

RESEARCH ARTICLE

# Comparison of dry and wet deposition of particulate matter in near-surface waters during summer

Yanan Wu<sup>1</sup>, Jiakai Liu<sup>1</sup>, Jiexiu Zhai<sup>1</sup>, Ling Cong<sup>1</sup>, Yu Wang<sup>1</sup>, Wenmei Ma<sup>1</sup>, Zhenming Zhang<sup>1\*</sup>, Chunyi Li<sup>2\*</sup>

**1** College of Nature Conservation, Beijing Forestry University, Beijing, China, **2** Institute of Wetland Research, Chinese Academy of Forestry, Beijing, China

\* [zhenmingzhang@bjfu.edu.cn](mailto:zhenmingzhang@bjfu.edu.cn) (ZM Zhang); [chunyili@126.com](mailto:chunyili@126.com) (CY Li)



**OPEN ACCESS**

**Citation:** Wu Y, Liu J, Zhai J, Cong L, Wang Y, Ma W, et al. (2018) Comparison of dry and wet deposition of particulate matter in near-surface waters during summer. PLoS ONE 13(6): e0199241. <https://doi.org/10.1371/journal.pone.0199241>

**Editor:** Fei Li, Zhongnan University of Economics and Law, CHINA

**Received:** November 6, 2017

**Accepted:** June 4, 2018

**Published:** June 21, 2018

**Copyright:** © 2018 Wu et al. This is an open access article distributed under the terms of the [Creative Commons Attribution License](https://creativecommons.org/licenses/by/4.0/), which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are credited.

**Data Availability Statement:** All relevant data are within the paper.

**Funding:** This research was supported by the Fundamental Research Funds for the Central Universities (2016JX05) (ZZ), Forestry Public Welfare Projects Special Fund of China (201304301), and Beijing Municipal Science and Technology Project (Z141100006014031).

**Competing interests:** The authors have declared that no competing interests exist.

## Abstract

Atmospheric particulate matter (PM) deposition which involves both dry and wet processes is an important means of controlling air pollution. To investigate the characteristics of dry and wet deposition in wetlands, PM concentrations and meteorological conditions were monitored during summer at heights of 1.5 m, 6 m and 10 m above ground level at Cuihu Wetland (Beijing, China) in order to assess the efficiency of PM<sub>2.5</sub> (particles with an aerodynamic size of <2.5 μm) and PM<sub>10</sub> (particles with an aerodynamic size of <10 μm) removal. The results showed: Daily concentrations of PM, dry deposition velocities and fluxes changed with the same variation trend. The daily average deposition velocity for PM<sub>10</sub> ( $3.19 \pm 1.18 \text{ cm}\cdot\text{s}^{-1}$ ) was almost 10 times that of PM<sub>2.5</sub> ( $0.32 \pm 0.33 \text{ cm}\cdot\text{s}^{-1}$ ). For PM<sub>2.5</sub>, the following dry deposition fluxes were recorded: 10 m ( $0.170 \pm 0.463 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) > 6 m ( $0.007 \pm 0.003 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) > 1.5 m ( $0.005 \pm 0.002 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ). And the following deposition fluxes for PM<sub>10</sub> were recorded: 10 m ( $2.163 \pm 2.941 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) > 1.5 m ( $1.565 \pm 0.872 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) > 6 m ( $0.987 \pm 0.595 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ). In the case of wet deposition, the relative deposition fluxes for PM<sub>2.5</sub> and PM<sub>10</sub> were 1.5 m > 10 m > 6 m, i.e. there was very little difference between the fluxes for PM<sub>2.5</sub> ( $0.688 \pm 0.069 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) and for PM<sub>10</sub> ( $0.904 \pm 0.103 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ). It was also noted that rainfall intensity and PM diameter influenced wet deposition efficiency. Dry deposition (63%) was more tilted towards removing PM<sub>10</sub> than was the case for wet deposition (37%). In terms of PM<sub>2.5</sub> removal, wet deposition (92%) was found to be more efficient.

## Introduction

The frequent occurrence of particulate matter (PM) pollution has led to many problems [1,2]. The emission of PM is one of the most important factors affecting climate [2] and health [3,4]. The presence of PM<sub>2.5</sub> is thought to have caused premature mortality in 1.27 million individuals in China [5,6]. Likewise, PM<sub>10</sub> may increase the risk of premature death [7] due to cardiovascular [8] and respiratory diseases [9].

Atmospheric deposition is an important means of controlling air pollution [10]. Atmospheric PM deposition involves both dry and wet processes. Dry deposition refers to the deposition of particles or gases from the atmosphere through the direct delivery of mass to the surface (i.e. via non-precipitation) [11]. On the other hand, wet processes are often referred to as rain or snow scavenging [12], with rain scavenging PM being generally classified as ‘rainout particles’ (serving as cloud-condensation nuclei or undergoing capture by cloud water) and as ‘washout’ (i.e. removal of below-cloud particles by raindrops as they fall) [13].

Studies have confirmed that dry deposition has the capacity to remove PM [14–16] and that the process of dry deposition is influenced by spatial fluctuations, surface-type differences, temporal changes, diurnal variations and meteorological conditions [14]. Wet deposition is also an important mechanism for reducing air pollution by the removal of PM [15]. A study showed that wet deposition accounted for 54–71% of PM<sub>1–20</sub> (particles with 1–20 μm diameter) deposition and it accounted for 76–86% of PM<sub>0.5–20</sub> (particles with 0.5–20 μm diameter) deposition [12]. According to a case study carried out in Guangzhou (China), the total annual flux of wet and dry depositions, representing the combined results for PM deposition in the urban area, was 34 g·m<sup>-2</sup>·yr<sup>-1</sup>, with 50% being attributed by wet deposition [15]. Therefore, dry and wet depositions can be regarded as important pathways for the elimination of PM from ambient air.

Wetlands are important ecological systems that perform vital ecological functions. A number of studies have demonstrated that wetlands play a role in reducing PM [17,18]. Considerable research attention has been focused on dry deposition as well as comparisons between various surface types [17,18]. Space–time variation has also been the focus of some studies. For example, PM<sub>2.5</sub> and PM<sub>10</sub> dry deposition fluxes at 10 m were higher than those measured at 6 m [16]. And deposition fluxes measured during the dry period (1.03 μg·m<sup>-2</sup>·s<sup>-1</sup>) were higher than those during the wet period (0.003 μg·m<sup>-2</sup>·s<sup>-1</sup>) and during normal humidity conditions (0.02 μg·m<sup>-2</sup>·s<sup>-1</sup>) [16]. Very little work has been carried out on the the ecological functions associated with dry and wet deposition in wetlands.

This study investigated the temporal and spatial variations in dry and wet deposition velocity in wetlands and compared the deposition fluxes of dry and wet deposition in order to assess the efficiency of PM<sub>2.5</sub> and PM<sub>10</sub> removal.

