different-intensity rains is necessary.

Air pollution events characterized by high $PM_{2.5}$ (particulate matter with aerodynamic diameter of 2.5 μm or less) concentration have frequently occurred in China during the last few decades, especially in the most developed and populated cities (Zhang and Cao, 2015), causing low visibility, adverse effects on health (Tie et al., 2009; Chen R. J. et al., 2013; Guo, 2016), and changes of weather and climate (Qian et al., 2006; Liao et al., 2015; Yang et al., 2016; Cai et al., 2017; Xu et al., 2017). In the mean time, the serve and persistent pollution events provide ideal cases for investigating below-cloud aerosol scavenging by rain (Luan et al., 2018).

Some recent studies in China have shown that the polluted aerosols could be evidently removed by precipitation (Feng and Wang, 2012; Zhao et al., 2015; Guo et al., 2016). However, the results are generally qualitative and the mechanisms involved are not well known. To fill this gap, we use long-term field data observed in Beijing city during March 2014–July 2016 by the Chinese Academy of Meteorological Sciences (CAMS) to quantitatively investigate the below-cloud aerosol scavenging process by different-intensity rains. The mechanism and impacts from raindrop size distribution, rain duration, and wind speed are also studied.

2. Instruments and methods

2.1 *Sampling site*

The observational site was built on the roof of a 20-m tall building (39°57′N, 116°20′E) at the campus of China Meteorological Administration (CMA) in Beijing city, northern China. It is located in the northwest part of urban Beijing, close to the West 3rd Ring Road, without any major sources nearby. Detailed descriptions of the site can be found in Luan et al. (2018). We used data of rain measured by laser disdrometer (Thies Clima, Germany) and $PM_{2.5}$ mass concentration measured by TEOM (Tapered Element Oscillating Microbalance) 1405-DF dichotomous ambient particulate monitor (Thermo Fisher Scientific, USA) from March 2014 to July 2016 at the observational site. The wind data are from Haidian automatic weather station (39°58′N, 116°17′E). The two sites are within a direct distance of 6 km, as shown in Fig. 1.

2.2 *Instruments*

The TEOM 1405-DF dichotomous ambient particulate monitor with FDMS (Filter Dynamics Measurement System) can continuously measure particulate matter mass concentration. The monitor draws ambient air through two TEOM filters at constant flow rate, continuously weighing the filters and calculating near real-time mass concentrations of both $PM_{2.5}$ and PM_{coarse} (2.5–10 μm). The FDMS unit dries the sample flow and automatically generates mass concentration measurement that account for both nonvolatile and volatile particulate matter components. The mass concentration is a 1-h average updated every 6 minutes. The resolution is 0.1 μg m^{-3}, and the precision is ± 2.0 μg m^{-3}. The filters were replaced when the filter loading percentage was near 100%.

The Thies laser disdrometer can simultaneously count and measure the size and fall velocity of hydrometerors, which has been used in many studies (Bloemink and Lanzinger, 2005; Fernández-Raga et al., 2009; de Moraes Frasson et al., 2011; Jameson et al., 2015; Chen et al., 2016). The laser source emits parallel light-beam in 780 nm. The measured rain particle diameter is in the range 0.125–8 mm and divided into 22 classes; the fall speed of rain particle is in the range 0–10 m s^{-1} and divided into 20 classes. The types of rain such as drizzle, rain, hail, snow, and mixed rain, are determined from the statistic proportion of all particles referring to diameter and velocity. Rain with a temperature of above 9°C is automatically accepted as liquid (exception: soft hail and hail), and with a temperature of below –4°C as solid. The calculated data are memorized over 1 min. The resolution of rain intensity is 0.001 mm h^{-1}. To minimize the measurement errors caused by strong winds, splashing, or margin fallers, particles outside ± 60% of the relationship of raindrop fall speed and diameter [$v = 965 - 1030\exp(-6D)$] (Atlas et al., 1973) were removed from the observed data, and the data containing fewer than 10 drops or with a rainfall rate less than 0.002 mm h^{-1} were also removed (Friedrich et al., 2013; Chen et al., 2016, 2017).

2.3 *Data processing*

The number concentration of rain particles was calculated based on the data measured by Thies disdrometer using Eq. (1),

$$N(D_i)=\sum_{j=1}^{20}\frac{n_{ij}}{A\Delta t V_j \Delta D_i}, \tag{1}$$

where n_{ij} denotes drop number for size bin i and velocity bin j, A (in m^2) for sampling area, Δt (in s) for sampling time, V_j (in m s^{-1}) for the fall speed of bin j, D_i (in mm) for the raindrop diameter of bin i, and ΔD_i (in mm) for the diameter interval. $N(D_i)$ (in m^{-3} mm^{-1}) is the number concentration of raindrops per unit volume with diameters from D_i to $D_i + \Delta D_i$.

