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Relationships between Aerosol and Raindrop Size Distributions during Rainfall Period (Changma) in Jeju Island, Korea

Woonseon Jung [1], Hyun Min Sung [1], Cheol-Hwan You [2,*], Hyeon-Joon Kim [3], Sung-Ho Suh [4], Dong-In Lee [2] and Ki-Ho Chang [1]

1 National Institute of Meteorological Sciences, 33, Seohobuk-ro, Seoqwipo, Seogwipo-si 63568, Korea; wsjung01@korea.kr (W.J.); sunghm122@korea.kr (H.M.S.); khchang@korea.kr (K.-H.C.)
2 Atmospheric Environmental Research Institute (AERI), Pukyong National University (PKNU), 45, Yongso-ro, Nam-gu, Busan 48513, Korea; leedi@pknu.ac.kr
3 Department of Civil and Environmental Engineering, College of Engineering, Chung-Ang University, 84, Heukseok-ro, Dongjak-gu, Seoul 06974, Korea; hjkim22@cau.ac.kr
4 NARO Space Center, Korea Aerospace Research Institute (KARI), 508, Haban-ro, Bongrae-myeon, Daejeon 59571, Korea; suhsh@kari.re.kr
* Correspondence: youch@pknu.ac.kr

Abstract: To investigate the variation in aerosol particles removed by rainfall, we determined the relationship between aerosols and raindrops observed on Jeju Island, Korea, during a heavy rainfall period (Changma) from 1–15 July 2012. Various instruments, including an aerodynamic particle sizer, rain gauge, and disdrometer, were installed at the Gosan meteorological observation site on Jeju Island. During the Changma period, precipitation between 0.7 and 25.4 mm per day was recorded, and large variations in aerosol and raindrop size distributions were observed. Aerosol removal by precipitation was indicated, and its scavenging effect was confirmed from the results. Three major parameters (Brownian diffusion, interception, and impaction) described the collision efficiency based on aerosol and raindrop size distributions. The variations in the scavenging coefficient and below-cloud scavenging rate produced similar results for the accumulated rain amount. Therefore, these field observations explained the relationship between aerosol and raindrop size distributions.

Keywords: changma; aerosol; rain; size distribution

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1. Introduction

The size of aerosol particles varies widely between 0.001 and 100 μm [1], and these particles play important roles in cloud condensation, precipitation, and climate change [2]. Atmospheric aerosols are scavenged by rainfall, and their size distributions are largely affected by variations in the dispersed raindrop size. The size distributions of aerosols and raindrops are often explained in terms of cloud and precipitation formation and have been the subject of several studies [3–5]. Additionally, theoretical models that include scavenging coefficients have been formulated [6–13]. However, few experiments using direct observations have been performed [14,15]. In previous modeling research, the Marshall–Palmer and Krigian–Mazin raindrop distributions were adopted for generalized raindrop distributions using the moments method. However, raindrops with small diameters collect aerosols in narrow distributions more efficiently [10]. Moreover, the precipitation scavenging effect decreased below 0.6 μm and was effective above 3.5 μm [16]. The scavenging rates of weak, moderate, and strong rainfalls were 5.1%, 38.5%, and 50.6%, respectively, and the scavenging effect of weak rainfall was influenced by the rainfall duration and wind speed [17]. A numerical model was used to calculate the scavenging coefficients according to aerosol origins, such as continents, rural areas, oceans, and cities [18]. This model found differences between the coefficients by comparing those derived from theory, observations, and other models [19]. Below-cloud scavenging mainly affects coarse particles (over 3 μm [13] or 5 μm [12]). Additionally, heavy rain (about 10 mm/h) results in high below-cloud scavenging efficiency. The minimum size for efficient collection increases with decreasing fall velocity and packing density [9]. Thus, the collection efficiency is a function of the terminal velocity. The most influential factor for the scavenging effect is the increase in aerosol particle size with raindrop diameter by the Brownian diffusion and interception processes [11]. Sensitivity tests have been performed in urban, rural, and desert environments [6] and have considered the temperature differences between the air and raindrop surfaces [7]. Charge is also important for determining the most influential particle size [8]. Despite various modeling studies, there is still a gap between the measured and calculated scavenging coefficients. Furthermore, observational research on the relationships between aerosol and raindrop size distributions is rare. Therefore, to understand the characteristics and underlying mechanical processes, basic observational studies on variations in aerosols removed by rainfall are required.

The Changma period (generally, from June to July) is an important rainy season in Korea and is part of the Asian–Pacific summer monsoon [20]. The Changma frontal precipitation system is accompanied by heavy rainfall, strong winds, and humidity [21], with the maximum long-term rainfall occurring over the southern Korean Peninsula [22]. Additionally, the indirect effects of aerosols during the Changma period differ compared to that during dry seasons [23]. In the Indian summer monsoon region, there are positive correlations between aerosol loading and cloud properties, as well as rainfall [24]. More aerosols can increase clouds over specific regions under ample humidity during the monsoon [25].

Therefore, the purpose of this study is to understand and statistically quantify the relationship between aerosol and raindrop size distributions, as observed on Jeju Island, Korea, in Changma frontal systems during heavy rainfall.

2. Data and Method

2.1. Data

Jeju Island is an elliptical island located in the southern part of the Korean Peninsula, with an area of 1833.2 km^2, 73 km in the east–west direction, and 31 km in the north–south direction. To examine the relationship between aerosol and raindrop size distributions, various instruments were installed at the Korea Global Atmosphere Watch Center (33.29° N, 126.16° E) on Jeju Island, Korea, during the field observation period. These included an aerodynamic particle sizer (APS), a rain gauge from the Korea Global Atmosphere Watch Center of the Korea Meteorological Administration, and particle size and velocity instrument (PARSIVEL) from Pukyong National University.