## Materials and methods

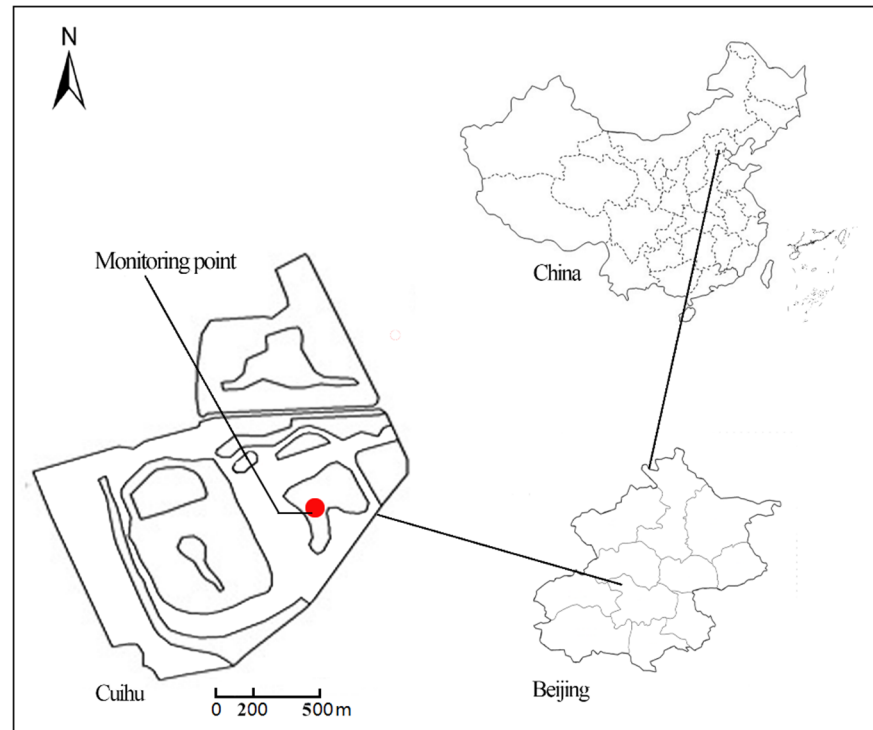
### Experimental sites

Cuihu National Urban Wetland Park (1.57 km<sup>2</sup>; 1.9 km × 1.2 km) is situated north of the Shangzhuang Reservoir, Haidian District, Beijing (Fig 1), which is in a warm temperate semi-humid monsoon climate zone. The annual average temperature is 12.38°C and the annual precipitation is 500–700 mm, 60% of which is concentrated in the wet summer period (July and August) [16]. The experiment was carried out on the “crane island” (116° 19′E, 40° 10′N) in Cuihu National Urban Wetland Park as shown in Fig 1.

### Experimental design

**Ethics statement.** This study has been licensed by Cuihu National Urban Wetland Park. And this study did not involve endangered or protected species.

**Dry deposition.** Particle concentration data parameters (TSP, PM<sub>10</sub> and PM<sub>2.5</sub>) were measured using Dustmate (Turnkey Co. Ltd, UK). The using of Dustmate for collecting the PM concentration was given in previous studies [16, 17, 18] and all above these researches have successfully used this instrument. Meteorological data (wind speed, temperature and relative humidity) were collected by a weather station (Nielsen-Kellerman Co. Ltd, USA). Both the



**Fig 1. Location of study site.** Reprinted from Ref. [16] under a CC BY license, with permission from Lijuan Zhu, original copyright 2016.

<https://doi.org/10.1371/journal.pone.0199241.g001>

Dustmate and the weather station were mounted at three height levels, which were 1.5 m (the average plant height in the wetland), 6 m (dry deposition layer) and 10 m (stable layer) above ground level. The sampling time was from 7:00 am to 7:00 pm during summer (June–August) in 2016. The reason for the summer sampling was the rainfall concentrated in summer and we could contrast dry deposition and wet deposition on this basis.

**Wet deposition.** The sampling time was the summer (June–August) of 2016. Rainfall events of greater than 8 h of duration were chosen to represent wet deposition in order to studied the process of wet deposition under longer rainfall and compared it with the dry deposition process. Rain collectors which were located at 1.5 m, 6 m and 10 m collected rainwater every 0.5 h to provide information on volume and intensity. A Dustmate system was used to measure particle concentration during rainfall events.

## Data analysis

**Dry deposition.** Dry deposition flux can be calculated as follows [19,20]:

$$F = v_d \cdot \Delta c \quad (1)$$

Where F is deposition flux;  $\Delta c$  is concentration difference between constant flux layer and deposition layer and  $V_d$  is deposition velocity. This deposition velocity can be defined as follows:

$$\frac{1}{v_d} = \frac{1}{V_C} + \frac{1}{V_D} - \frac{V_g}{V_C \cdot V_D} \quad (2)$$

Where  $V_g$  is the gravitational settling speed (based on dry particle diameters);  $V_C$  is total

transfer velocity in the constant flux layer and  $V_D$  is the total transfer velocity in the dry deposition layer, calculated as follows:

$$V_g = \rho_p \cdot C_c \cdot d_p^2 \cdot g / 18 \cdot \mu_a \tag{3}$$

$$V_c = V'_c + V_g \tag{4}$$

$$V_D = V'_D + V_g \tag{5}$$

Where  $C_c$  is the Cunningham correlation factor;  $\rho_p$  is density of particles (equivalent to particle concentration);  $d_p$  is particle diameter;  $\mu_a$  is air dynamic viscosity;  $V'_c$  is transfer velocity (without gravity) in the constant flux layer and  $V'_D$  is transfer velocity (without gravity) in the dry deposition layer. These can be calculated as follows:

$$C_c = 1 + \frac{2\lambda}{d_p} \cdot (1.257 + 0.4e^{-0.55 \cdot d_p / \lambda}) \tag{6}$$

$$V'_c = \frac{1}{1 - k} \cdot C_d \cdot u(z) \tag{7}$$

$$V'_D = -\alpha \cdot m + k^{-1} \cdot C_d \cdot u(z) \cdot Sc^{-1/2} + 10^{-3} \cdot St \tag{8}$$

Where  $\lambda$  is mean free path of air (65 nm);  $\alpha$  is a constant [ $10^3 \text{ cm} \cdot \text{s}^{-1} / (1 \text{ g} \cdot \text{cm}^{-2} \cdot \text{s}^{-1})$ ];  $C_d$  is the drag coefficient, calculated as follows:  $C_d = [(1.3 \pm 0.3) \times 10^{-3}]$ ;  $Sc$  is Schmidt number and  $St$  is Stokes number, calculated as follows:

$$S_c = \frac{V_a}{D_B} = \frac{\mu_a}{\rho_p} \tag{9}$$

$$S_t = \tau_p \cdot u(z) / d_n \tag{10}$$

Where  $V_a$  is the air kinematic viscosity;  $D_B$  is the Brown diffusion coefficient;  $u(z)$  is the average wind velocity;  $d_n$  is the dimension of the vegetation element for wetlands ( $d_n = 1$ ) [20,21];  $\tau_p$  is the particle relaxation time, which can be presented as follows:

$$\tau_p = \rho_p \cdot C_c \cdot d_p^2 / 18 \cdot \mu_a \tag{11}$$

**Wet deposition.** Wet deposition flux is calculated as follows [20,21]:

$$P_{wet_i} = Cr_i \times \frac{\text{Rain}}{t} \tag{12}$$

Where  $P_{wet_i}$  is the wet deposition flux,  $Cr_i$  is the concentration of particles, Rain is rainfall and  $t$  is the time of collection.

## Results

### Temporal and spatial variation in dry deposition

Fig 2 shows two curves relating to daily variation in PM2.5 and PM10 deposition velocity, both of which indicate similar changes. The highest value for deposition velocity was recorded at 8:00. PM2.5 deposition velocity decreased during the next three hrs and then changed in a