Rainfall rate (R) data were collected every minute. In order to match the $PM_{2.5}$ mass concentration data, the minute rainfall rate was calculated into hourly rainfall rate, which is the rainfall amount in 1 h. Generally, a rainy hour is that the hourly rainfall amount is equal or larger than 0.1 mm. The rainfall rate represents the

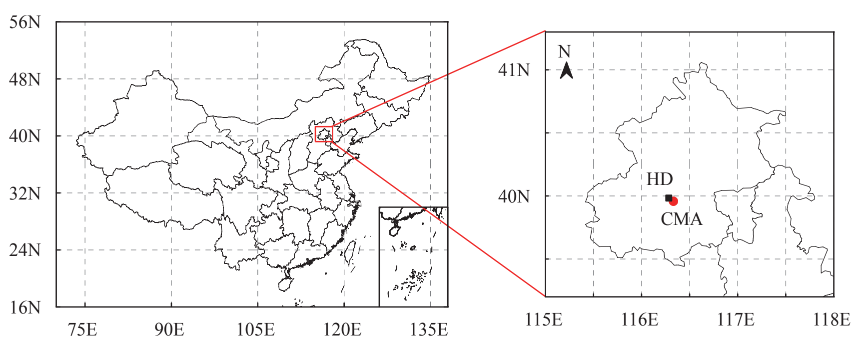

Fig. 1. Geographical location of the observational sites. CMA and HD represent China Meteorological Administration and Haidian automatic weather station, respectively.

rain intensity.

For the convenience of calculation and analysis, the $PM_{2.5}$ scavenging ratio (ΔC) was defined as the change of $PM_{2.5}$ mass concentration before and during the rain, as shown in Eq. (2) (Feng and Wang, 2012; Olszowski, 2016),

$$\Delta C = \frac{C_b - C_p}{C_b} \times 100\%, \quad (2)$$

where ΔC (%) is the the $PM_{2.5}$ scavenging ratio, C_b (μg m^{-3}) is $PM_{2.5}$ concentration before the rain, and C_p (μg m^{-3}) is hourly $PM_{2.5}$ concentration averaged on rainfall time of Δt. The positive ΔC indicates a decrease of $PM_{2.5}$ mass concentration during the rain, while the negative ΔC indicates an increase of $PM_{2.5}$ mass concentration during the rain. To consider the data representation, C_b is the average of $PM_{2.5}$ mass concentration within 3 h before the rain and C_p is the average of $PM_{2.5}$ mass concentration during the rain. In fact, there is little difference to calculate C_b when using the $PM_{2.5}$ data within 1 h before the rain.

In order to compare the variations of $PM_{2.5}$ concentration due to rain events with that of $PM_{2.5}$ concentration in rain-free periods, the $PM_{2.5}$ changed ratio (ΔC_{dry}) in rain-free periods before rain events was also calculated by using Eq. (3).

$$\Delta C_{dry} = \frac{C_d - C_b}{C_d} \times 100\%, \quad (3)$$

where C_d is the average of $PM_{2.5}$ mass concentration between 3 and 6 h in rain-free period before rain events. The positive ΔC_{dry} indicates a decrease tendency of $PM_{2.5}$ mass concentration before rain events, while the negative ΔC_{dry} indicates an increase tendency of $PM_{2.5}$ mass concentration before rain events.

Herein, we adopted the definition of a rain event proposed by Chen B. J. et al. (2013). A rain event is defined as the consecutive rainy hours. If the rain-free period is equal to or longer than 1 h, we regard it as two rain events. If the rain-free period is smaller than 1 h, we regard it as one rain event. According to glossary of meteorology (American Meteorological Society, 2019), we delineated the rain events using three categories: light rain (R: 0.1–2.5 mm h^{-1}), moderate rain (R: 2.6–7.6 mm h^{-1}), and heavy rain (R: > 7.6 mm h^{-1}). In order to analyze the dependence of aerosol scavenging on rain events, we chose the rain events in the pollution condition that the $PM_{2.5}$ concentration was higher than 35 μg m^{-3} before the rain. For the purpose of avoiding the influence from strong change of ambient environment, we defined that the rain events with wind speed larger than 4 m s^{-1} were rejected. When the temperature decreased more than 6°C in any adjacent hours from the rain-free period to the end of the rain, we regarded it as the change in $PM_{2.5}$ concentration due to the cold frontal system.

3. Results

3.1 *Statistical characteristics of rainfall*

Figure 2 shows the total duration and amount of all rain measurements and selected rain measurements from March 2014 to July 2016 in Beijing city. A total of 576 rain hours were observed with rainfall accumulation of 1530.5 mm, in which there were 372 hours of rain measurement with rainfall accumulation of 1117.4 mm in selected rain events. All rain events occurred from March to November. A total of 190 rain events were obtained and 117 rain events were selected in the observed period. Figure 3 shows the histogram of the rain duration and intensity of each rain event and selected rain event. As shown in Fig. 3a, each rain event lasted from tens of minutes to several hours. About 80% of the rain events had rain duration less than 5 h while those with duration more than 10 h were only 3%. As shown in Fig. 3b, the average rainfall rate of each rain event was in the range

