PM$_{2.5}$ decrease with precipitation as revealed by single-point ground-based observation

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Abstract

This study analyzed the observed data of precipitation and PM$_{2.5}$ concentration collected over two and a half years at Tsujido, Fujisawa City, to examine a quantitative relationship between the two quantities. It was observed that the PM$_{2.5}$ concentration decreased as precipitation increased. A composite analysis revealed that the PM$_{2.5}$ concentration on average decreases by 20.99% 1 h after the onset of precipitation as compared to that 1 h before, which is statistically significant based on a paired $t$-test. The PM$_{2.5}$ concentrations decreased over 8 h period from 5 h ($-$5 h) before to 3 h (+3 h) after the onset of precipitation, with a particularly dramatic decrease at the onset of precipitation. Analysis of PM$_{2.5}$ scavenging rates by precipitation intensity showed that the scavenging rate was 2.03% for light precipitation events, 28.15% for moderate precipitation events, and 26.75% for heavy precipitation events, indicating that the scavenging rate increased with moderate precipitation intensity and above. It is suggested that the deposition processes of rainout and washout are effective in reducing PM$_{2.5}$ concentration with precipitation.

KEYWORDS

aerosols, observational data analysis, physical phenomenon, physical phenomenon, rainfall, tools and methods

1 | INTRODUCTION

Aerosols affect the Earth’s radiation budget by changing the optical properties of clouds (indirect effect, Twomey, 1977), which influence radiative processes through an albedo effect. An increase in the concentration of atmospheric aerosols increases the number and concentration of cloud particles, as aerosols serve as cloud condensation nuclei and ice nuclei. As a result, aerosols affect precipitation on the ground, and influence radiative processes through cloud lifetime, and changes in cloud cover and volume (e.g., Miyamoto, 2021; Miyamoto et al., 2020; Sato et al., 2015). This is called the “lifetime effect” (Albrecht, 1989). In the Coupled Model Inter-comparison Project (CMIP6), the major contributors to the effective radiative forcing spread are the instantaneous radiative forcing and the aerosol-cloud interaction, with the aerosol forcing spread ranging from $-0.63$ to $-1.37$ W m$^{-2}$ (Smith et al., 2020). Thus, the aerosol–cloud interaction is the most uncertain process in predicting global warming. Therefore, a deeper understanding of aerosol–cloud interaction is required.

Observational studies have indicated causal relationships inferred from correlation of a cloud-related quantity...
with aerosol content, but it is difficult to identify the effect of aerosols due to the complexity of the aerosol–cloud interaction and the variety of meteorological fields (Brenguier et al., 2003; George & Wood, 2010). As a result, quantitative understanding based on observations is not fully established. A lack of understanding of the detailed mechanisms governing the efficiency of aerosol removal in moist convection is the major source of uncertainty in numerical simulations of aerosol spatial distribution (Textor et al., 2006).

There are two mechanisms by which aerosols are actually removed from the atmosphere: dry deposition and wet deposition. The latter indicates that atmospheric aerosols are removed from the ground surface by precipitation, in which aerosol particles become cloud condensation nuclei and ice nuclei or are directly trapped in cloud droplets, and the below-cloud scavenging process, in which particles below the cloud base are scavenged by precipitation.