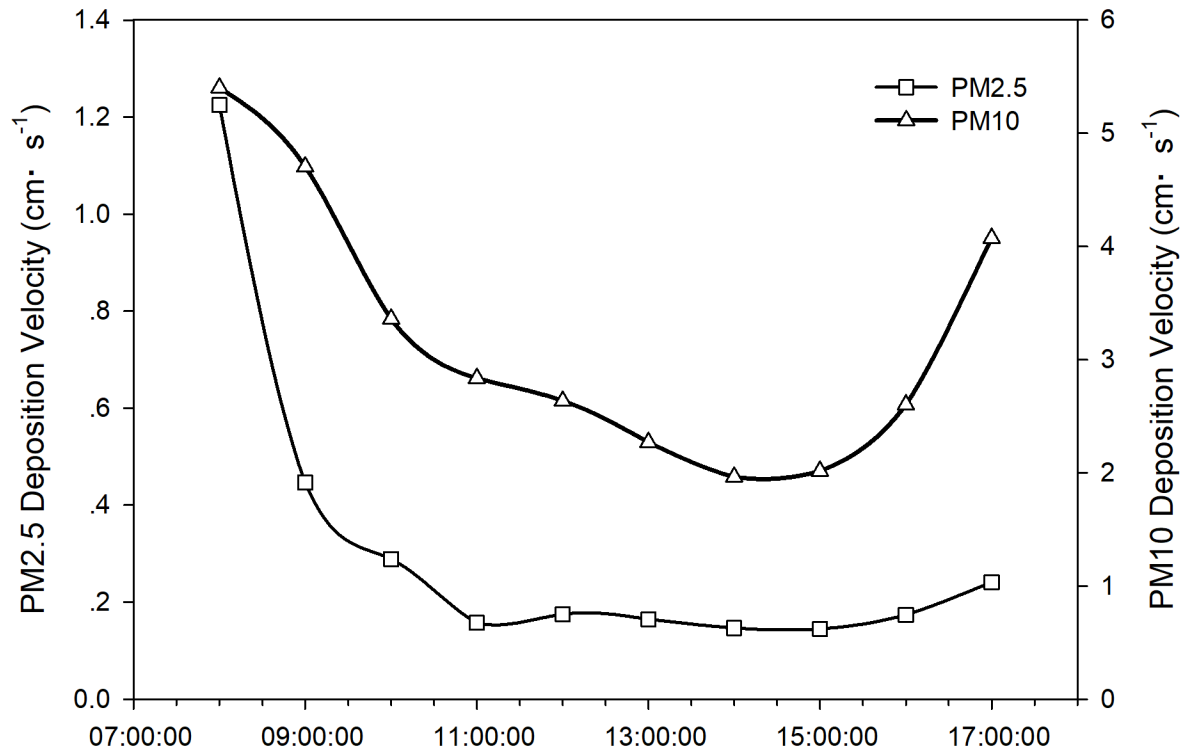


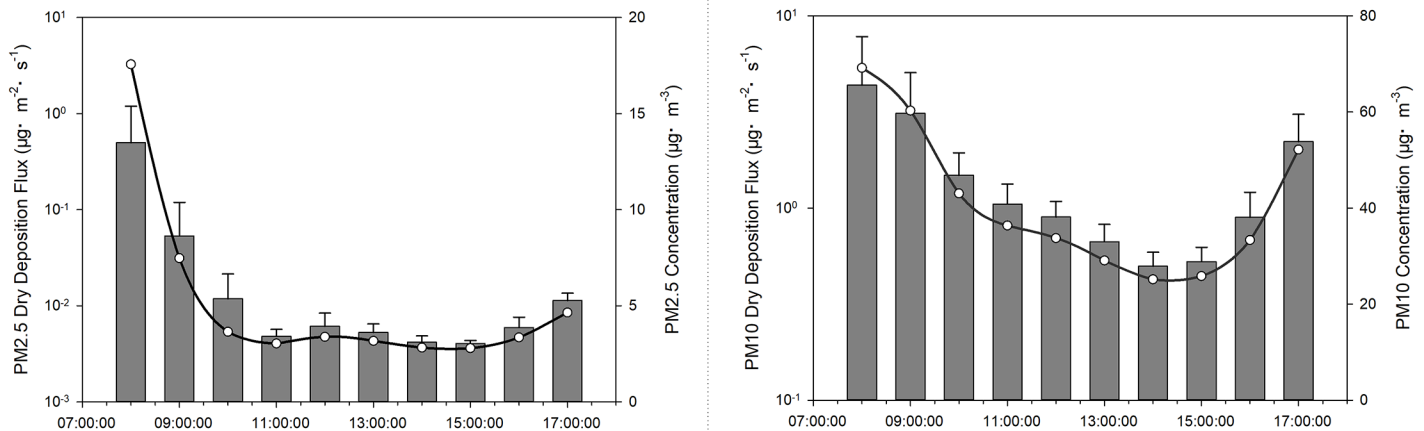
Fig 2. Daily variation in PM deposition velocity.

<https://doi.org/10.1371/journal.pone.0199241.g002>

stable manner, while PM10 deposition velocity continued to decrease until 15:00, after which there was a slight increase in PM2.5 deposition velocity and a greater increase in PM10 deposition velocity. Although the trends associated with these variations were similar, greater differences were noted between the deposition velocities of PM2.5 and PM10. The deposition velocities of PM10 were greater than those of PM2.5. The daily average deposition velocity of PM10 was  $3.19 \pm 1.18 \text{ cm}\cdot\text{s}^{-1}$ , almost 10 times that of PM2.5 ( $0.32 \pm 0.33 \text{ cm}\cdot\text{s}^{-1}$ ). Minimum PM2.5 and PM10 deposition velocities were  $0.14 \text{ cm}\cdot\text{s}^{-1}$  and  $1.96 \text{ cm}\cdot\text{s}^{-1}$ , respectively, both of which were recorded between 14:00 and 15:00.

The relationship between PM concentration and dry deposition flux is shown in Fig 3. These results suggest that variation trends in PM concentrations followed the same pattern as that associated with dry deposition flux. When PM concentrations were high, dry deposition flux increased; after a period of time, the concentration of PM in the air decreased, resulting in a decrease in dry deposition flux. Because of the low dry deposition flux, PM concentration again increased and accordingly the dry deposition flux increased. PM concentration and dry deposition flux thus had a mutual influence on each other. The highest PM2.5 and PM10 dry deposition flux values were recorded at 8:00, as were the concentrations of PM2.5 and PM10. Average dry deposition fluxes of PM2.5 and PM10 were  $0.06 \pm 0.15 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$  and  $1.57 \pm 1.28 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ , respectively. These results clearly indicate that the dry deposition flux of PM10 was much higher than that of PM2.5.

Information on the spatial variation in PM2.5 and PM10 deposition flux is summarised in Fig 4. PM2.5 deposition flux presented a downward and an upward trend, and then declined and rose again when it reached 10 m and 1.5 m, respectively. These changing tendencies resulted in an initial peak of 6 m, which later declined. The highest value occurred at 17:00 at both 6 m and 1.5 m. When it reached the height of 10 m, the highest flux occurred at 8:00. The maximum



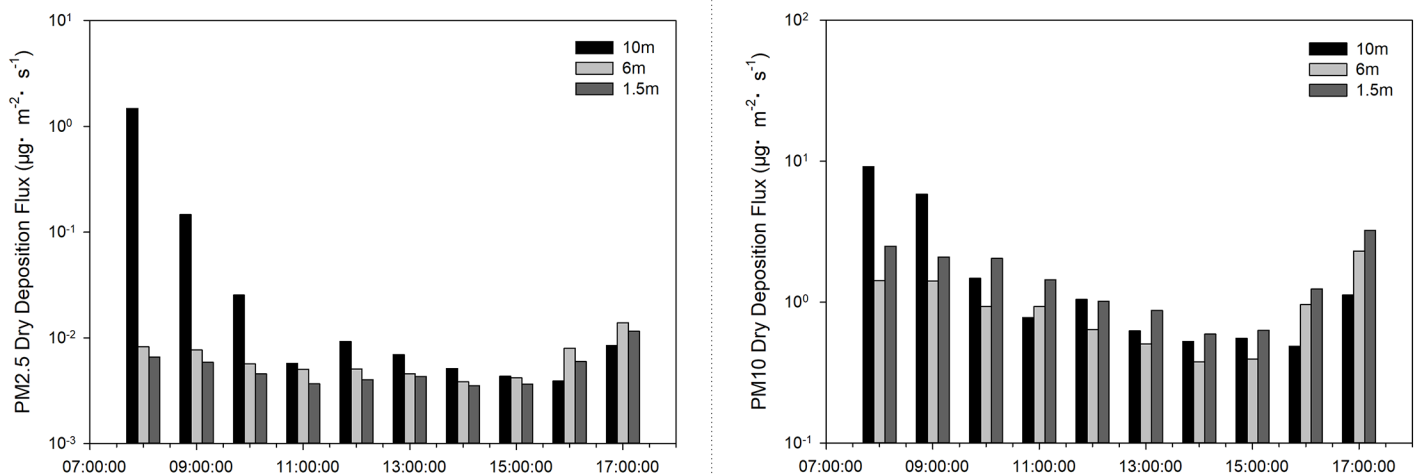
**Fig 3. PM concentration and dry deposition flux.**

<https://doi.org/10.1371/journal.pone.0199241.g003>

deposition flux ( $1.48 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) for PM2.5 occurred at 10 m, which was considerably greater than that recorded at 6 m ( $0.007 \pm 0.003 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) and at 1.5 m ( $0.005 \pm 0.002 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ). With respect to PM10, the deposition flux at 10 m initially increased and then decreased. At heights of 1.5 m and 6 m the flux pattern changed in a similar manner to that associated with PM2.5 in which the highest fluxes occurred at 1.5 m and 10 m. The highest values at the three height levels occurred at the same time, as was the case for PM2.5. However, the maximum flux was  $9.18 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ , almost 6 times that recorded for PM2.5.