The APS spectrometer (TSI, Shoreview, MN, USA, Model: 3221), which provides accurate size distributions of aerosol particles with aerodynamic diameters from 0.5 to 20 μm using 52 channels, was used to determine aerosol size distributions with 3 s time resolution. The PARSIVEL (OTT, Kempten, Germany, Model: 2), which measures raindrop sizes ranging from 0.2 to 25 mm using 32 channels and fall velocities from 0.2 to 20 m/s using 32 classes, was used to determine raindrop size distribution with 3 s time resolution. The disdrometer (OTT, Kempten, Germany, Mode: 2) was used to identify eight different precipitation types (drizzle, mixed drizzle/rain, rain, mixed rain/snow, snow, snow grains, freezing rain, and hail). The rain gauge measured rainfall amounts with a precision of 0.1 mm.

Four precipitation cases during the Changma period from 1–15 July 2012, were selected and analyzed (Table 1). In each case, the Changma frontal system was located near Jeju Island, and the cases of this study had similar humid meteorological conditions. The humidity, wind speed, and wind direction of Cases 1–4 were 92.0 ± 2.3%, 5.4 ± 2.2 m/s, and southerly wind. Thus, Cases 1–4 showed similar meteorological environment conditions. Additionally, the accumulated rainfall amounts gradually increased from 0.7 (Case 1) to 25.4 mm (Case 4) per day. It is known that rainfall amounts vary greatly between the onset and peak during the rainy season in Korea [26].

Table 1. Selected date and rain amount.

Case	Date	Rain Amount (mm)
Case 1	15 July 2012	0.7
Case 2	14 July 2012	3.4
Case 3	13 July 2012	13.5
Case 4	11 July 2012	25.4

2.2. Method

In this study, the correlation and scavenging coefficients were calculated for each aerosol and raindrop size to understand the relationship between their distributions.

Correlation coefficients are widely used in statistical analyses to indicate the fit of a linear relationship between two variables and are defined as the covariance of the variables divided by the product of their standard deviations [27,28]. The formula for the Pearson correlation coefficient r is as follows:

$$r = \frac{\sum_{i=1}^{n}\left(X_i - \overline{X}\right)\left(Y_i - \overline{Y}\right)}{\sqrt{\sum_{i=1}^{n}\left(X_i - \overline{X}\right)^2}\sqrt{\sum_{i=1}^{n}\left(Y_i - \overline{Y}\right)^2}}, \quad (1)$$

where n is the number of observations, and X and Y are the aerosol and raindrop size distributions, respectively.

The scavenging coefficient is a parameterization for the rate of the loss of gases or aerosols from the atmosphere by their incorporation into larger drops. Scavenging coefficients are generally a function of time, location, rainfall characteristics, and aerosol size distribution. The rate of below-cloud scavenging is often referred to as the "washout" rate. The scavenging coefficient $\Lambda(d_p)$ and the below-cloud scavenging rate $W_{bc}(d_p)$ are derived as follows [1]:

$$\Lambda(d_p) = \int_0^\infty \frac{\pi}{4} {D_p}^2 U_t(D_p) \mathrm{E}(D_p, d_p) N(D_p) \mathrm{d}D_p, \quad (2)$$

$$W_{bc}(d_p) = n(d_p)\mathrm{d}d_p \int_0^\infty \frac{\pi}{4} {D_p}^2 U_t(D_p) \mathrm{E}(D_p, d_p) N(D_p) \mathrm{d}D_p, \quad (3)$$

where D_p and d_p are the raindrop and aerosol diameters, respectively, $N(D_p)$ and $n(d_p)$ are the number distributions for raindrops and aerosols, respectively, $U_t(D_p)$ is the fall-velocity of the raindrops, and $\mathrm{E}(D_p, d_p)$ is the collision efficiency. The fall-velocity, $U_t(D_p)$, is determined using an empirical formula [29]. The collision efficiency, $\mathrm{E}(D_p, d_p)$, is defined as the ratio of the total number of collisions to the total number of particles in the droplet's effective cross-sectional area. The collision efficiency, $\mathrm{E}(D_p, d_p)$ is expressed as follows [1]:

$$\mathrm{E}(D_p,\ d_p) = \frac{4}{R_e S_c}\left[1 + 0.4\, {R_e}^{\frac{1}{2}} {S_c}^{\frac{1}{3}} + 0.16\, {R_e}^{\frac{1}{2}} {S_c}^{\frac{1}{2}}\right] + 4\varnothing\left[\omega^{-1} + \left(1 + 2{R_e}^{\frac{1}{2}}\right)\varnothing\right] + \left(\frac{S_t - S^*}{S_t - S^* + \frac{2}{3}}\right)^{\frac{3}{2}}, \quad (4)$$

where R_e is the Reynolds number of the raindrop based on its radius, S_c is the Schmidt number of the collected particle, $\varnothing$ and ω are the ratios of the diameters and viscosities, respectively, and S_t and S^* are the Stokes number and critical Stokes number of the collected particle, respectively. The first term is the contribution from Brownian diffusion, the second expresses interception, and the third represents impaction. The scavenging coefficient is calculated from the aerosol and raindrop size distributions using Equations (2)–(4).

3. Results and Discussion

3.1. Aerosol and Raindrop Size Distributions

Figure 1 shows the particle number concentrations and diameters of raindrops and aerosol particles. The raindrop number concentration decreased with an increase in diameter (Figure 1a). For the low rainfall amount (Case 1), large raindrops showed low or non-existent number concentrations. Comparatively, the number concentration of large-diameter raindrops is high for the heavy rainfall case (Case 4). This change in number concentrations occurs at a critical rainfall amount of approximately 1 mm, as in a previous study on the relationship between raindrop number concentration and diameter [30]. The aerosol number concentration also decreased overall for larger aerosol diameters (Figure 1b). The aerosol number concentration for low rainfall amounts was small, whereas it was large for high rainfall. Overall, the distribution of aerosol and raindrop number concentrations showed similar patterns.