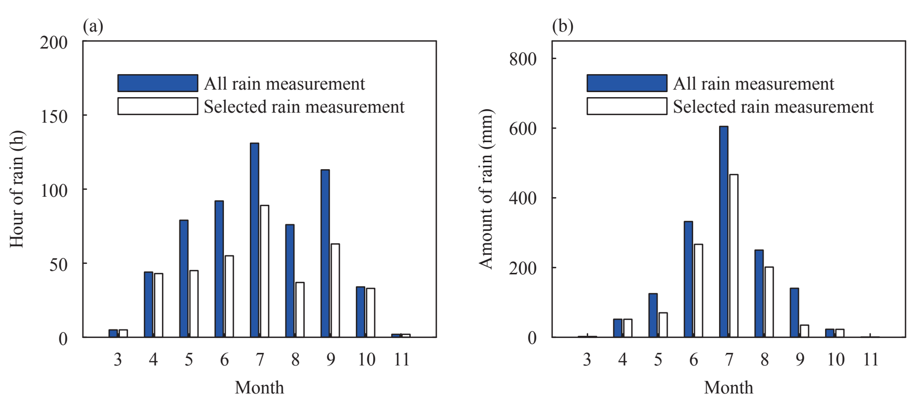

Fig. 2. (a) The total duration and (b) amount of all rain measurements and selected rain measurements during March 2014–July 2016.

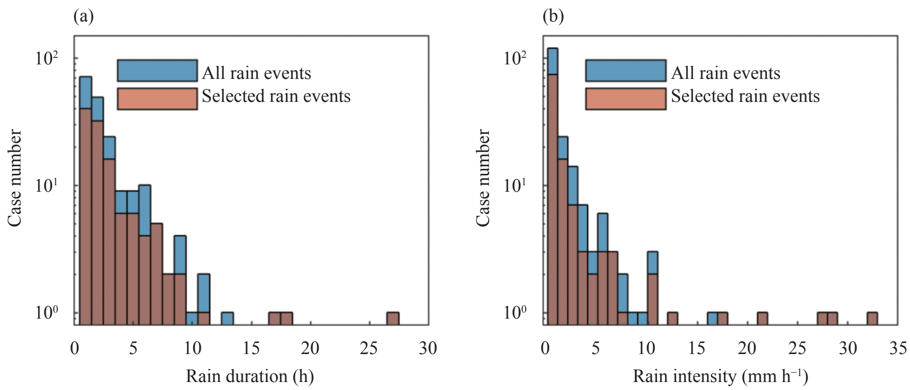

Fig. 3. Histograms of (a) the rain duration and (b) rain intensity of all rain events and selected rain events.

of 0–33 mm h^{-1} and most of them were 0–1 mm h^{-1}. There were 94 light events, 14 moderate events, and 9 heavy rain events in the selected events. The mean rainfall rate and standard deviation (SD) for these three groups were 0.6 ± 0.6 mm h^{-1}, 4.4 ± 1.4 mm h^{-1}, and 18.6 ± 9.3 mm h^{-1}, respectively. Light rain accounted for 80% of all selected rain cases.

3.2 *Effect of rain intensity on $PM_{2.5}$ scavenging ratio*

The relationship between $PM_{2.5}$ mass concentration and rainfall rate from March 2014 to July 2016 is shown in Fig. 4. During the observational period, the hourly rainfall rate was at the range of 0.1–80 mm h^{-1}. It is seen that the dominant rain hours were those with rainfall rates less than 10 mm h^{-1}. The rainfall rates greater than 40 mm h^{-1} were rarely observed. It can be seen that the $PM_{2.5}$ concentration substantially decreased with the increase of rainfall rate. The small rainfall rates ($R < 1$ mm h^{-1}) were closely associated with very high $PM_{2.5}$ concentrations.

Figure 5 shows the relationship between rain case numbers and $PM_{2.5}$ scavenging ratio (ΔC) for different rain intensities during 2014–16 in Beijing city. For light rain, the negative and positive $PM_{2.5}$ scavenging ratios almost accounted for 50%, respectively. The scavenging ratio for most light rain cases was primarily distributed from –20% to 20%. It is indicated that the $PM_{2.5}$ scavenging efficiency by light rain was relatively low. For moderate and heavy rain, although the case numbers were small, more positive and larger aerosol scavenging ratios