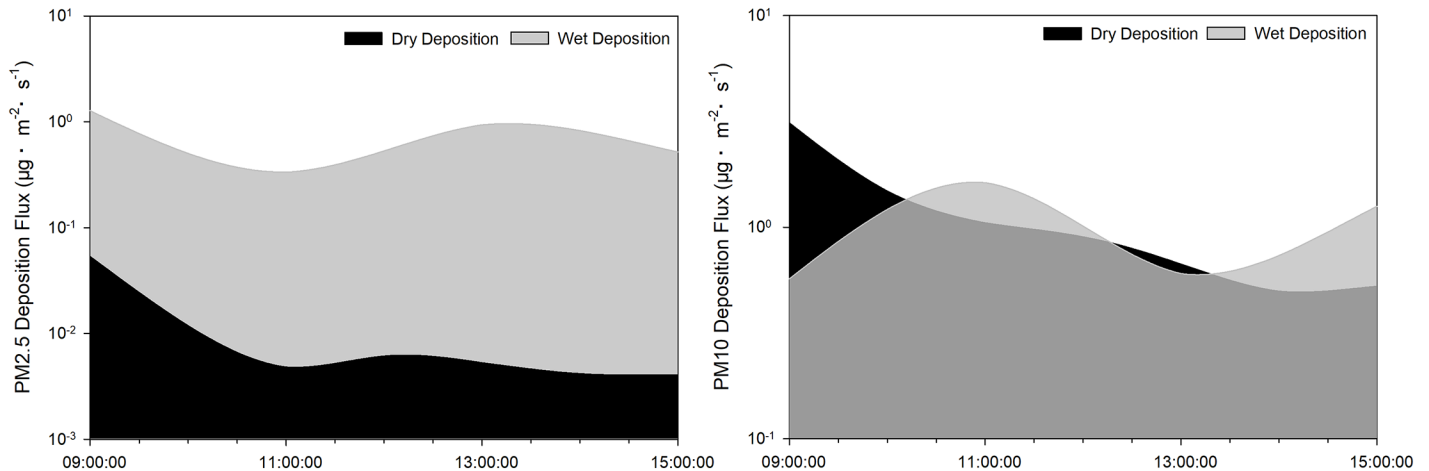
### Temporal and spatial variation in wet deposition

As shown in Fig 5, the wet deposition flux curves for PM2.5 and PM10 are changeable. PM2.5 deposition flux reached a peak at the onset of rain, after which it fell sharply. The minimum flux of PM2.5 occurred after four rainy hours, when the PM10 wet deposition flux underwent an increase. Thereafter, the PM2.5 flux increased and then declined. Opposite trends were



**Fig 4. PM dry deposition flux on different height levels.**

<https://doi.org/10.1371/journal.pone.0199241.g004>



**Fig 5. Temporal variation of dry and wet deposition.**

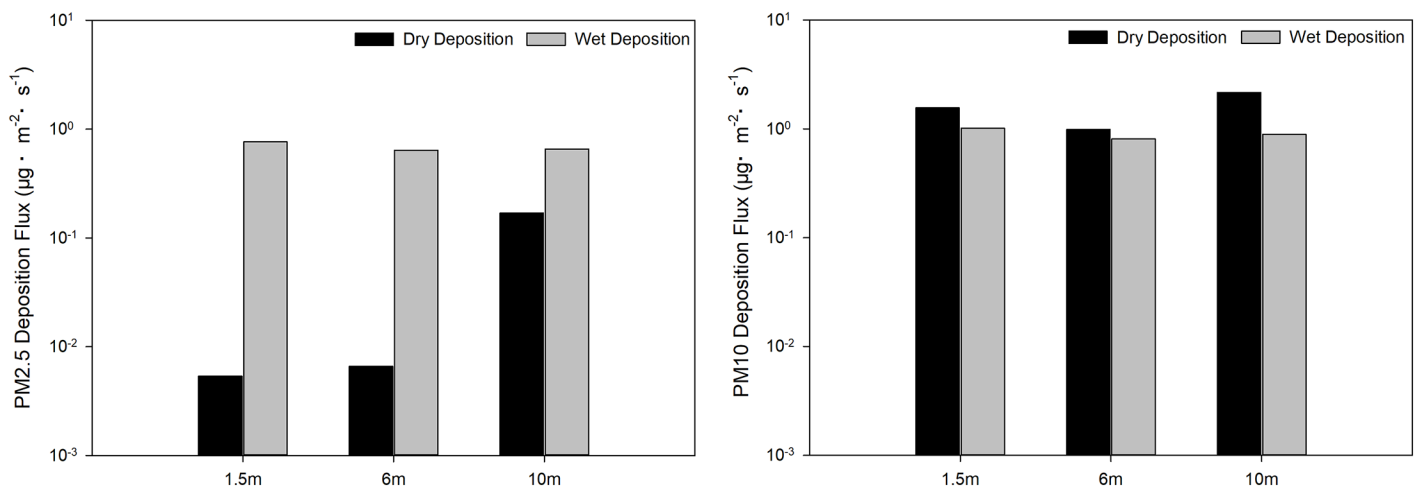
<https://doi.org/10.1371/journal.pone.0199241.g005>

noted in the case of PM10. The PM10 deposition flux during the precipitation process thus varied in an opposite manner to that observed for PM2.5. Deposition flux of PM2.5 declined from  $0.34 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$  to  $1.27 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ . In the case of PM10, the maximum and minimum wet deposition fluxes were  $1.62 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$  and  $0.57 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ , respectively. Unlike dry deposition, there was very little difference between the values for wet deposition flux for PM2.5 and PM10, being between PM2.5 ( $0.77 \pm 0.42 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) and PM10 ( $1.01 \pm 0.52 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ), respectively.

As illustrated in Fig 6, the wet deposition flux trends ( $1.5 \text{ m} > 10 \text{ m} > 6 \text{ m}$ ) were similar for PM2.5 or PM10, with minimal differences between 1.5m, 6m and 10m. The flux of PM10 ( $0.904 \pm 0.103 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) was, however, slightly larger than that of PM2.5 ( $0.688 \pm 0.069 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ).

### Comparison of dry and wet deposition

A comparison of temporal variation between dry and wet deposition is illustrated in Fig 5. PM2.5 deposition flux was changeable for wet deposition, within a range of  $0.335\text{--}1.273 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ . The flux reached a maximum at the start of precipitation and fell to a minimum



**Fig 6. Spatial variation of dry and wet deposition.**

<https://doi.org/10.1371/journal.pone.0199241.g006>

level after 2 h. Thereafter, the curve remained steady until 13:00, after which it declined. The changing tendency resulted in two 'V'-shaped graphs, one positive, and the other an inverted 'V'. In the case of dry deposition, the values for PM<sub>2.5</sub> remained steady. The wet deposition flux ( $0.766 \pm 0.422 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) was nearly 60 times the dry deposition flux ( $0.013 \pm 0.018 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ). Accordingly, wet deposition for PM<sub>2.5</sub> was much more obvious than dry deposition. In contrast, wet deposition flux was steady and the dry deposition flux was changeable in the case of PM<sub>10</sub>. Dry deposition flux ( $1.18 \pm 0.92 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) declined during the whole process, from 3.12 to  $0.50 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$  and made no big difference with wet deposition flux ( $1.01 \pm 0.52 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ).

Variation in dry and wet deposition at different height levels is illustrated in Fig 6. In the case of PM<sub>2.5</sub>, the order of dry deposition flux is  $10 \text{ m} > 6 \text{ m} > 1.5 \text{ m}$  and the flux at 10 m ( $0.17 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ) was far greater than that measured at 6 m and 1.5 m. In the case of wet deposition, the sequence trend was as follows:  $1.5 \text{ m} > 10 \text{ m} > 6 \text{ m}$ . However, there were no significant differences between these values. Wet deposition flux was, however, greater than dry deposition flux at each height level. In the case of PM<sub>10</sub>, the values for dry deposition flux were  $1.57 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ,  $0.99 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$  and  $2.16 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$  at 1.5 m, 6 m and 10 m, respectively. The flux at 10 m was highest, which was consistent with the results obtained for PM<sub>2.5</sub>. The values of PM<sub>10</sub> wet deposition flux were  $1.01 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ,  $0.81 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$  and  $0.89 \mu\text{g}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$  at 1.5 m, 6 m and 10 m, respectively.