In the wet deposition processes, many studies have confirmed the PM$_{2.5}$ concentration scavenging below clouds by precipitation, especially in China where air pollution is a serious social problem (Baе et al., 2012; Li et al., 2017; Luan et al., 2019; Mircea et al., 2000; Pranesha & Kamra, 1997). It has been shown that there is a correlation between the rate of change of PM$_{2.5}$ concentration and precipitation intensity by estimating polydisperse scavenging coefficients for scavenging processes below-cloud (Mircea et al., 2000) and a significant negative correlation between PM$_{2.5}$ concentration and precipitation (Li et al., 2017). In addition, the change in PM$_{2.5}$ concentration is related to the size of raindrops, and raindrops with a diameter of 1 mm or more play a major role in the removal of PM$_{2.5}$ with a diameter of 1–2 μm (Pranesha & Kamra, 1997). Hence, the size distribution of raindrops is an important factor in the removal efficiency of the below-cloud aerosols (Baе et al., 2012). However, most of these studies focused on the removal rate and particle size of aerosols below the cloud base and few studies examined the duration time scale of deposition, that is, how long it takes to reduce PM$_{2.5}$ concentration associated with precipitation events, and the amount of deposition to quantitatively clarify the deposition process. Therefore, the purpose of this study was to analyze the observation data for precipitation and PM$_{2.5}$ in Tsujido, Fujisawa City, Kanagawa Prefecture, Japan, and to clarify the relationship between the two quantities. The observation data and methodology for the analyses are presented in Section 2. A discussion on the obtained results with a comparison with previous studies and a summary are provided in Sections 4 and 5, respectively.

## 2 | DATA AND METHODOLOGY

The PM$_{2.5}$ mass concentration data for 1 h at the Meiji Municipal Center in Fujisawa City, Kanagawa Prefecture, collected by the Ministry of the Environment's Wide-area Monitoring System for Air Pollutants (http://soramame.taiki.go.jp) was utilized in this study. For continuous monitoring, an automatic measuring machine is used, which is recognized to provide values equivalent to the mass concentration measured by the filter collection-mass method (cf. National Institute for Environmental Studies, Japan). The analysis period was from 0:00 on October 1, 2017 to 24:00 on March 31, 2020. Surface precipitation data for 1 h were obtained from the Automated Meteorological Data Acquisition System (AMeDAS) at Tsujido, Fujisawa City. The time in the manuscript shows the local time in Japan. The observation station is located near the coast and approximately 50 km from the center of Tokyo. Hence, the meteorological features at Tsujido are quite similar to those around Tokyo of which the annual mean of precipitation is about 1500 mm. The yearly average of precipitation and the PM$_{2.5}$ concentration at the station are approximately 1500 mm and 10 μg m$^{-3}$, respectively (figure not shown). Compared with Beijing (Luan et al., 2019), Tsujido has about twice as much annual precipitation and about one-third lower PM$_{2.5}$ concentration.

We conducted a composite analysis for the time variation of the PM$_{2.5}$ concentration during precipitation. First, the individual events in which the precipitation was greater than 0.5 mm were extracted. Then, to analyze the change in PM$_{2.5}$ concentration before and after the onset of precipitation, the average was calculated at each time from 12 h before (−12 h) to 12 h after (+12 h) the onset of precipitation. To examine whether there is a statistically significant difference in the PM$_{2.5}$ concentration before and after the onset of precipitation, a paired $t$-test was conducted. Specifically, the difference in the PM$_{2.5}$ concentration for the 2 h around the onset of precipitation (i.e., from −1 h to +1 h) was compared with the difference for another 2 h before the onset (i.e., from −3 h to −2 h). Significance was set at $p < 0.05$.

We analyzed the relationship between the PM$_{2.5}$ scavenging rate and precipitation using the following equation, which defines the PM$_{2.5}$ scavenging rate ($\Delta C$) as the change in PM$_{2.5}$ concentration before and during precipitation (Feng & Wang, 2012; Luan et al., 2019; Olszowski, 2016).

$$\Delta C = \frac{C_b - C_p}{C_b} \times 100\%,$$  \hspace{1cm}(1)$$

where $C_b$ is the average PM$_{2.5}$ concentration within 3 h before the onset of precipitation, and $C_p$ is the average PM$_{2.5}$ concentration during precipitation, respectively.
Thus, larger values of $\Delta C$ indicate that more amount of PM$_{2.5}$ is scavenged due to precipitation. $\Delta C$ below $-100$ and $\Delta C$ above $100$ were excluded. We also used the $\Delta C_{dry}$ equation (Luan et al., 2019) to analyze changes in PM$_{2.5}$ during no precipitation.