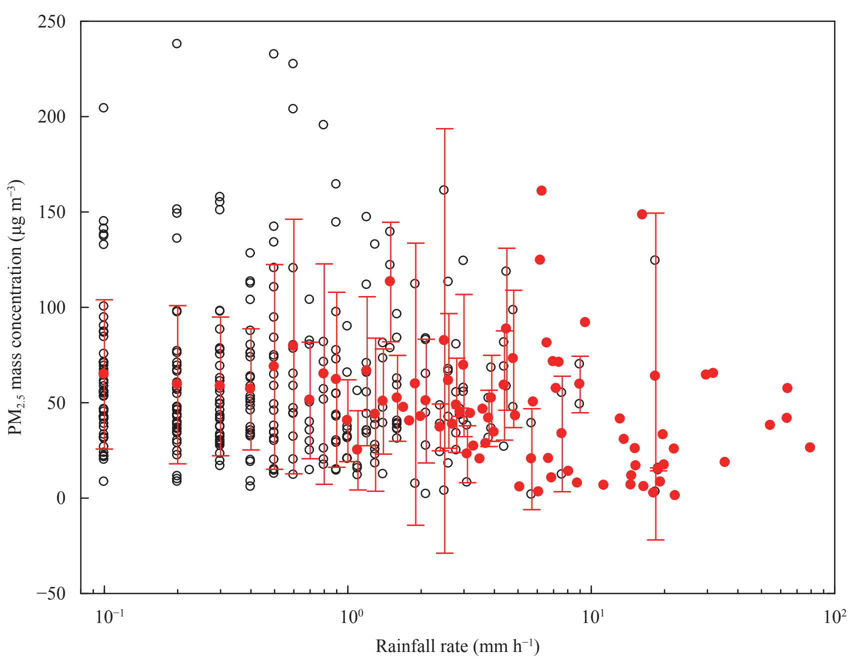

Fig. 4. Relationship between $PM_{2.5}$ mass concentration and rainfall rate during 2014–16 in Beijing city. The red dots and error bars represent the mean values and standard deviations.

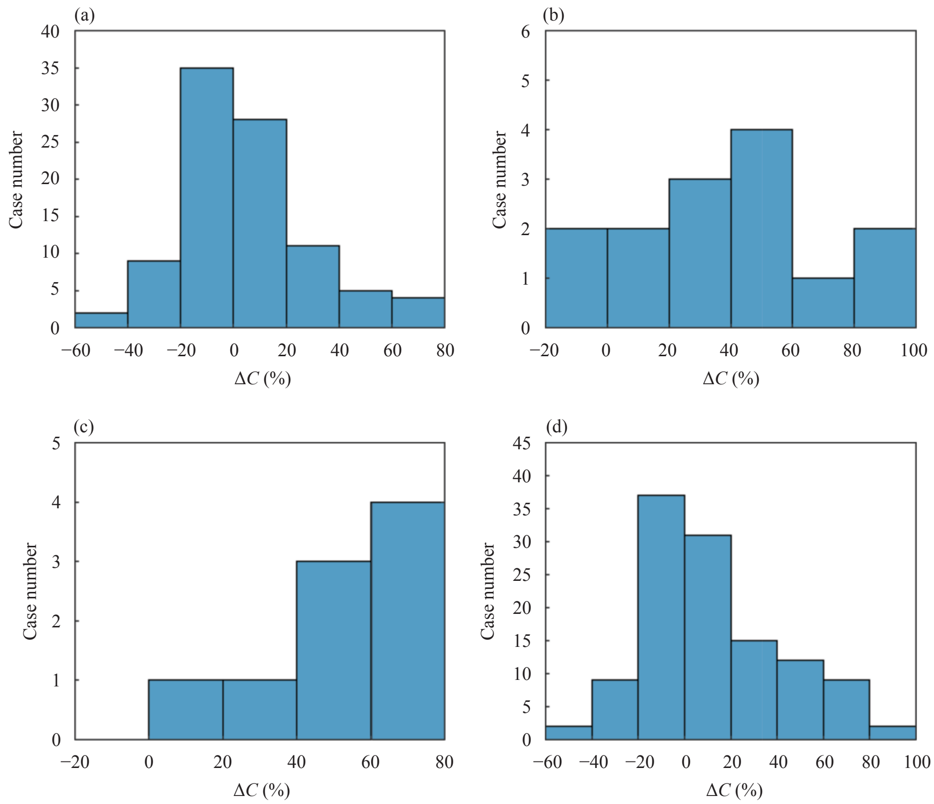

Fig. 5. Relationship between the rain case numbers and $PM_{2.5}$ scavenging ratio for different-intensity rains during 2014–16 in Beijing city: (a) light rain events, (b) moderate rain events, (c) heavy rain events, and (d) all rain events.

could be clearly seen in the figures. More than 40% of $PM_{2.5}$ could be removed for about 10%, 50%, and 78% cases in light, moderate, and heavy rain events, respectively. The higher rain intensity was more efficient in removal of $PM_{2.5}$.

Table 1 summarizes the rain case numbers and $PM_{2.5}$ scavenging ratio for different rain intensities during 2014–16 in Beijing city. It shows that the mean $PM_{2.5}$ scavenging ratios were 5.1% ± 25.7%, 38.5% ± 29.0%, and 50.6% ± 21.2% for light, moderate and heavy rain cases, respectively, indicating that the higher rain intensity was more efficient in removing the polluted aerosols in atmosphere. The mean positive and negative $PM_{2.5}$ scavenging ratios accounted for about 50% in light rain cases. However, the $PM_{2.5}$ scavenging ratio could be as high as 79%, which indicates that in some situations, the light rain event could also have high aerosol scavenging efficiency. The reason will be further explained in the later analyses. For moderate and heavy rain cases, the case numbers of $PM_{2.5}$ increases were relatively small. The maximum $PM_{2.5}$ scavenging ratio could be over 83%.