## Discussion

### Meteorological factors and dry deposition

Meteorological factors, such as wind speed, temperature and relative humidity, can influence friction velocity and atmospheric stability [22]. Deposition velocity and PM concentration were also strongly affected by friction velocity and atmospheric stability [23–25]. Therefore, we can conclude that meteorological factors have an important impact on deposition velocity. The relationship between dry deposition velocity and meteorological factors was investigated by means of correlation analysis between dry deposition velocity and meteorological parameters, as illustrated in Fig 7 (at a confidence level of 95%).

The mean wind speed, temperature and relative humidity at the sampling site were 0.16 m/s, 24.99°C and 32.52%, respectively. These results indicated that there was no significant association between dry deposition velocity and wind speed of PM (Fig 7a and 7b). However, this result did not conform with the results of other studies [26,27] and the relevant explanations made by some physical models, such as firm theory and the resistance model [28–30]. Furthermore, wind speed could also influence the particles accumulation on leaves [31]. Wind speed was the strongest meteorological factor, which influenced dry deposition velocity [26] and had a positive correlation with the dry deposition velocity of TSP [22,26–29]. The results of our study may have been affected by the low wind speed conditions that prevailed during the sampling period.

The dependence of temperature on dry deposition velocity is illustrated in Fig 7c and 7d, which indicates the following correlation coefficients between dry deposition velocity and temperature:  $-0.72$  for PM<sub>2.5</sub> and  $-0.78$  for PM<sub>10</sub>. Other results indicate that PM<sub>2.5</sub> ( $R = 0.78$ ; Fig 7e) and PM<sub>10</sub> ( $R = 0.82$ ; Fig 7f) deposition velocities were significantly positively correlated with relative humidity. Hence, PM deposition velocity had a significant correlation with temperature and relative humidity. PM deposition velocity was negatively correlated with temperature and positively correlated with relative humidity. Similar results were reported in previous studies, which indicated a strong negative correlation between temperature and dry deposition velocity of TSP [22] and that an increase in relative humidity could lead to a significant increase in particle deposition rate, due to the increase in particle size [32].



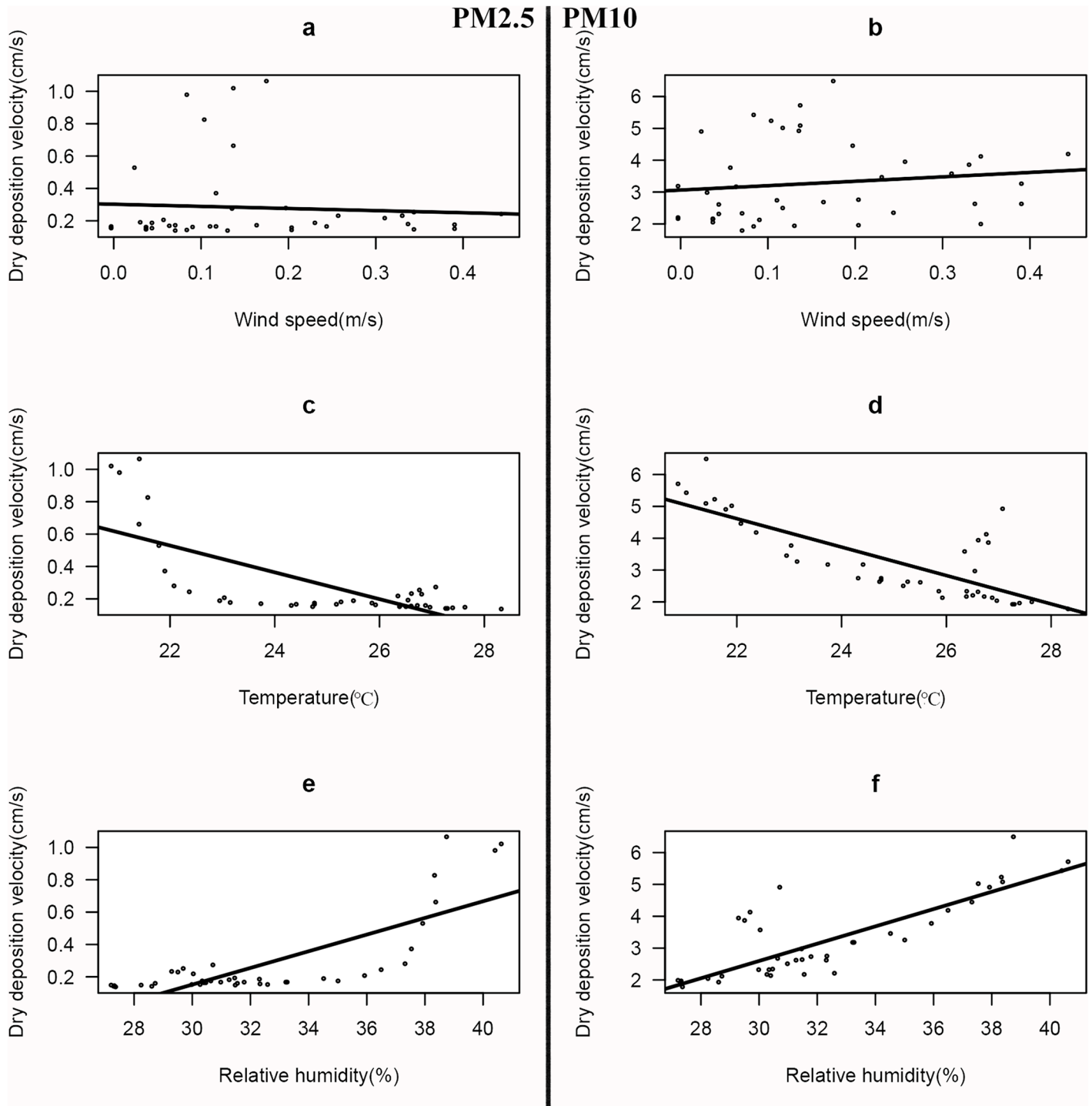


Fig 7. Relations between PMs dry deposition velocity and meteorological factors.

<https://doi.org/10.1371/journal.pone.0199241.g007>

### Rainfall intensity and wet deposition

As shown in Fig 8, PM wet deposition flux and rainfall intensity were changeable during precipitation. Variation in PM2.5 deposition flux was consistent with that in rainfall intensity. In