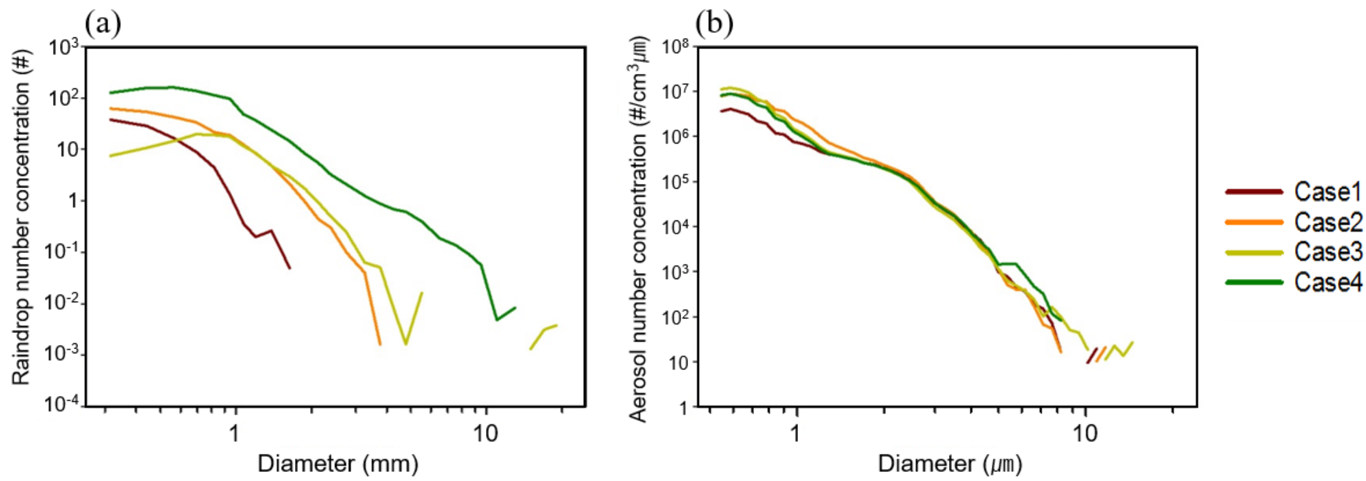

Figure 1. The number concentration of (**a**) raindrop and (**b**) aerosol in Cases 1 to 4.

To further examine this relationship, the correlation coefficient was calculated for each recorded channel, which showed that the aerosols were more scavenged by raindrops in the larger channels. Although the correlation coefficients showed small values, their distribution clearly indicated a relationship between the distributions (Figure 2). Case 1 (the lowest rainfall amount) and Case 4 (the highest rainfall amount) showed a negative correlation coefficient for small aerosol particles (less than 1.037 μm) and positive values for large channels (Figure 2a). In contrast, Case 2 (Figure 2b) and Case 3 (Figure 2c) showed positive correlation coefficients for aerosol particles smaller than 1.382 and 0.898 μm, respectively, and negative values for large channels. The correlation coefficients were affected by the characteristics of each rainfall event. Therefore, their trends were diverse. However, the critical aerosol diameter of approximately 1 μm played an important role in precipitation, as shown in Figure 2.

3.2. Below-Cloud Scavenging Processes with Aerosol Characterization

Wet deposition refers to natural processes that scavenge aerosols by precipitation and deliver them to the ground [1]. The definitions before and after precipitation were classified before and after passing through the observation site of the precipitation system. The aerosol number concentration decreased after precipitation throughout every size range, and higher rain amounts caused a greater decrease (Figure 3), thus, suggesting a scavenging effect.

To estimate the scavenging coefficient for below-cloud scavenging processes using aerosol and raindrop size distributions from each channel, the collision efficiency, which is a key parameter in scavenging processes, was calculated. The collision efficiency is determined by the Brownian diffusion, interception, and impaction parameters (Figure 4). The collision efficiency increased with decreasing raindrop diameter and increasing aerosol diameter. In particular, higher values (approximately 1) occurred for raindrop diameters below 1 mm and aerosol diameters over 10 μm (Figure 4a). Brownian diffusion was marginally affected by aerosol diameters less than 1 μm, and raindrop sizes less than 0.5 mm (Figure 4b). However, interception and impaction were affected by large aerosol diameters (over 10 µm). Interception was affected by aerosol diameters over 1 µm, and raindrop sizes less than 1 mm (Figure 4c). Impaction increased with decreasing raindrop diameter and increasing aerosol diameter and showed higher values (approximately 1) for raindrop diameters of approximately 1 mm and aerosol diameters greater than 10 µm (Figure 4d). Notably, the trends in impaction and collision efficiency were almost identical.