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

where $C_d$ is the average PM$_{2.5}$ concentration for no precipitation events from 3 to 6 h before the onset of precipitation.

We divided maximum precipitation of individual events into three categories for the analysis of the PM$_{2.5}$ scavenging rate by precipitation intensity: light precipitation events were defined as those with the precipitation less than 2.5 mm h$^{-1}$, moderate precipitation events as from 2.6 to 7.6 mm h$^{-1}$, and heavy precipitation events as 7.7 mm h$^{-1}$ or more, respectively (cf. American Meteorological Society, 2019; Luan et al., 2019). When the duration of no precipitation is more than three consecutive hours, it is considered as a separate precipitation event.

3 | RESULTS

3.1 | PM$_{2.5}$ concentrations decrease with precipitation

Figure 1 shows the scatter plot of one-hour precipitation and PM$_{2.5}$ concentration observed at Tsujido for the 2.5-year analysis period. It shows that overall the higher the precipitation, the lower the PM$_{2.5}$ concentration. In particular, when the 1-h precipitation was greater than 15 mm, the PM$_{2.5}$ concentration never exceeded 20 $\mu$g m$^{-3}$. In contrast, when the 1-h precipitation was 0 mm, the PM$_{2.5}$ concentration was widely distributed.

Figure 2 shows the time variation of 1 h of precipitation and PM$_{2.5}$ concentration for two events, observed at Tsujido from 3:00 to 13:00 on April 8, 2019 and from 19:00 on October 7 to 6:00 on October 8, 2019, which are typical examples of PM$_{2.5}$ concentrations decreasing with precipitation. For both events, as precipitation began, the PM$_{2.5}$ concentration decreased, and as the precipitation continued, the PM$_{2.5}$ concentration decreased further. The PM$_{2.5}$ concentration also decreased before the onset of precipitation. After the precipitation completed, the PM$_{2.5}$ concentration was less than 50% of the original value before the precipitation began.

Figure 3 shows the composited temporal change in PM$_{2.5}$ concentration from 12 h before to 12 h after the onset of precipitation. The composites were constructed from all precipitation events with one-hour precipitation greater than 0.5 mm observed at Tsujido during the analysis period. The PM$_{2.5}$ concentrations for each event were
normalized by the value at the onset of precipitation. The total number of precipitation events during the analysis period was 237.

The average of PM$_{2.5}$ concentration decreased by 20.99\% from 1 h (−1 h) before to 1 h (+1 h) after precipitation, with a dramatic decrease at the onset of precipitation. The average of PM$_{2.5}$ concentrations 12 h before the onset of precipitation are higher than those for 12 h after the onset of precipitation. The average of PM$_{2.5}$ concentrations decreased over 8 h from 5 h (−5 h) before to 3 h (+3 h) after the onset of precipitation with the drastic drop from −1 h to +1 h. Then, the average of PM$_{2.5}$ concentration gradually increased after +4 h. The standard deviation of the PM$_{2.5}$ concentration did not significantly change over time. A paired $t$-test for these differences showed that there were statistically significant differences between the PM$_{2.5}$ concentrations at −1 h and at +1 h of precipitation onset. In addition, the difference between the PM$_{2.5}$ concentrations for the 2 h from −1 h to +1 h of precipitation onset and −10 h to −8 h, −9 h to −7 h, −8 h to −6 h, and −7 h to −5 h, that is, the differences without precipitation, were also shown to be statistically significant.

3.2 | Relationship between precipitation intensity and PM$_{2.5}$ scavenging rates

Figure 4 shows the frequency distribution of PM$_{2.5}$ scavenging rates $\Delta C$ for events with three different categories of precipitation intensity, namely light, moderate, and heavy precipitation events, and all events. Light precipitation events account for 62.37\% of all precipitation events, moderate precipitation events for 23.39\%, and heavy precipitation events for 14.24\%. The averages of $\Delta C$ and their standard deviations were $2.03\% \pm 38.59\%$ for the light precipitation events, $28.15\% \pm 34.80\%$ for the moderate precipitation events, and $26.75\% \pm 33.69\%$ for heavy precipitation events, respectively. Thus, the scavenging rate increased with precipitation intensity.