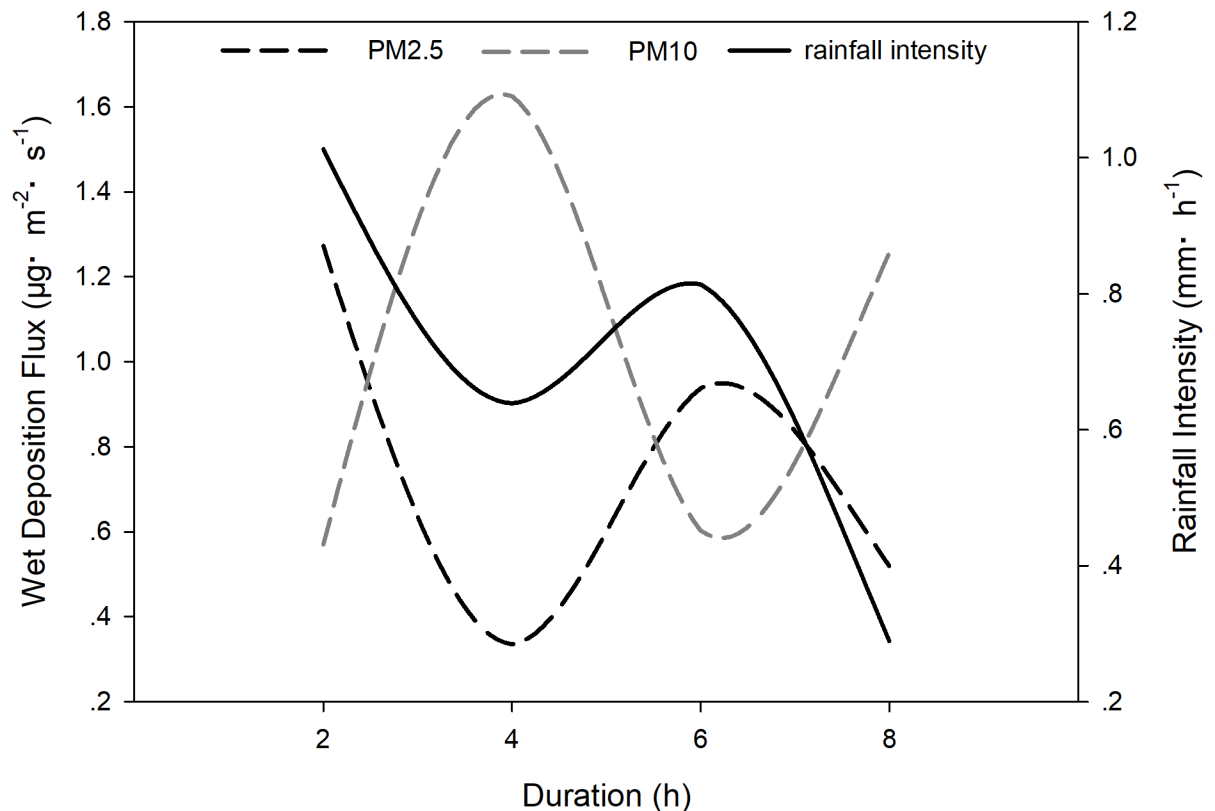


Fig 8. Relations between PMs wet deposition flux and rainfall intensity.

<https://doi.org/10.1371/journal.pone.0199241.g008>

contrast, variation in PM10 deposition flux was adverse to that in rainfall intensity. Rainfall intensity thus had opposite effects on PM2.5 and PM10. There were, however, no obvious differences between wet deposition fluxes of PM2.5 and PM10.

Wet deposition can reduce air pollution by removing PM and other pollutants [33]. The efficacy of this removal is related to the duration and intensity of the precipitation event [33–36].

Results of further research have indicated a negative correlation between precipitation rates and PM2.5 concentrations [36–40]. Increases in precipitation can cause an increase in wet deposition of PM2.5 and their gaseous precursors [41]. Results obtained in our study supported the conclusion that precipitation intensity is positively correlated with PM2.5 deposition efficiency.

Our results also indicated a linear relationship between the intensity and duration of rainfall and the value of the PM10 removal coefficient [42]. The scavenging coefficient  $\Lambda(S^{-1})$  was considered to be the most important parameter characterising the scavenging effects of particles [43,44]. Field experiment average scavenging coefficients of PM10 for different rain intensities are listed in Table 1 [45–48]. Unlike previous studies [45–48], the changed tendency of wet

Table 1. Average PM10 scavenging coefficient for different rain intensity.

Rain intensity (mm/h)	Bae et al. (2001)	Baklanov (2001)	Chate (2003)	Zhao (2006)
0.2–0.5(Light Rain)	8.50E-04	1.01E-03	1.20E-03	1.60E-03
0.5–4.0(Moderate Rain)	2.18E-03	1.38E-03	1.78E-03	3.63E-03
>4.0(Heavy Rain)	5.80E-03	1.90E-03	3.40E-03	7.20E-03

<https://doi.org/10.1371/journal.pone.0199241.t001>

deposition for PM<sub>10</sub> had an opposite variation to rainfall intensity. This may be due to the limited range of rainfall intensity (0.29–1.01 mm/h) during the study period.

## Concentration and deposition of PM

As shown in Fig 3, the concentration of PM reached a maximum at 8:00, mainly because temperature inversion limited the diffusion of pollutants during the morning [49]. Due to the high relative humidity, which caused a high density of vapour, secondary aerosols formed over wetlands in the late afternoon [50].

Variation trends in dry deposition velocities of PM<sub>2.5</sub> were similar to those of PM<sub>10</sub> (Fig 2), owing to the PM deposition being significantly correlated with the PM concentration [50]. Particle size had a great influence on dry deposition velocity, which tended to increase with particle size [12], i.e. the larger the particle, the higher the settling rate [51,52]. Changes in the range of velocities may be influenced by meteorological conditions, particle size distribution, particle morphology and chemical composition [12]. As shown in Figs 2 and 3, changing trends and the level of dry deposition flux followed the same laws as those of velocity since both the concentration and the deposition velocity had an impact on deposition flux [16]. As a result of the influence of gravity acceleration, deposition velocity increases with an increase in height [53]. Maximum deposition fluxes of PM<sub>2.5</sub> and PM<sub>10</sub> thus occurred at the height of 10 m. While the PM<sub>10</sub> deposition fluxes of 1.5 m were higher than those of 6 m in our study. It may be because of the particle dry deposition is a dynamic process, which may also be affected by spatial fluctuations, surface-type differences, temporal changes, diurnal variations and meteorological conditions [14]. And the low surface was also greatly influenced by human activity.

In the case of wet deposition, rain scavenging generally takes place via Brownian diffusion, inertial impaction, diffusiophoresis, thermophoresis and electrical charge effects [54–56]. The process of rain scavenging is affected by many factors, including raindrop size distribution and intensity, particle size distribution and concentration, the chemical and physical properties of droplets and atmospheric temperature regimes [57,58]. The PM<sub>10</sub> wet deposition fluxes at each height level were greater than those associated with PM<sub>2.5</sub>, which were shown in Fig 6. It has been confirmed that there exists a ‘Greenfield gap’ between particles (of sizes ranging from 1 to 2  $\mu\text{m}$ ) during the collection process of aerosol particles [59,60]. Particles of diameters of 0.1 to 2  $\mu\text{m}$  were too small to effectively get collected by inertial impaction and too large for Brownian diffusion, but the effect of inertial impaction and Brownian diffusion were much more efficient for particles of diameters of  $<0.1 \mu\text{m}$  and  $>2 \mu\text{m}$  [61]. This may explain the results that indicated greater collection efficiency for PM<sub>10</sub> than for PM<sub>2.5</sub>. The PM<sub>2.5</sub> wet deposition flux changed with changes in rainfall intensity during precipitation, but this was not the case for the PM<sub>10</sub> wet deposition flux. This phenomenon was mainly caused by the sensitivity of the scavenging coefficient to the aerosol size [61] and limitation of the range of rainfall intensity.

Compared to dry deposition, wet deposition was more effective in scavenging PM<sub>2.5</sub>, a result that was supported by a previous study [12]. While a dry process can remove PM<sub>10</sub> more effectively, the result was consistent with the conclusion that dry deposition flux was more skewed towards coarse/large particles than wet deposition flux [12].

## Conclusions

Based on this investigation, the following conclusions can be made:

Similar trends were noted for dry deposition, daily variation in concentration, deposition velocity, and flux for PM: they peaked at 8:00 and then decreased, but a slight revival was noted at 15:00. These activities therefore appeared to influence each other. It must be pointed

out that the values of PM10 were higher than those of PM2.5 and the maximum occurred at 10 m. For PM2.5, the order of dry deposition fluxes was 10 m > 6 m > 1.5 m and for PM10 the flux order was 10 m > 1.5 m > 6 m. In the case of wet deposition, PM10 deposition flux values at each height level were more than PM2.5 deposition flux values, and the sequences of both PM2.5 and PM10 deposition fluxes were as follows: 1.5 m > 10 m > 6 m. In comparison with dry deposition, wet deposition was more efficient in terms of PM2.5 removal. Nevertheless, dry deposition was more tilted towards PM10 than wet deposition.

The dry deposition velocity of PM had a significant negative correlation with temperature. The correlation coefficients of dry deposition velocity and temperature were  $-0.72$  for PM2.5 and  $-0.78$  for PM10, and were positively correlated with relative humidity. The correlation coefficients of dry deposition velocity and relative humidity were  $0.78$  for PM2.5 and  $0.82$  for PM10. Rainfall intensity and PM diameter were important factors that influenced wet deposition efficiency. Many aspects of this process require further research.

