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Scavenging of aerosol particles by large water drops
3. Washout coefficients, half-lives, and rainfall depths

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Abstract. Using the average values of collection efficiencies obtained from our experiments [*Pranesha and Kamra*, 1996, this issue] washout coefficients for the drops in the diameter (D) range $3.6 \le D \le 5.0$ mm collecting micron-sized aerosol particles have been calculated when the drops are neutral, charged, or falling in an electric field. Compared with the neutral case, the values of washout coefficients are higher in both electrical cases, the increase being more pronounced for smaller particles. Washout coefficients show a maximum for a drop charge of 10^{-12} to 10^{-11} C. With an increase in electric field, the washout coefficients increase linearly for ~1-μm particles, show a maximum for ~4-μm particles, and change insignificantly for ~7-μm particles. Combining our experimental values of collection efficiencies with the theoretical collision efficiencies of *McGann and Jennings* [1991] for smaller drops, washout coefficients, half-lives, and rainfall depths have been computed for the raindrop size distribution extending from 0.1- to 5 mm-diameter. Results show that raindrops of diameter >1 mm contribute dominantly in removing particles of diameter 1-2 μm and their contributions increase with the rainfall rate. When the effect of the raindrops of diameter > 1 mm is included, the values of washout coefficient increase by about 2 orders of magnitude for particles of diameter 1-2 μm and by about 1order of magnitude for particles of diameter > 2 μm. It can be concluded from the estimates of rainfall depth that a heavy rainfall over a short duration is more efficient in removing the particles of diameter < 2.2 μm, whereas a lower rainfall spread over a longer duration is more efficient in removing the particles of diameter > 2.2 μm.

1. Introduction

In the atmosphere, the size of aerosol particles range over several orders of magnitude. Sizes of drizzle and raindrops also vary by more than an order of magnitude, and the raindrop size distribution changes with rainfall rate. Moreover, the raindrops in electrically active clouds are charged and fall under high electric fields. Since various physical mechanisms that are important in drop-particle interactions are strongly dependent on the size and charge of the drops and particles, they contribute to the scavenging process in various degrees. Because of the superimposition of various mechanisms, the removal of aerosol particles by precipitation scavenging in the atmosphere is complicated. The scavenging or washout coefficient gives a quantitative estimate of scavenging of aerosol particles. However, direct measurements of the scavenging coefficient in the atmosphere are difficult. Some estimates of scavenging coefficients reported in the literature [e.g., *Lai et al.*, 1978; *McGann and Jennings*, 1991] have been obtained using the experimentally measured or theoretically derived collision efficiencies. Such estimates help in better understanding the physics of the scavenging process. However, because of the nonvalidity of the assumption of sphericity in the case of large drops which undergo distortion and shape oscillations, the raindrop size distribution is generally terminated at rather small values of drop size in these theoretical studies. For example, *McGann and Jennings* [1991] terminate the drop size at 1-mm diameter, and *Lai et al.* [1978] terminated the drop size at 2-mm diameter. These upper limits for raindrop size seem to be appropriate for low rainfall rates. However, in heavy rainfalls from thunderclouds, a large number of drops exceed these sizes. In this paper we present the contributions of large raindrops to washout coefficients computed from the collection efficiency results of our laboratory experiments, reported earlier [*Pranesha and Kamra*, 1996, this issue] (hereinafter referred to as PK 1 and PK 2, respectively). We also examine the effect of electrical forces on washout coefficients for the cases when the drops are electrically charged and when the neutral drops fall through aerosol in the presence of a horizontal electric field. In addition, we present estimates of washout coefficients for the raindrop sizes from 0.1-mm to 5-mm diameter using our experimental values of collection efficiency for larger drops and the theoretical collision efficiency results of *McGann and Jennings* [1991] for smaller drops.

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2. Washout Coefficients

In discrete form the washout coefficient $\Lambda(d_p)$ for particles of diameter d_p can be written as

$$\Lambda(d_p) = \Sigma_j\, (\pi/4)\, D_j^2\, V_j\, E(D_j, d_p)\, N_j \tag{1}$$

where D_j is the diameter of jth drop, V_j is its terminal velocity, N_j is the number density of raindrops of diameter between D_j and $(D_j + \Delta D_j)$, and $E(D_j, d_p)$ is the collection efficiency of drops of diameter D_j for particles of diameter d_p. The raindrop number density N_j in a size range is computed from *Marshall and Palmer* [1948] size distribution of raindrops, and the terminal velocity V_j is computed from the formulas given by *Beard* [1976]. Equation (1) is integrated with discrete values of the quantities involved.