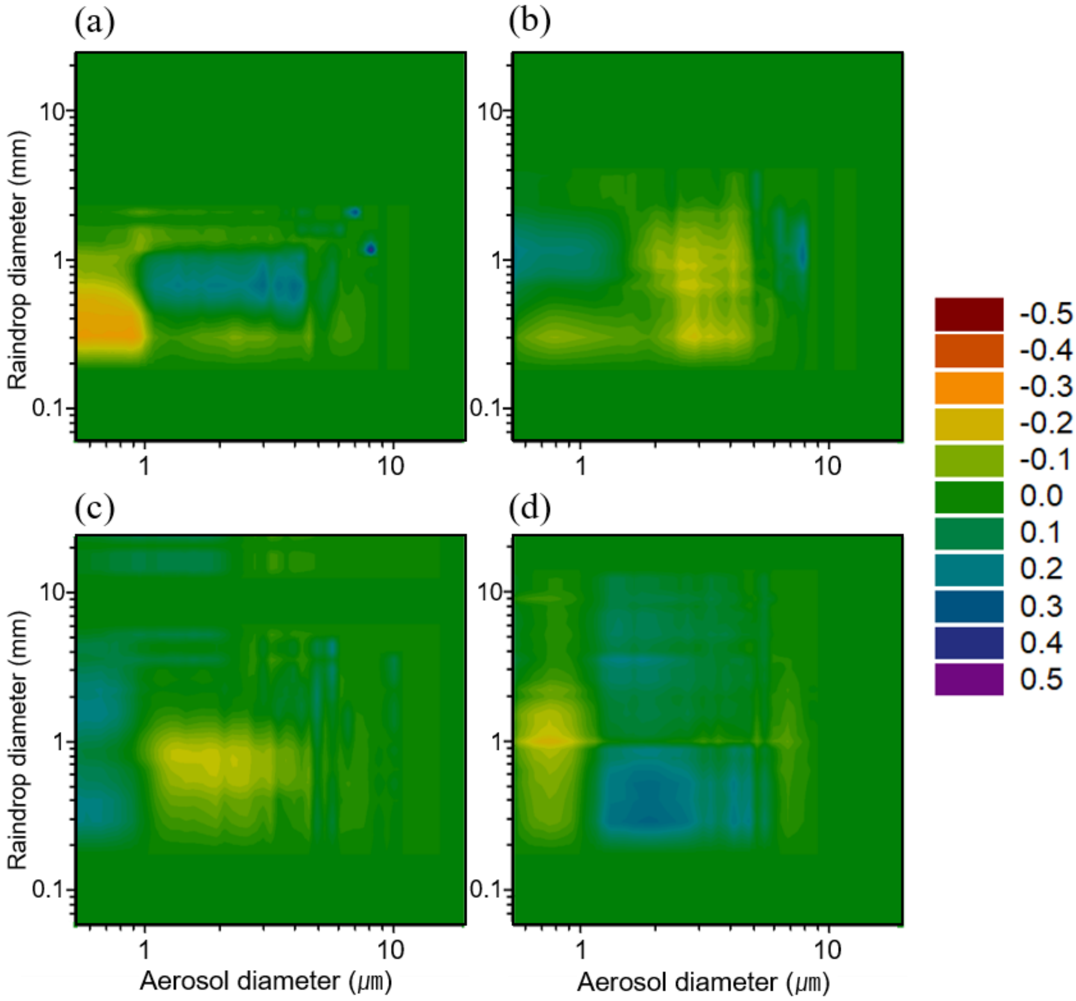

Figure 2. Distribution of correlation coefficient between aerosol and raindrop diameter from (**a**–**d**) (cases 1 to 4).

Therefore, the results indicate that the collision efficiency may contribute to the impaction of a particle captured by a raindrop. The variations in collision efficiency, Brownian diffusion, interception, and impaction with aerosol diameter were calculated for several raindrop diameters: 0.187, 0.312, 0.562, 1.062, 2.75, 5.50, 11.0, and 24.5 mm (Figure 5). However, it is mechanically impossible to exceed 10 mm, as the drop breaks at that diameter [31]. Therefore, raindrops less than 10 mm were considered in this study. The collision efficiency value steadily increased for aerosol diameters less than 3.5 µm, and sharply increased for diameters greater than 3.5 and up to 6 µm (Figure 5a). The aerosol size threshold required to increase collision efficiency was small when raindrops were less than 1 mm, whereas it was large when raindrops increased to over 1 mm. A sudden drop in collision efficiency occurred at a raindrop diameter of 2 mm. Moreover, Brownian diffusion decreased, and interception increased with increasing aerosol size (Figure 5b,c). Additionally, the impaction for aerosol diameters over 3.5 µm was calculated, and a change occurred at a raindrop diameter of 2 mm (Figure 5d). Overall, the results showed that the collision efficiency for aerosol diameters 0.5–20 µm was affected by the impaction parameter for aerosol diameters over 3.5 µm. Notably, the trends in interception, impaction, and collision efficiency were almost identical.

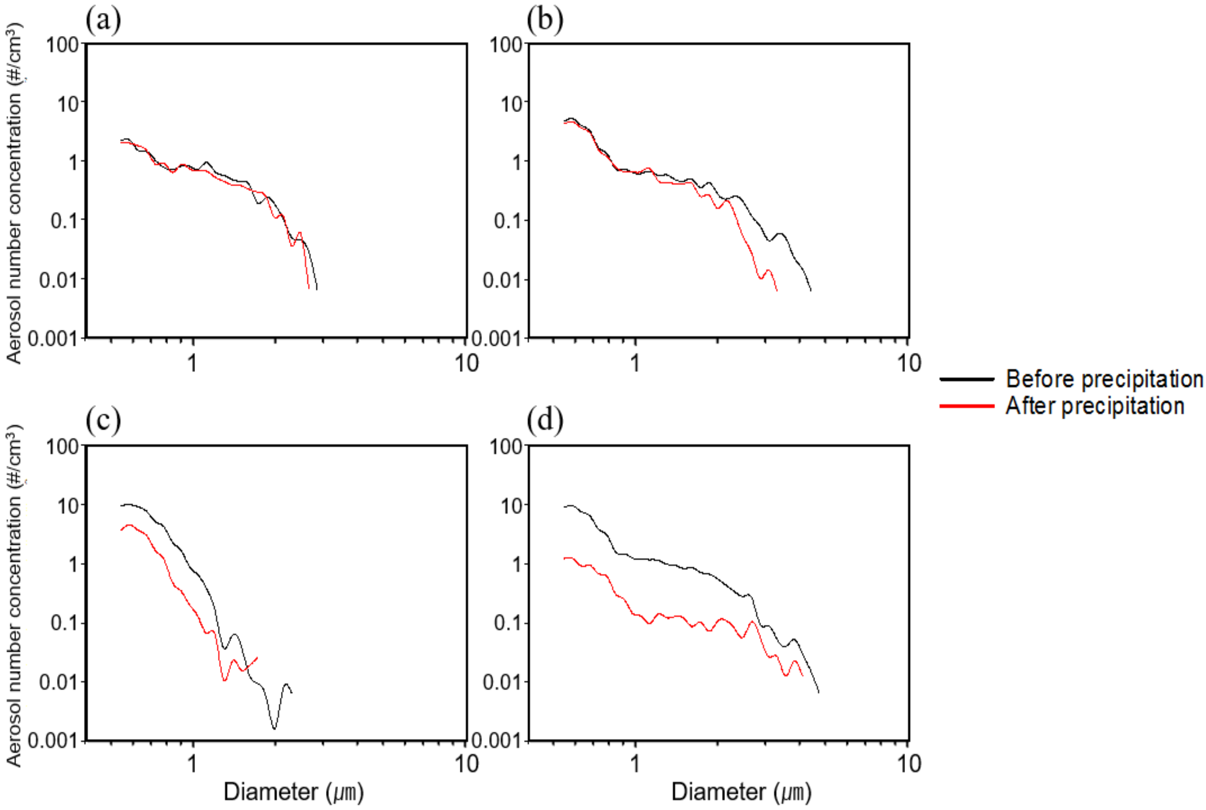

Figure 3. Observed aerosol number concentration before and after precipitation from (**a**–**d**) (cases 1 to 4).