In order to understand the uncertainty of the above analysis method, the $PM_{2.5}$ changed ratio (ΔC_{dry}) in rain-free periods before the rain events was also calculated (Table 2 and Fig. 6). The positive ΔC_{dry} indicates a decreasing tendency of $PM_{2.5}$ mass concentration before the

Table 1. Summary of rain case numbers and $PM_{2.5}$ scavenging ratios for different rain intensities

Rain intensity	Rain event number	$PM_{2.5}$ scavenging ratio ΔC (%)		
		Mean ± SD	Maximum	Minimum
Light rain	94 (total)	5.1 ± 25.7	78.8	–50.9
	48 (decreased)	23.4 ± 21.8	78.8	0.4
	46 (increased)	–14.0 ± 12.0	–0.2	–50.9
Moderate rain	14 (total)	38.5 ± 29.0	83.9	–10.4
	12 (decreased)	46.0 ± 23.6	83.9	5.6
	2 (increased)	–6.7 ± 5.3	–2.9	–10.4
Heavy rain	9 (total)	50.6 ± 21.2	70.9	10.5
	9 (decreased)	50.6 ± 21.2	70.9	10.5

Note: The “decreased” (“increased”) in brackets indicates the number of rain cases with decreased (increased) $PM_{2.5}$ concentration during the rain. The “total” in brackets indicates the sum of rain case numbers for decreased and increased $PM_{2.5}$ concentrations during the rain.

rain, while the negative ΔC_{dry} indicates an increasing tendency of $PM_{2.5}$ mass concentration before the rain. The average ΔC_{dry} and ΔC were –8.4% ± 32.3% and 12.6% ± 30.0%, respectively, indicating that there was an increasing tendency of $PM_{2.5}$ concentration during rain-free periods before rain events and $PM_{2.5}$ concentration also decreased after rain events. As seen in Table 2, about 32.5% of events presented an increasing tendency of $PM_{2.5}$ concentration during rain-free period and $PM_{2.5}$ concentration decreased after rain events. In this situation, the rain played an evident role in $PM_{2.5}$ scavenging. About 30.0% of events presented a decreasing tendency of $PM_{2.5}$ concentration during rain-free periods and $PM_{2.5}$ concentration also decreased after rain events. In this situation, although ΔC_{dry} and ΔC were both positive, the maximum value of ΔC_{dry} (44%) was much smaller than that of ΔC (84%). This indicates that the contribution of rain on $PM_{2.5}$ scavenging is more important.

From analyses above, we can see that for moderate and heavy the rain events, $PM_{2.5}$ can be obviously scavenged by rain events. However, for light the rain events, the results had large difference. In light rain events, about 50% cases could decrease $PM_{2.5}$ apparently, and other 50% cases had no obvious effect. Even for some cases the $PM_{2.5}$ concentration was increased. In order to understand this phenomenon, we further investigate the impacts of the raindrop size distribution, the rain duration, and wind speed on aerosol scavenging ratio.

Table 2. Statistical percentage of the $PM_{2.5}$ changing ratio (ΔC_{dry}) in rain-free periods before rain events and $PM_{2.5}$ scavenging ratio (ΔC) by rain

		Percentage of the rain events
$\Delta C > 0$	$\Delta C_{dry} < 0$	32.5%
	$\Delta C_{dry} > 0$	30.0%
$\Delta C < 0$	$\Delta C_{dry} < 0$	32.5%
	$\Delta C_{dry} > 0$	17.5%

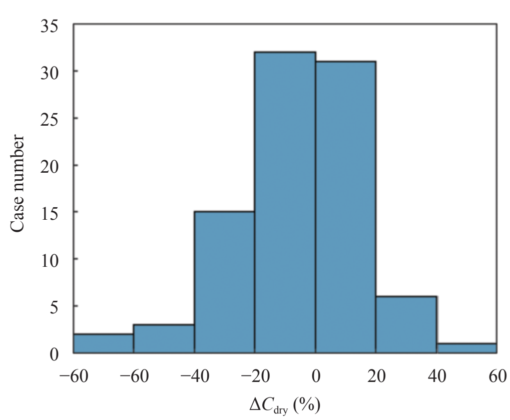

Fig. 6. Relationship between the case numbers and the $PM_{2.5}$ changed ratio (ΔC_{dry}) in rain-free periods before rain events during 2014–16 in Beijing city.

3.3 *Effect of raindrop size distribution on $PM_{2.5}$ scavenging ratio*

Figure 7 shows the mean raindrop size distributions of light, moderate, and heavy rain events. As the rain intensity increased, the distributions of raindrops shifted to larger drops, and the total raindrop number concentration also increased. Thus, the collection efficiency was increasing due to the increased number of raindrops for higher rain intensity. The liquid water contents of the light, moderate, and heavy rain were 0.063 ± 0.061, 0.305 ± 0.128, and 1.302 ± 0.671 g m^{-3}, respectively.

Mean size distributions of raindrops for light rain cases with $PM_{2.5}$ concentration increased and decreased during the rain are shown in Fig. 8. It is seen that the size distributions of raindrops for the two situations had no apparent differences, indicating that the size distribution of raindrops was not an important factor to cause the different below-cloud scavenging ratios in the light rain events. This can be easily understood since the light rain events usually have relatively stable raining process, and

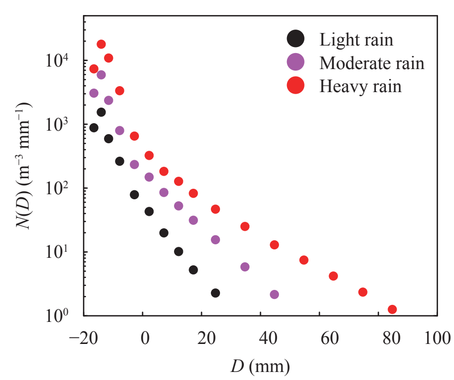

Fig. 7. The mean raindrop size distributions of light, moderate, and heavy rain events.