![FIGURE 3](image3.png)

**FIGURE 3** Composited time series of PM$_{2.5}$ concentration from 12 h before and after the onset of precipitation for the events with one-hour precipitation was greater than 0.5 mm. The PM$_{2.5}$ concentrations for individual event are normalized by the value at the onset of precipitation. The black dot and line indicate the average of the normalized value and the error bars indicate the half of standard deviation ($0.5 \sigma$) at each time.

![FIGURE 4](image4.png)

**FIGURE 4** Frequency distributions of PM$_{2.5}$ scavenging rates by precipitation intensity for (a) all precipitation events, (b) light precipitation events, (c) moderate precipitation events, and (d) heavy precipitation events.
Figure 5 shows the box plots of total precipitation and maximum precipitation in the individual precipitation events as a function of ΔC. It shows a positive relationship between the median of both total precipitation and maximum precipitation and ΔC. The highest median of total precipitation was 15 mm at 40%–60% scavenging rate (ΔC). The highest median of maximum precipitation was 4.5 mm at 40%–60% scavenging rate (ΔC).

Quantile regression analysis was also conducted for both total precipitation and maximum precipitation, and the regression line at the 50% tile was shown in the figure. The regression lines of total precipitation and maximum precipitation were $y = 0.07x + 4.16$ and $y = 0.02x + 1.71$, respectively. The result also shows that stronger precipitation results in a larger amount of scavenging of PM$_{2.5}$ concentration.

4 | DISCUSSION

The scatter plot in Figure 1 shows that PM$_{2.5}$ concentration is low as precipitation is high, suggesting the presence of the following two processes. The first is the nucleation scavenging in which aerosols were taken up by cloud droplets in the cloud (rainout). Whereas there are a number of processes in the in-cloud scavenging, previous studies indicated that the nucleation scavenging is dominant among the others (Alheit et al., 1990; Flossmann et al., 1985). This would play a role to decrease the PM$_{2.5}$ concentration prior to the onset of precipitation in the time variation analysis (Figure 2) and composite analysis (Figure 3). The second is that aerosols were taken up by precipitation particles below the cloud base (washout), that is, impaction scavenging. This appears to play a role to decrease the PM$_{2.5}$ concentration after the onset of precipitation in the composite analysis (Figure 3).

The composite analysis showed a decrease of 20.99% in PM$_{2.5}$ concentration in the 1 h before and after the onset of precipitation (Figure 3), which is also likely to be due to the deposition processes of rainout and washout. Since there is a statistically significant difference in the reduction of PM$_{2.5}$ concentration in the 1 h before and after the precipitation onset (Figure 3) and the PM$_{2.5}$ scavenging rate increases with increasing precipitation intensity (Figures 4 and 5), it is suggested that the more effective the deposition is due to the accelerated collisional growth of cloud droplets during the washout process. Another possible cause of the decrease in PM$_{2.5}$ concentration is advection. Although the contribution of advection terms should be examined to consider the budget of PM$_{2.5}$ concentration using observed data for winds, the spatial distribution of PM$_{2.5}$, which is also required to calculate the advection term, was not available. To further support these results, we calculated the correlation coefficient between maximum precipitation and ΔC using a scatter plot, and the correlation coefficient was 0.25, indicating a weak correlation. In addition, we used the ΔC$_{dry}$ equation defined in (2) to analyze the change in PM$_{2.5}$ during no precipitation. The average and standard deviation of ΔC$_{dry}$ was $-8.86% \pm 45.28\%$, while that of ΔC was $11.66% \pm 39.08\%$. The result showed that PM$_{2.5}$ concentrations tended to increase when there was no precipitation before precipitation, and PM$_{2.5}$ concentrations also decreased after precipitation, which is consistent with Luan et al. (2019).