3.3. Scavenging Coefficient and Below-Cloud Scavenging Rate

The scavenging effect of precipitation and interception and impaction processes affecting collision efficiency were estimated. The scavenging coefficient gradually increased with the rainfall amount (Figure 6a). In all cases, the coefficient increased smoothly for small-diameter aerosols (less than 3.5 µm), and sharply increased for large-diameter aerosols (over 3.5 µm), which was a similar trend to the impaction distribution. According to Equation (2), the high rainfall case showed a high scavenging value because of the large raindrop size, fall velocity, collision efficiency, and raindrop number concentration. The scavenging rate is particularly affected by collision efficiency (Figure 6a) around the threshold value. The collision efficiency sharply increased at an aerosol diameter of 3.5 µm, indicating that particles over 3.5 µm have the scavenging capacity. The below-cloud scavenging rate did not correspond to rainfall amounts (Figure 6b) because of the aerosol number concentration in Equation (3). The below-cloud scavenging rate of aerosols decreased smoothly in all cases but sharply increased for aerosols over 3.5 µm in diameter. It was the most efficient at removing particles with diameters of approximately 0.6 µm and greater than 3.5 µm [16].

The black lines in Figure 7a,b indicate the mean scavenging coefficient and mean below-cloud scavenging rate per day, respectively, and bar graphs show the daily accumulated rainfall amount. Here, the scavenging coefficient and below-cloud scavenging rate increased as the rain amount increased. Cases 1 and 4 showed significant variations between the two factors because of the large difference in rain amount. This indicates that the higher the amount of rain, the greater the variation between the two factors. Furthermore, Cases 2 and 3 showed a similar pattern. Therefore, we investigated the time series of the scavenging effect to determine the cause, except precipitation.

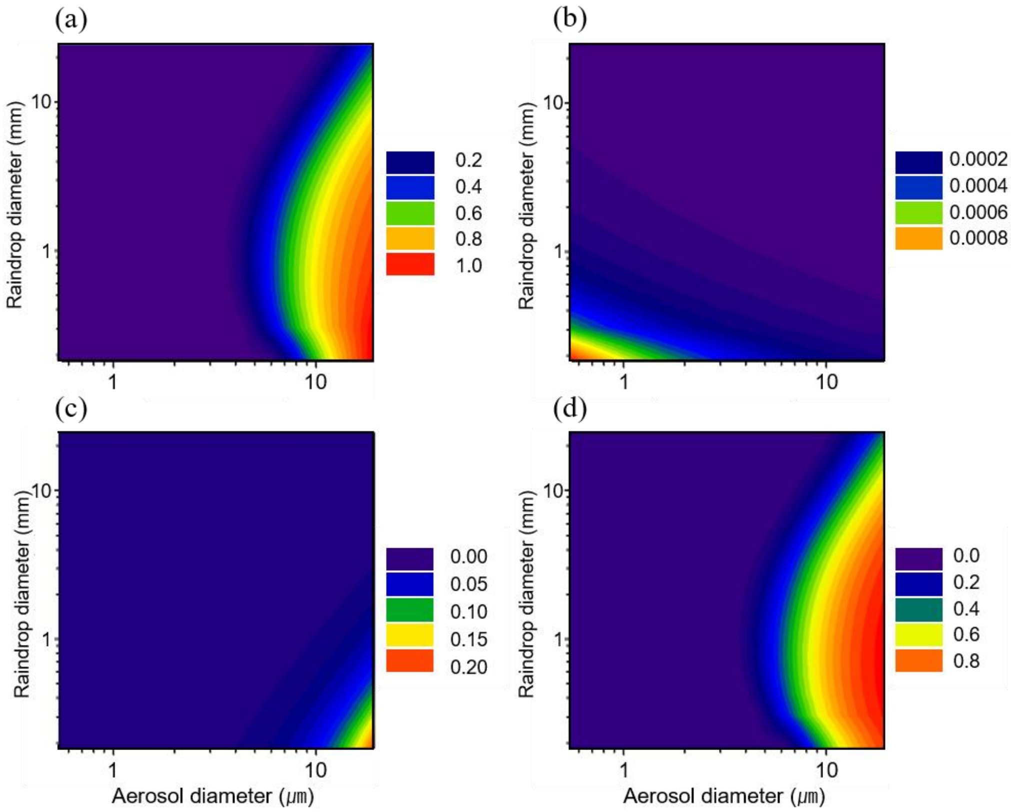

Figure 4. Distribution of (**a**) collision efficiency, (**b**) Brownian diffusion, (**c**) interception, and (**d**) impaction as a function of aerosol and raindrop diameters.