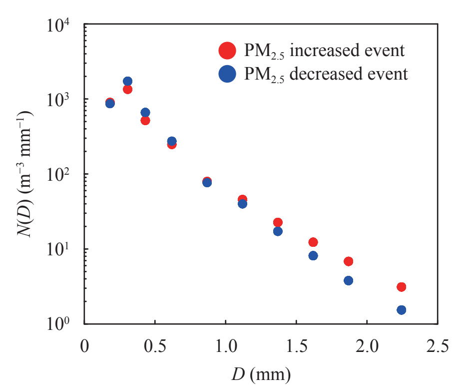

Fig. 8. Mean raindrop size distributions for both cases with $PM_{2.5}$ concentration decreased and increased in light rain events.

the variation of raindrop size distribution is also small during rain period. Andronache (2003) also found that the scavenging coefficient had a weak dependence on the particular raindrop size distributions for the same rain intensity.

3.4 *Effect of rain duration on $PM_{2.5}$ scavenging ratio*

We investigated all rain events in this study, which had different durations (Fig. 3a). To investigate the impact of rain duration on $PM_{2.5}$ scavenging ratio, Figs. 9a–d show the variations of $PM_{2.5}$ scavenging ratio with rain duration for different rain intensities in case of $PM_{2.5}$ concentration decreased during the rain. It shows that the rain duration had more important impact on $PM_{2.5}$ scavenging ratio in light rain event than that in moderate and heavy rain. For light rain event, the scavenging ratio increased from less than 20% to about 40% after 3 h, and then it almost kept constant. For moderate rain event, the scavenging ratio kept about 50% from 1 to 6 h. After 6 h, the scavenging ratio decreased to about 30%. For heavy rain event, the scavenging ratio increased rapidly to 60% at 3 h. On average for all rain events, the scavenging ratio could increase from less 20% to about 40% within 6 h, and after 6 h, it slightly decreased. It means that the rain duration was an important factor to the below-cloud $PM_{2.5}$ scavenging ratio, in particular, for the light rain event that lasted for more than 3 h. The one-dimensional model simulations done by Zhang et al. (2004) also reported similar results. The values of scavenging ratio were more decentralized in the light rain than those in moderate and heavy rains, which was also observed in the research about PM_{10} scavenging by rain in Poland (Olszowski, 2016).

In contrast, Figs. 9e–g illustrate the variations of $PM_{2.5}$ scavenging ratio with rain duration for different rain intensities in case of increased $PM_{2.5}$ concentration during rain. It was found that the rain duration had very small influence on the increase of $PM_{2.5}$ concentration during the rain events. The raindrop evaporation may have contributed to the slightly increase of $PM_{2.5}$ concentration near the ground surface.

3.5 *Effect of wind speed on $PM_{2.5}$ scavenging ratio*

The wind speed is an important meteorological variable affecting aerosol concentrations (Barmpadimos et al., 2011). In particular, it can affect the measured aerosol particle concentrations at a site. The average wind speeds of the rain events were in the range of 0–3.4 m s^{-1}. About 84% rain events had the wind speed less than 2.0 m s^{-1}. Figure 10 shows that relationship between $PM_{2.5}$ scavenging ratio and the difference of wind speed for different rain intensities. Here, the wind speed difference was the wind speed during the rain minus that before the rain. The negative value indicates a decrease of wind speed during the rain, while the positive value indicates an increase of wind speed during the rain. The