In addition, while the average ΔC in light precipitation events was small, light precipitation events account for the majority (62.37%) of all precipitation events in this study, and this supports previous results that light precipitation climatologically controls aerosols wet removal (Wang, Xia, Liu, et al., 2021; Wang, Xia, Zhang, 2021).

The mean PM$_{2.5}$ scavenging rate and the standard deviations in Beijing (Luan et al., 2019), where air pollution is serious, were $5.1\% \pm 25.7\%$, $38.5\% \pm 29.0\%$, and $50.6\% \pm 21.2\%$ for light, moderate, and heavy precipitation events, respectively, whereas in Tsujido, they were $2.03\% \pm 38.59\%$, $28.15\% \pm 34.80\%$, and $26.75\% \pm 33.69\%$, respectively, indicating that the scavenging rate was lower in Tsujido than in Beijing.

In case of light precipitation events, there are many cases where the scavenging rate is negative, suggesting that this is due to secondary production of aerosols, hygroscopic growth on rainy days, or advection and replacement of air masses. The increase in the composited PM$_{2.5}$ concentration after precipitation
suggests that the aerosols that escaped deposition are rolled up from the ground after precipitation. This can occur when aerosols are supplied by the wind or by secondary production.

Considering that variables other than precipitation may also affect PM$_{2.5}$ concentrations, we conducted a composite analysis of wind speed for 12 h before and after precipitation at the same observation point as precipitation (figure not shown). However, no significant difference was found before and after the onset of precipitation. Therefore, the result of the analysis for wind speed supports that wet deposition plays an important role in the rapid decrease of PM$_{2.5}$ around the onset of precipitation.

The composite analysis showed that the average of PM$_{2.5}$ concentration recovered to approximately 81.20% of the value 12 h before the onset of precipitation. The PM$_{2.5}$ concentration stopped decreasing 3 h after the onset of precipitation. Whereas we need to extend the analysis period to see completely recover the PM$_{2.5}$ concentration, the results suggest the time scale of wet deposition is approximately a couple of hours to several hours.

This study analyzed the observed data of surface PM$_{2.5}$ concentration and precipitation at a single-point and showed a significant decrease in PM$_{2.5}$ concentration due to precipitation, which would be due to wet deposition. However, various detailed processes affect the surface PM$_{2.5}$ concentration, and these effects have not been discarded in the present analyses. In order to further elucidate the detailed mechanism on the decrease in surface PM$_{2.5}$ concentration, it is necessary to directly measure aerosol particle size distribution.

5 | CONCLUSION

In this study, we analyzed the relationship between precipitation and PM$_{2.5}$ concentration at Tsujido, Kanagawa, Japan, for the two and a half years from October 1, 2017 to March 31, 2020. The PM$_{2.5}$ concentration is low as precipitation is high. The composite analysis for all the events revealed that the PM$_{2.5}$ concentration decreased by 20.99% from 1 h before to 1 h after the onset of precipitation. A paired t-test showed that the difference in the PM$_{2.5}$ concentration around the precipitation onset was statistically significant compared with the difference for 2 h before the onset of precipitation. The PM$_{2.5}$ concentrations decreased over 8 h period from 5 h (−5 h) before to 3 h (+3 h) after the onset of precipitation and then gradually increased afterward. In particular, the PM$_{2.5}$ concentration was drastically decreased at the onset of the precipitation. As the precipitation intensity increased, the PM$_{2.5}$ scavenging rate ($\Delta C$) increased, but the scavenging rate ($\Delta C$) for each intensity in Tsujido was lower for all intensity categories than in Beijing, where there is a pollution problem. The results based on observation data suggest that the deposition process plays a key role in reducing the PM$_{2.5}$ concentration at the surface.

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AUTHOR CONTRIBUTIONS

Risako Fujino: Conceptualization; data curation; formal analysis; investigation; methodology; visualization; writing – original draft. Yoshiaki Miyamoto: Conceptualization; funding acquisition; project administration; supervision; writing – review and editing.

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