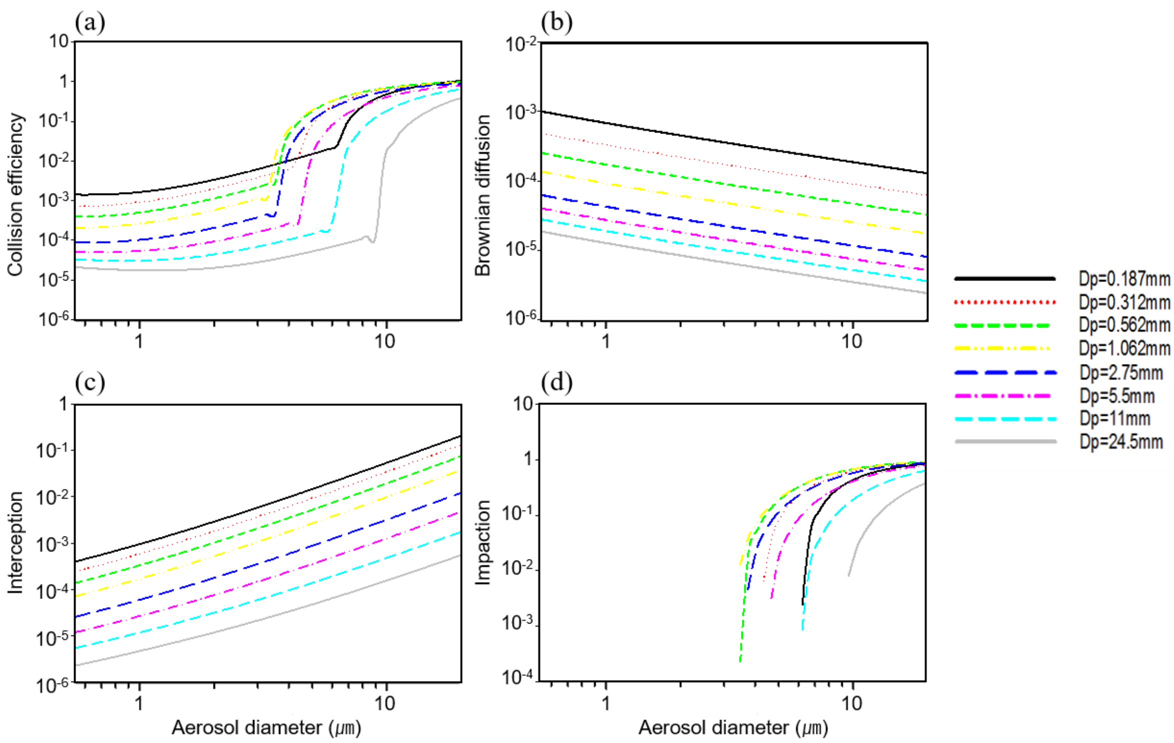

Figure 5. Variation of (**a**) collision efficiency, (**b**) Brownian diffusion, (**c**) interception, and (**d**) impaction versus aerosol diameter for several raindrops.

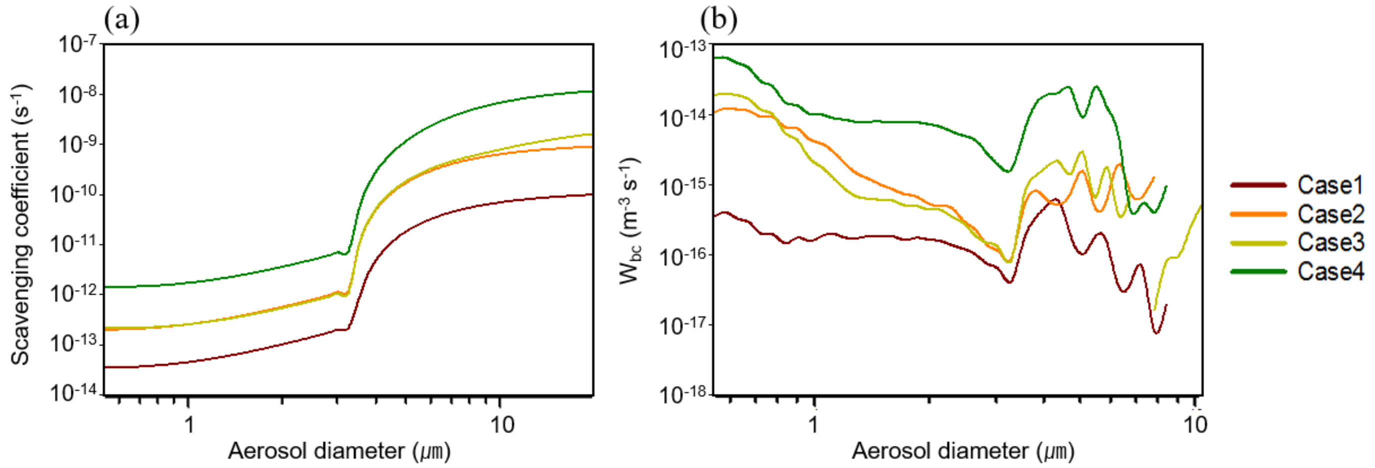

Figure 6. Variation of (**a**) scavenging coefficient and (**b**) below-cloud scavenging rate as a function of aerosol diameter in Cases 1 to 4.

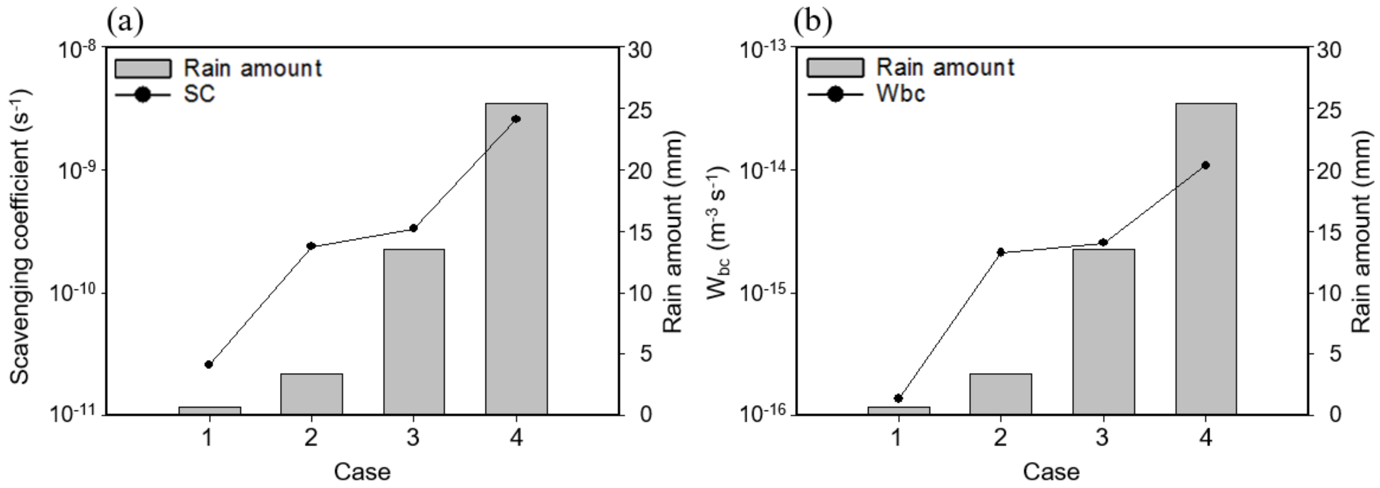

Figure 7. Comparison of the (**a**) mean scavenging coefficient and (**b**) mean below-cloud scavenging rate and rain amount in Cases 1 to 4.