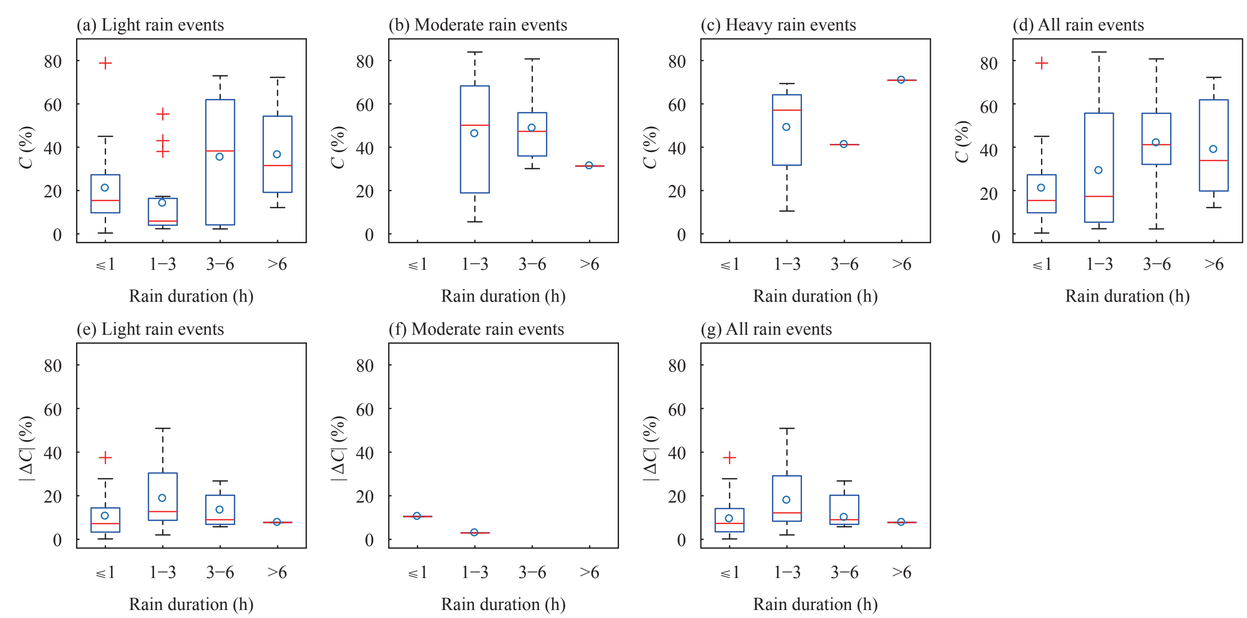

Fig. 9. Variations of the $PM_{2.5}$ scavenging ratio with rain duration for different-intensity rains. (a–d) The rain events with $PM_{2.5}$ concentration decreased after the rain. (e–g) The rain events with $PM_{2.5}$ concentration increased after the rain. The blue circles indicate the average value. The red lines indicate the median value. The box boundaries represent the first and third quartiles. The black lines above and under the box indicate the maximum and minimum values. $|\Delta C|$ is the absolute value of the $PM_{2.5}$ scavenging ratio.

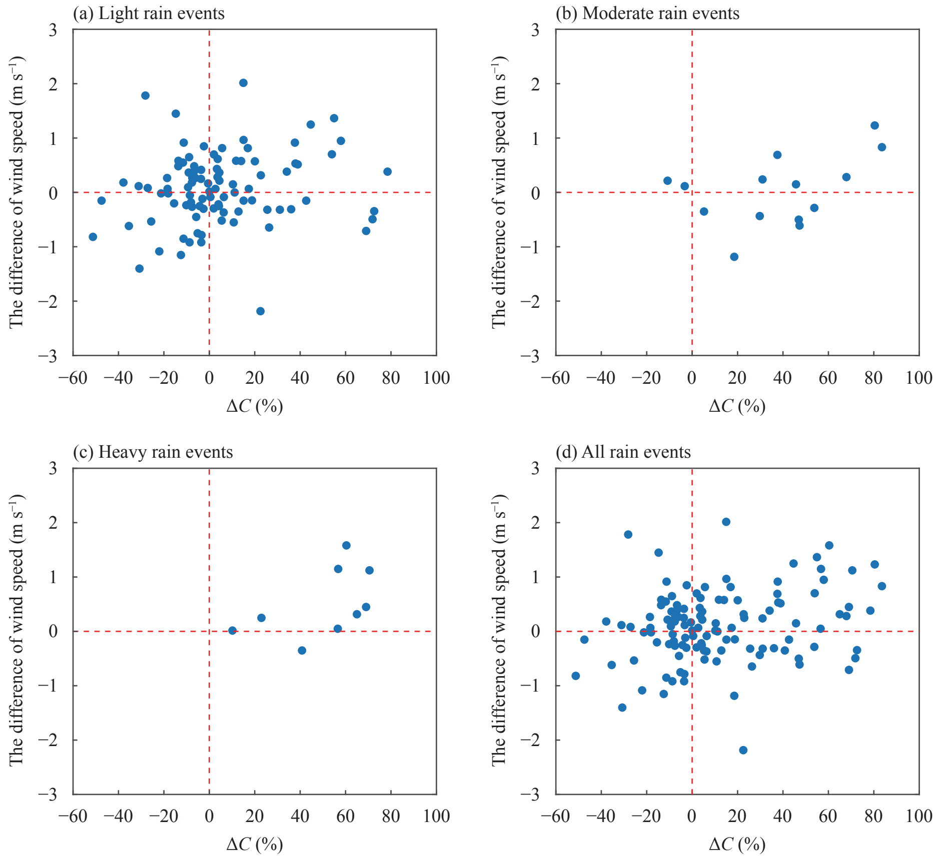

Fig. 10. Relationship between $PM_{2.5}$ scavenging ratio and the difference of wind speed (the wind speed during the rain minus that before the rain) for different-intensity rains during 2014–16 in Beijing city: (a) light rain events, (b) moderate rain events, (c) heavy rain events, and (d) all rain events.

maximum and minimum of the wind speed difference were 2.0 and –2.2 m s^{-1}, respectively. As shown in Fig. 10a, light rain events with positive $PM_{2.5}$ scavenging ratio and positive wind speed difference account for 26%, in which the higher $PM_{2.5}$ scavenging ratio generally corresponded well with the higher wind speed difference. It is indicated that the increased wind speed during rain had an important contribution on the enhancement of $PM_{2.5}$ scavenging process. Moreover, light rain events with negative $PM_{2.5}$ scavenging ratio and negative wind speed difference account for 24%. In this situation, the higher absolute value of $PM_{2.5}$ scavenging ratio generally corresponded well with the higher absolute value of wind speed difference. This indicates that the increased $PM_{2.5}$ concentration during light rain is due to the decreased wind speed.