In summary, the effects of dry and wet deposition in wetland on the concentrations of atmospheric particulates were evaluated and compared in our study. This work provided the data foundation for measures which could help improving air quality. The removal efficiency of PM2.5 by wet deposition was remarkable, indicating that it is necessary to control the air pollutant emissions during the dry weather season. What's more, some previous studies [62–64] focus on the relationship between deposition and metals. It was surprisingly found that there was no significant correlation between metals in PM10 and rainfall except for vanadium and nickel. And It was difficult to predict metal scavenging by rainfall characteristics [62]. These processes are not considered in our study, therefore, more attention should be paid to further investigations and field studies about the metals change during the wet deposition.

## Acknowledgments

This research was supported by the Fundamental Research Funds for the Central Universities (2016JX05), Forestry Public Welfare Projects Special Fund of China (201304301), and Beijing Municipal Science and Technology Project (Z141100006014031).

## Author Contributions

**Conceptualization:** Zhenming Zhang.

**Data curation:** Ling Cong, Yu Wang.

**Formal analysis:** Jiakai Liu, Jiexiu Zhai, Yu Wang, Wenmei Ma.

**Investigation:** Yanan Wu, Ling Cong.

**Methodology:** Yanan Wu.

**Project administration:** Zhenming Zhang.

**Resources:** Chunyi Li.

**Software:** Jiakai Liu, Jiexiu Zhai, Wenmei Ma.

**Writing – original draft:** Yanan Wu, Jiakai Liu.

**Writing – review & editing:** Yanan Wu, Zhenming Zhang.

## References

1. Baik NJ, Yong PK, Moon KC. Visibility study in Seoul, 1993. *Atmos Environ.* 1996; 30: 2319–2328.

2. Law KS, Stohl A. Arctic air pollution: Origins and impacts. *Science*. 2007; 315: 1537–1540. <https://doi.org/10.1126/science.1137695> PMID: 17363665
3. Kesavachandran C, Pangtey BS, Bihari V, Fareed M, Pathak MK, Srivastava AK, et al. Particulate matter concentration in ambient air and its effects on lung functions among residents in the National Capital Region, India. *Environ Monit Assess*. 2013; 185: 1265–1272. <https://doi.org/10.1007/s10661-012-2630-0> PMID: 22527464
4. Li MH, Fan LC, Mao B, Yang JW, Choi AM, Cao WJ, et al. Short-term Exposure to Ambient Fine Particulate Matter Increases Hospitalizations and Mortality in COPD A Systematic Review and Meta-analysis. *Chest*. 2016; 149: 447–458. <https://doi.org/10.1378/chest.15-0513> PMID: 26111257
5. Apte JS, Marshall JD, Cohen AJ, Brauer M. Addressing Global Mortality from Ambient PM2.5. *Environ Sci Technol*. 2015; 49: 8057–8066. <https://doi.org/10.1021/acs.est.5b01236> PMID: 26077815
6. Lim SS, Vos T, Flaxman AD, Danaei G, Shibuya K, Adair-Rohani H, et al. A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: a systematic analysis for the Global Burden of Disease Study 2010. *Lancet*. 2012; 380: 2224–2260. [https://doi.org/10.1016/S0140-6736\(12\)61766-8](https://doi.org/10.1016/S0140-6736(12)61766-8) PMID: 23245609
7. Mundt KA. An examination of the Environmental Protection Agency risk assessment principles and practices: a brief commentary on section 4.1.3 of the EPA March 2004 Staff Paper. *Hum Exp Toxicol*. 2006; 25: 19–21. <https://doi.org/10.1191/0960327106ht580oa> PMID: 16459710
8. Zhang LL, Gao YX, Dao X, Wang C, Teng E. Composition and Distribution of Elements in Air Particulate Matters during Heating Season of Beijing-Tianjin-Hebei Megacities, China. *Environmental Monitoring in China*. 2014; 30: 53–61.
9. Chen R, Kan H, Chen B, Huang W, Bai Z, Song G, et al. Association of Particulate Air Pollution With Daily Mortality. *Am J Epidemiol*. 2012; 175: 1173–1181. <https://doi.org/10.1093/aje/kwr425> PMID: 22510278
10. Shannigrahi AS, Fukushima T, Ozaki N. Comparison of different methods for measuring dry deposition fluxes of particulate matter and polycyclic aromatic hydrocarbons (PAHs) in the ambient air. *Atmos Environ*. 2005; 39: 653–662.
11. Dolske DA, Gatz DF. A field comparison of methods for the measurement of particle and gas deposition. *J Geophys Res*. 1985; 90: 2076–2084.
12. Tai AY, Chen LA, Wang X, Chow JC, Watson JG. Atmospheric deposition of particles at a sensitive alpine lake: Size-segregated daily and annual fluxes from passive sampling techniques. *Sci Total Environ*. 2017; 579: 1736–1744. <https://doi.org/10.1016/j.scitotenv.2016.11.117> PMID: 27932212
13. Chate DM, Rao P, Naik MS, Momin GA, Safai PD, Ali K. Scavenging of aerosols and their chemical species by rain. *Atmos Environ*. 2003; 37: 2477–2484.
14. Zellner R. *Atmospheric Chemistry: Fundamentals and Experimental Techniques*, J. Wiley and Sons, New York, Chichester, Brisbane, Toronto and Singapore 1986. 1098 Seiten, Preis: £ 57.45. *Berichte der Bunsengesellschaft für physikalische Chemie*. 1986; 90: 1244.
15. Guo L, Bao L, She J, Zeng EY. Significance of wet deposition to removal of atmospheric particulate matter and polycyclic aromatic hydrocarbons: A case study in Guangzhou, China. *Atmos Environ*. 2014; 83: 136–144.
16. Zhu LJ, Liu JK, Cong L, Ma WM, Ma W, Zhang ZM. Spatiotemporal Characteristics of Particulate Matter and Dry Deposition Flux in the Cuihu Wetland of Beijing. *Plos One*. 2016; 11: e0158616. <https://doi.org/10.1371/journal.pone.0158616> PMID: 27437688
17. Qiu DD, Liu JK, Zhu LJ, Mo LC, Zhang ZM. Particulate matter assessment of a wetland in Beijing. *Journal of Environmental Sciences*. 2015; 36: 93–101.
18. Liu JK, Zhu LJ, Wang HH, Yang YL, Liu JT, Qiu DD, et al. Dry deposition of particulate matter at an urban forest, wetland and lake surface in Beijing. *Atmos Environ*. 2016; 125: 178–187.
19. Petroff A, Mailliat A, Amielh M, Anselmetti F. Aerosol dry deposition on vegetative canopies. Part I: Review of present knowledge. *Atmos Environ*. 2008; 42: 3625–3653.
20. Slinn SA, Slinn WGN. Predictions for particle deposition on natural waters. *Atmos Environ*. 1980; 14: 1013–1016.
21. Zhang L, Gong S, Padro J, Barrie L. A size-segregated particle dry deposition scheme for an atmospheric aerosol module. *Atmos Environ*. 2001; 35: 549–560.
22. Chen L, Peng S, Liu J, Hou Q. Dry deposition velocity of total suspended particles and meteorological influence in four locations in Guangzhou, China. *Journal of Environmental Sciences*. 2012; 24: 632–639.
23. Tai APK, Mickley LJ, Jacob DJ. Correlations between fine particulate matter (PM2.5) and meteorological variables in the United States: Implications for the sensitivity of PM2.5 to climate change. *Atmos Environ*. 2010; 44: 3976–3984.