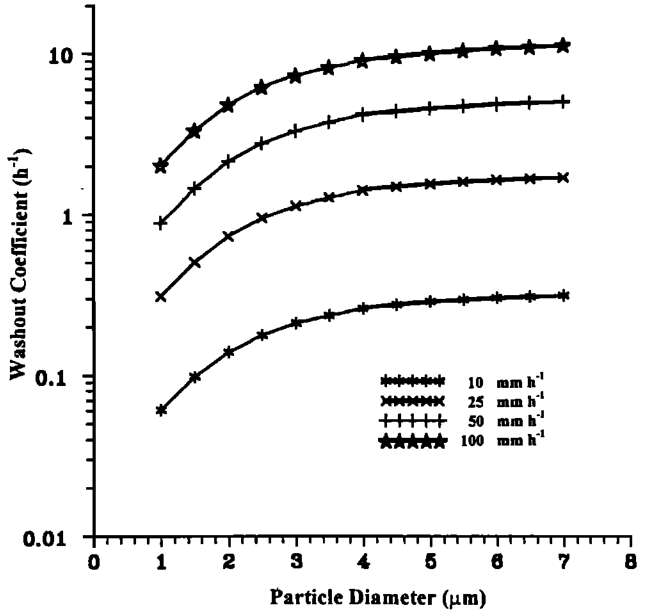

Figure 1. Washout coefficient for $3.6 \leq D \leq 5.0$ mm drops as a function of particle size for different rainfall rates.

Unlike in the work of PK 1 and PK 2 where each data point has been plotted, here we calculate the average of two to three data points for each drop-particle pair. Using these average collection efficiency values (reported by PK 1), we compute the washout coefficients for the part of raindrop size distribution $3.6 \leq D \leq 5$ mm for rainfall rates of 10, 25, 50, and 100 mm h^{-1} and present them in Figure 1. For a given rainfall rate, the washout coefficient increases rather rapidly as the particle size increases from 1 to 4 µm and then asymptotically settles down to almost a constant value for larger particles. To get an estimate of the contribution of drops of diameter 3.6-5 mm in scavenging the aerosol particles, we have computed the washout coefficients for raindrop size distribution extending from 0.1- to 5-mm diameter for the four rainfall rates mentioned above. In these calculations, in addition to our average experimental values of collection efficiency for 3.6- to 5-mm-diameter drops, we also use the theoretical collision efficiencies of *McGann and Jennings* [1991] for drops of diameter 0.1-1 mm at relative humidity of 50%. The collision efficiencies of 1- to 3-mm-diameter drops are obtained by extrapolating the values given in Figure 2 of *McGann and Jennings* [1991] by replotting them as a function of drop size. However, as noted by these authors, the extrapolated values show that the collision efficiencies tend to become independent of drop size for drops greater than 0.4-mm diameter and particles greater than 4 µm. Even though no experimental results are available in this size range, these values roughly agree with the extrapolated collision efficiencies used by *Horn et al.* [1988] for 2.5-mm drops. Figure 2 shows the variation of these washout coefficients with the particle size for four rainfall rates. It also shows the results of *McGann and Jennings* [1991] for a rainfall rate of 10.2 mm h^{-1} both excluding and including the contributions due to extrapolated values of collision efficiencies. In both cases, the results show an increase in washout coefficients with the increase in particle size. The washout coefficients presented by *Seinfeld* [1986] based on the results of *Slinn* [1983] are normalized with respect to rainfall rate. These washout coefficients are computed for polydisperse aerosol particles collected by monodisperse raindrops. For the case of monodisperse particles, their washout coefficients show a similar trend. However, when the contributions of raindrops of 0.1-3 mm in diameter are calculated and added to our new results, the washout coefficient for particles < 2-µm diameter increases by about 2 orders of magnitude, and the washout coefficient for particles > 2-µm diameter increases by about 1 order of magnitude. For particles of diameter 1 to 2 µm on the right side of Greenfield gap, the collection efficiency values increase sharply with the increasing particle size. For particles larger than 2-µm diameter the collection efficiency increases at a slower rate and settles down at values less than unity. This results in the higher increase in washout coefficient for particles of diameter < 2 µm when 1- to 5-mm-diameter drops are included.