The time series of the scavenging coefficients for several aerosol diameters (0.542, 1.037, 2.129, 5.048, 10.37, 15.96, and 19.81 µm) and the mean scavenging coefficient were also similar to the rain rates in all cases (Figure 8). The effect of the scavenging coefficient depends on the aerosol size distribution. Moreover, large aerosol particles can perform scavenging processes, as described above. The time series of the below-cloud scavenging rate for several aerosol diameters (0.542, 1.037, 2.129, 5.048, 10.37, 15.96, and 19.81 µm) and the mean below-cloud scavenging rate were also similar to the rain rates in all cases (Figure 9). The effect of the below-cloud scavenging rate was also dependent on the aerosol size distribution, as it is affected by the aerosol number concentrations. The pattern in the mean below-cloud scavenging rates was similar to that in the scavenging coefficients. In Cases 2 and 3, the scavenging coefficient and below-cloud scavenging rate showed large values during high rainfall periods, but the trend was unclear.

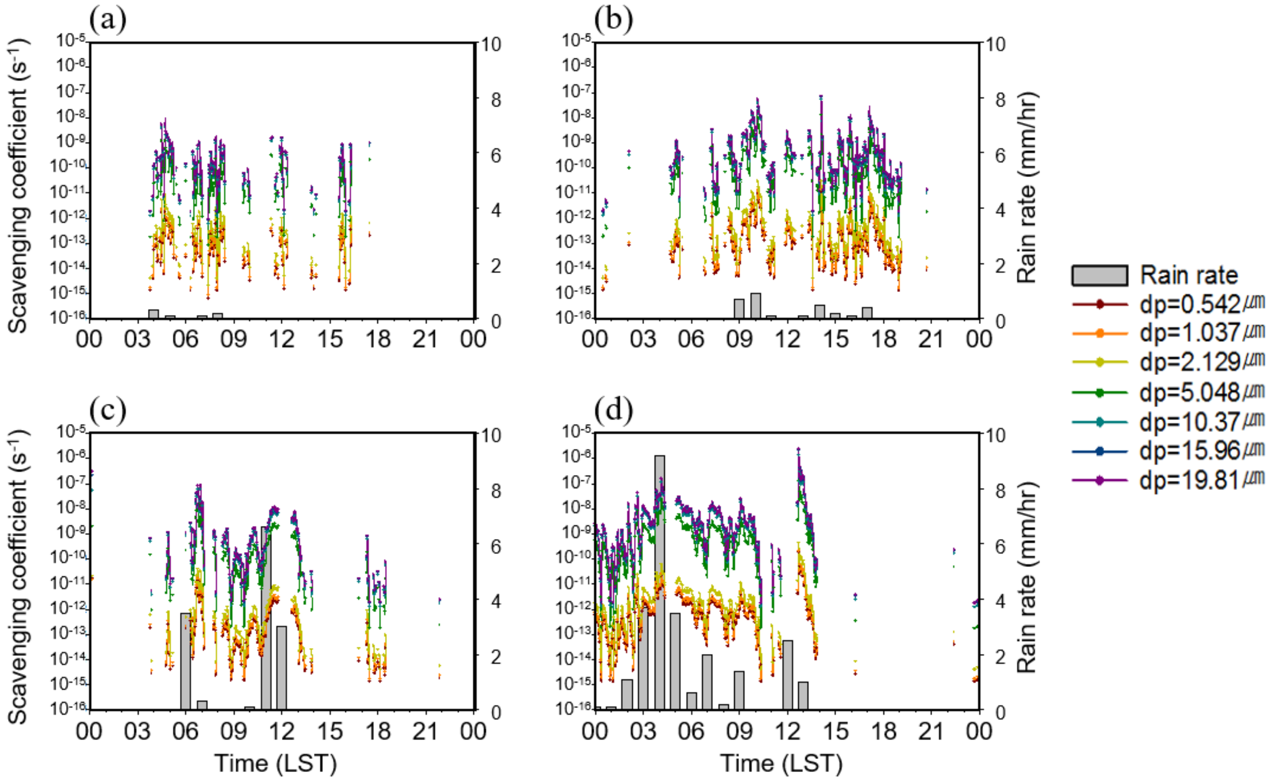

Figure 8. Time series of the scavenging coefficient and rain rate for several aerosol diameters from (**a**–**d**) (cases 1 to 4).