24. Mammarella I, Rannik Ü, Aalto P, Keronen P, Vesala T, Kulmala M. Long-term aerosol particle flux observations. Part II: Particle size statistics and deposition velocities. *Atmos Environ*. 2011; 45: 3794–3805.
25. Connan O, Maro D, Hébert D, Roupsard P, Goujon R, Letellier B, et al. Wet and dry deposition of particles associated metals (Cd, Pb, Zn, Ni, Hg) in a rural wetland site, Marais Vernier, France. *Atmos Environ*. 2013; 67: 394–403.
26. Fang G, Chiang H, Chen Y, Xiao Y, Wu C, Kuo Y. A measurement of summertime dry deposition of ambient air particulates and associated metallic pollutants in Central Taiwan. *Environ Geochem Hlth*. 2015; 37: 233–249.
27. Odabasi M, Muezzinoglu A, Bozlaker A. Ambient concentrations and dry deposition fluxes of trace elements in Izmir, Turkey. *Atmos Environ*. 2002; 36: 5841–5851.
28. Schwarzenbach RP, Gschwend PM, Imboden DM. Environmental organic chemistry: illustrative examples, problems, and case studies. *J Contam Hydrol*. 1996; 23: 361–362.
29. Hicks BB, Baldocchi DD, Meyers TP, Hosker RP, Matt DR. A preliminary multiple resistance routine for deriving dry deposition velocities from measured quantities. *Water Air Soil Pollut*. 1987; 36: 311–330.
30. Liu JK, Yan GX, Wu YN, Wang Y, Zhang ZM, Zhang MX. Wetlands with greater degree of urbanization improve PM2.5 removal efficiency. *Chemosphere*. 2018; 207: 601–611. <https://doi.org/10.1016/j.chemosphere.2018.05.131> PMID: 29843037
31. Yan GX, Liu JK, Zhu LJ, Zhai JX, Cong L, Ma WM, et al. Effectiveness of wetland plants as biofilters for inhalable particles in an urban park. *J Clean Prod*. 2018; 194: 435–443.
32. Mohan SM. An overview of particulate dry deposition: measuring methods, deposition velocity and controlling factors. *Int J Environ Sci Te*. 2016; 13: 387–402.
33. Guo L, Bao L, She J, Zeng EY. Significance of wet deposition to removal of atmospheric particulate matter and polycyclic aromatic hydrocarbons: A case study in Guangzhou, China. *Atmos Environ*. 2014; 83: 136–144.
34. González CM, Aristizábal BH. Acid rain and particulate matter dynamics in a mid-sized Andean city: the effect of rain intensity on scavenging. *Atmos Environ*. 2012; 60: 164–171.
35. Dawson JP, Adams PJ, Pandis SN. Sensitivity of PM2.5 to climate in the Eastern US: a modeling case study. *Atmos Chem Phys*. 2007; 7: 4295–4309.
36. Lecoeur E, Seigneur C. Dynamic evaluation of a multi-year model simulation of particulate matter concentrations over Europe. *Atmos Chem Phys*. 2013; 13: 4319–4337.
37. Hedegaard GB, Brandt J, Christensen JH, Frohn LM, Geels C, Hansen KM, et al. Impacts of Climate Change on Air Pollution Levels in the Northern Hemisphere with Special Focus on Europe and the Arctic. *Atmospheric Chemistry & Physics*. 2008; 8: 568–576.
38. Jacob DJ, Winner DA. Effect of climate change on air quality. *Atmos Environ*. 2009; 43: 51–63.
39. Jiménez-Guerrero P, Montávez JP, Gómez-Navarro JJ, Jerez S, Lorente-Plazas R. Impacts of climate change on ground level gas-phase pollutants and aerosols in the Iberian Peninsula for the late XXI century. *Atmos Environ*. 2012; 55: 483–495.
40. Manders AMM, Meijgaard EV, Mues AC, Kranenburg R, Uift LHV, Schaap M. The impact of differences in large-scale circulation output from climate models on the regional modeling of ozone and PM. *Atmos Chem Phys*. 2012; 12: 9441–9458.
41. Megaritis AG, Fountoukis C, Charalampidis PE, Denier van der Gon HAC, Pilinis C, Pandis SN. Linking climate and air quality over Europe: effects of meteorology on PM2.5 concentrations. *Atmos Chem Phys*. 2014; 14: 10283–10298.
42. Olszowski T. Changes in PM10 concentration due to large-scale rainfall. *Arab J Geosci*. 2016; 9: 160.
43. Radke LF, Hobbs PV, Eltgroth MW. Scavenging of aerosol particles by precipitation. *Journal of Applied Meteorology*. 1980; 19: 715–722.
44. SAI. User's guide to the regional modeling system for aerosols and deposition (REMSAD) version 8. Systems Applications International: San Rafael, California, USA. 2005.
45. Bae SY, Jung CH, Kim YP. Derivation and verification of an aerosol dynamics expression for the below-cloud scavenging process using the moment method. *J Aerosol Sci*. 2010; 41: 266–280.
46. Baklanov A, Sørensen JH. Parameterisation of radionuclide deposition in atmospheric long-range transport modelling. *Physics and Chemistry of the Earth, Part B: Hydrology, Oceans and Atmosphere*. 2001; 26: 787–799.
47. Chate DM, Rao P, Naik MS, Momin GA, Safai PD, Ali K. Scavenging of aerosols and their chemical species by rain. *Atmos Environ*. 2003; 37: 2477–2484.
48. Zhao HB, Zheng CG. Monte Carlo solution of wet removal of aerosols by precipitation. *Atmos Environ*. 2006; 40: 1510–1525.

49. Guo JC, Qi S, Shen YK, Wu JL. PM2.5 mass concentration changes of two kinds urban forest and its correlation with meteorological factors. *J Soil Water Conserv.* 2014; 28: 88–93.
50. Wu Z, Wang C, Xu J, Hu L. Air-borne anions and particulate matter in six urban green spaces during the summer. *Journal of Tsinghua University(Science and Technology).* 2007; 47: 2153–2157.
51. Khan FI, Abbasi SA. Cushioning the impact of toxic release from runaway industrial accidents with greenbelts. *J Loss Prevent Proc.* 2000; 13: 109–124.
52. Khan FI, Abbasi SA. Effective design of greenbelts using mathematical models. *J Hazard Mater.* 2001; 81: 33–65. PMID: [11118683](#)
53. Li WQ. *Fundamentals of aerosol pollution chemistry.* Yellow River Conservancy Press: Zhengzhou. 2010: 226–227.
54. Santachiara G, Prodi F, Belosi F. Atmospheric aerosol scavenging processes and the role of thermo- and diffusio-phoretic forces. *Atmos Res.* 2013; 128: 46–56.
55. Chate DM, Rao P, Naik MS, Momin GA, Safai PD, Ali K. Scavenging of aerosols and their chemical species by rain. *Atmos Environ.* 2003; 37: 2477–2484.
56. Olszowski T. Concentration changes of PM10 under liquid precipitation on conditions. *Ecol Chem Eng S.* 2015; 22: 363–378.
57. Davenport HM, Peters LK. Field studies of atmospheric particulate concentration changes during precipitation. *Atmos Environ.* 1978; 12: 997–1008.
58. Zhao HB, Zheng CG. Monte Carlo solution of wet removal of aerosols by precipitation. *Atmos Environ.* 2006; 40: 1510–1525.
59. Greenfield SM. Rain Scavenging of Radioactive Particulate Matter from the Atmosphere. *Journal of Atmospheric Sciences.* 2010; 14: 115–125.
60. Wang PK, Grover SN, Pruppacher HR. On the effect of electric charges on the scavenging of aerosol particles by clouds and small raindrops. *Journal of Atmospheric Sciences.* 1978; 35: 1735–1743.
61. Chate DM, Pranesha TS. Field studies of scavenging of aerosols by rain events. *J Aerosol Sci.* 2004; 35: 695–706.
62. Lamprea K, Percot S, Ruban V, Maro D, Roupsard P, Millet M. Characterization of atmospheric deposition in a small suburban catchment. *Environ Technol.* 2011; 32: 1141–1149. <https://doi.org/10.1080/09593330.2010.528045> PMID: [21882566](#)
63. Percot S, Ruban V, Roupsard P, Maro D, Millet M. Use of beryllium-7 as a surrogate to determine the deposition of metal and polycyclic aromatic hydrocarbon through urban aerosols in Nantes, France. *Atmos Environ.* 2013; 74: 338–345.
64. Omrani M, Ruban V, Ruban G, Lamprea K. Assessment of atmospheric trace metal deposition in urban environments using direct and indirect measurement methodology and contributions from wet and dry depositions. *Atmos Environ.* 2017; 168: 101–111.