Figures 10b, c indicate that wind speed generally had positive influence on $PM_{2.5}$ scavenging ratio in moderate and heavy rain events, but this positive influence was not as obvious as that in light rain events. This is because that the wind speed difference between before and during rain process for moderate and heavy rain events was not so large as that for light rain events. Therefore, the wind speed was another important factor to influence the $PM_{2.5}$ scavenging process in light rain events except for rain duration.

The averaged wind speed difference for rain events with $PM_{2.5}$ decreased and increased for different rain intensities is shown in Table 3. In the light rain, the averaged wind speed difference in the rain events with $PM_{2.5}$ concentration decreased after the rain was 0.15 ± 0.7 m s^{-1} and that in the rain events with $PM_{2.5}$ concentration increased after the rain was –0.04 ± 0.7 m s^{-1}. This further indicates that the increased $PM_{2.5}$ concentration during light rain should be caused by the decreased wind speed. The wind could play a critical role in sharply decreasing aerosol concentration, especially in the very pollutant condition.

4. Conclusions

The haze events characterized by high $PM_{2.5}$ concentration often occurred in the highly industrial and populated regions in the world. Below-cloud aerosol scaven-

Table 3. The averaged wind speed difference of the rain events with $PM_{2.5}$ decreased and increased for different-intensity rains

$PM_{2.5}$ concentration	Wind speed for light rain (m s^{-1})	Wind speed for moderate rain (m s^{-1})	Wind speed for heavy rain (m s^{-1})	Wind speed for all rains (m s^{-1})
Decreased	0.15 ± 0.7	–0.01 ± 0.7	0.49 ± 0.6	0.17 ± 0.7
Increased	–0.04 ± 0.7	0.15 ± 0.1	–	–0.03 ± 0.8
Total	0.06 ± 0.7	0.01 ± 0.6	0.49 ± 0.6	0.09 ± 0.7

ging process by precipitation is an important mechanism for cleaning the polluted aerosols in the atmosphere in these regions. The issue is widely concerned since it is closely related to air quality and human health. Most previous studies on this issue were conducted based on limited field measurement data, laboratory experiments, and theoretical methods, and the relevant results were not fully consistent and the mechanism involved in this process have not been well known. In this study, we conducted quantitative investigation of $PM_{2.5}$ scavenging ratio by rains in different intensities, based on long-term multi-site measurements from March 2014 to July 2016 in Beijing city, northern China.

To quantify the below-cloud aerosol scavenging process by rain, $PM_{2.5}$ scavenging ratio was defined and calculated as the change in $PM_{2.5}$ mass concentration before and during the rain for different rain intensities. We found that the mean $PM_{2.5}$ scavenging ratios were 5.1% ± 25.7%, 38.5% ± 29.0%, and 50.6% ± 21.2% for light, moderate, and heavy rain events, respectively, indicating that the rain event of higher rain intensity was more efficient in removing the polluted aerosols in the atmosphere. The $PM_{2.5}$ concentration could be apparently decreased for more than 85% rain cases at moderate and heavy rain intensities. The maximum aerosol scavenging ratio by moderate and large rain intensities could reach as high as over 83%. This is because a higher rainfall rate denotes a larger number concentration of raindrops at the levels below cloud and thus a greater chance of collision with aerosol particles.

However, the $PM_{2.5}$ concentration was not overall obviously decreased since nearly 50% of the cases were light rains; it was even increased in some cases. To understand this phenomenon, we further investigated the influence of raindrop size distribution, rain duration, and wind speed on the $PM_{2.5}$ scavenging ratio, and found that raindrop size distribution had less influence on the $PM_{2.5}$ scavenging ratio for light rain intensity. But the rain duration and wind speed had stronger influence on the $PM_{2.5}$ scavenging ratio for light rain intensity and less influence on that for moderate and heavy rain intensities. The stronger wind speed and longer rain duration were responsible for the higher $PM_{2.5}$ scavenging ratio in some light rain intensity cases. The increased aerosol concentrations that occurred in some light rain cases were due to the decreased wind speed.

Since most of the scavenging ratios were within 20%, the atmospheric $PM_{2.5}$ scavenging by rain events in Beijing was limited. This is because the accumulation mode of aerosol particles accounts for important contribution to the $PM_{2.5}$ mass concentration. Moreover, the raindrop collection efficiency for accumulation mode particles was minimum in previous experimental and theoretical studies. Due to the limitation of observational data, the effect of aerosol particle size on scavenging process by rain events is not analyzed in this study. Thus, we cannot compare the roles of different-intensity rains on aerosol size-dependent scavenging. Moreover, we only analyze the impacts of rain duration and wind speed on $PM_{2.5}$ concentration for light rain events. The other factors should be examined in future investigation.

The findings of this study are important to understand the mechanism of the below-cloud aerosol scavenging during different-intensity precipitation events. In particular, variations in aerosol concentration for altered aerosol particle sizes during different-intensity rain cases are worth of further investigation. An additional possible application of this study is to evaluate and assess the efficiency of artificial removal of aerosol particles in a local polluted area.

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