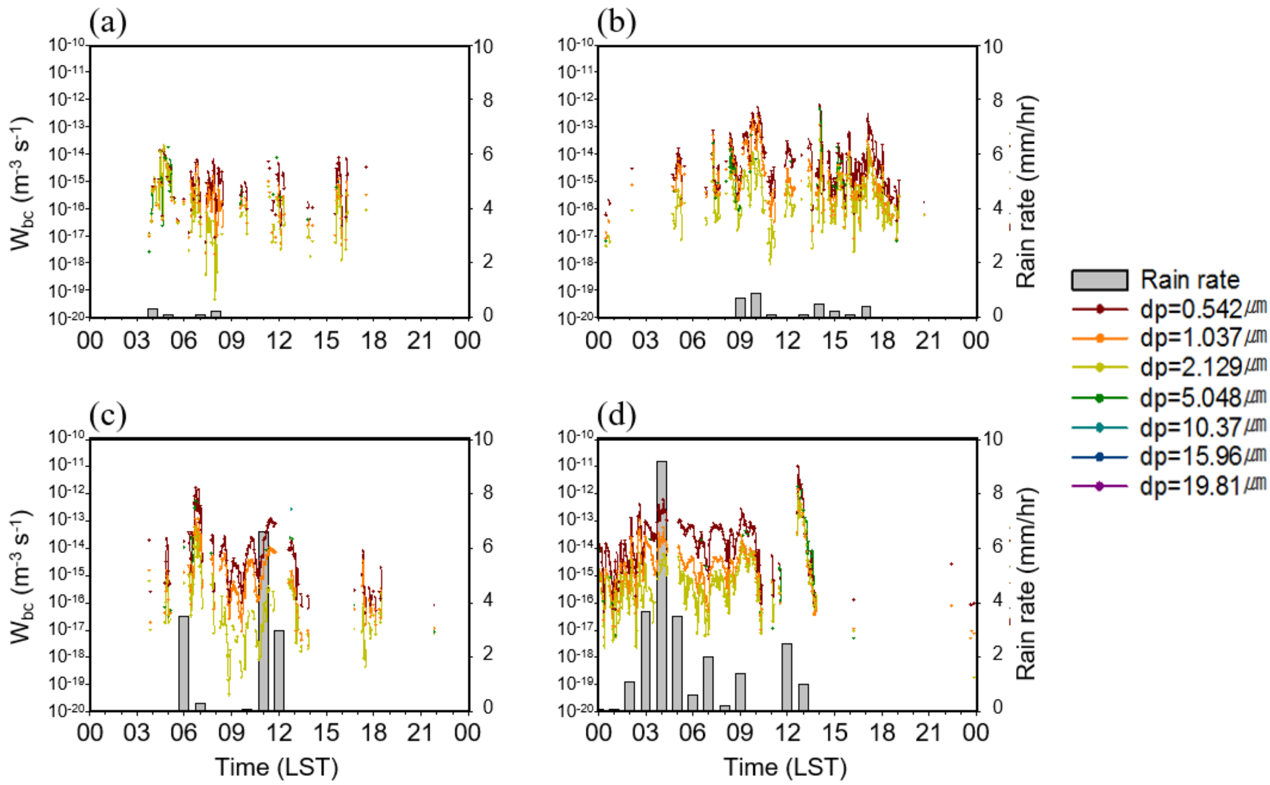

Figure 9. Time series of the below-cloud scavenging rate and rain rate for several aerosol diameters from (**a**–**d**) (cases 1 to 4).

4. Summary and Conclusions

To investigate the relationship between aerosol and raindrop size distributions, we performed observational experiments in Gosan, Jeju Island, Korea, during the Changma period from 1–15 July 2012. Rainfall amounts between 0.7 and 25.4 mm were recorded in four cases.

Overall, the raindrop and aerosol number concentrations decreased for large diameters. Although the correlation coefficients were low, their distribution clearly indicated a relationship between aerosol and raindrop size distributions. Although the correlation coefficient for each case represents diverse trends, a critical aerosol diameter of 1 μm played an important role in precipitation. The collision efficiency was determined by Brownian diffusion, interception, and impaction parameters. Collision efficiency increased with decreasing raindrop diameter and increasing aerosol diameter and showed higher values (approximately 1) for raindrop diameters less than 1 mm and aerosol diameters greater than 10 μm. The trends in the collision efficiency and impaction were almost identical, which reflects the contribution of impaction to the collision efficiency for a particle captured by a raindrop. Similar trends were observed between scavenging coefficients and rainfall amounts, but below-cloud scavenging rates showed different trends depending on rainfall amounts. Therefore, the scavenging process was influenced by the diffusion parameter in low rainfall cases (Figure 10). Conversely, scavenging was mainly affected by interception and impaction parameters in high rainfall cases (Figure 10). In humid environments (such as the Changma period) with over 92% relative humidity, the major scavenging mechanism appears to depend on the amount of rain. Under low rainfall conditions, the diffusion process plays a primary role in the scavenging effect. However, the interception and impaction processes have a significant effect on scavenging in the case of heavy rainfall.

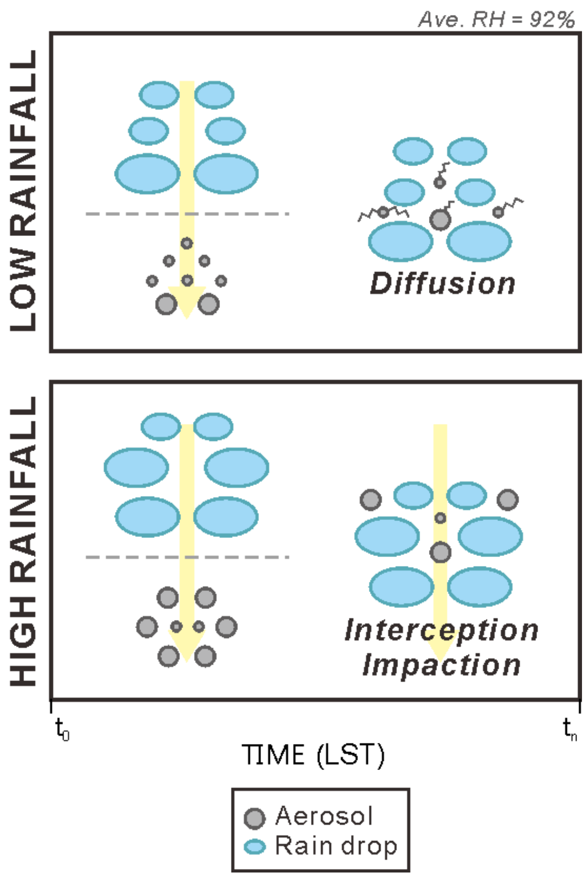

Figure 10. Schematic diagram of relationship between aerosol and raindrop particles. The gray and blue circles indicate aerosol and raindrop particles, respectively. Droplet number and size indicate the number concentration and drop size, respectively.

This study provides meaningful observational results on the relationship between aerosol and raindrop size distributions during natural events. The results show that Brownian diffusion, interception, and impaction are the primary mechanisms without considering phoresis. This study paves the way for understanding the mechanisms affecting aerosol and raindrop distributions in detail. For this, understanding the scavenging effect of electrical charge on small particles, field observations, and statistical analyses are warranted for various meteorological phenomena and rainfall events.

Author Contributions: Conceptualization, W.J.; methodology, W.J.; software, W.J.; validation, W.J., H.M.S., C.-H.Y., H.-J.K. and S.-H.S.; formal analysis, W.J.; investigation, W.J., H.M.S., C.-H.Y., H.-J.K. and S.-H.S.; data curation, W.J. and D.-I.L.; writing—original draft preparation, W.J.; writing—review and editing, all authors; funding acquisition, W.J. and K.-H.C.; All authors have read and agreed to the published version of the manuscript.

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