3. Half-Life and Rainfall Depth

At any time t during a scavenging event, the concentration $N(d_p,t)$ of particles of diameter d_p is related to initial concentration $N(d_p,0)$ by the relation

$$N(d_p,t) = N(d_p,0)\exp(-\Lambda(d_p)t) \tag{2}$$

The half-life period corresponding to the scavenging of half the initial particle concentration is given by

$$t_{1/2} = \frac{0.693}{\Lambda(d_p)} \tag{3}$$

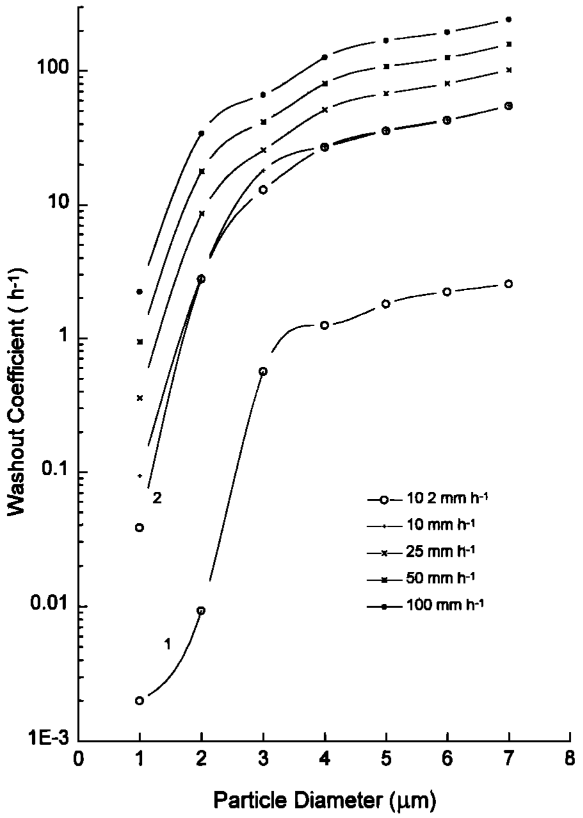

Figure 2. Washout coefficient for $0.1 \leq D \leq 5.0$ mm as a function of particle size for different rainfall rates. Curve 1 shows the results of *McGann and Jennings* [1991] for $0.1 \leq D \leq 1$ mm, and the curve 2 shows the results of McGann and Jennings plus a contribution from $1 \leq D \leq 3$ mm drops calculated from extrapolated collision efficiencies of McGann and Jennings.

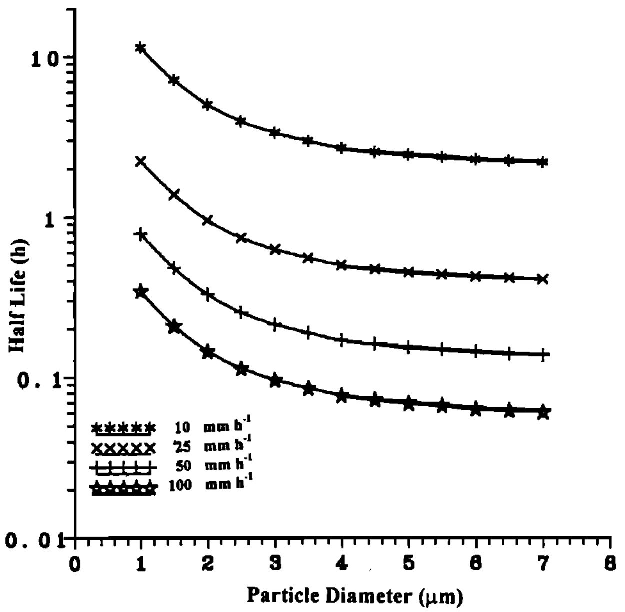

Figure 3. Half-life period versus particle size for scavenging by 3.6 ≤ D ≤ 5.0 mm drops as a function of rainfall rate.

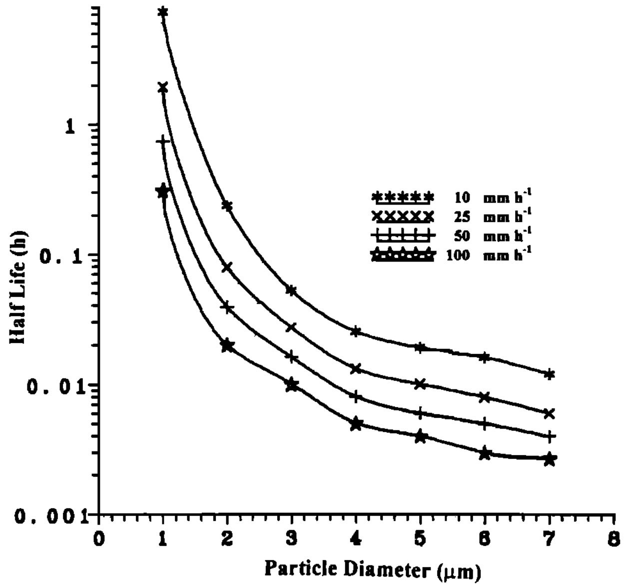

Figure 5. Half-life period versus particle size for scavenging by 0.1 ≤ D ≤ 5.0 mm drops as a function of rainfall rates.

and the corresponding rainfall depth is given by

$$h_{1/2} = R \times t_{1/2} \tag{4}$$

where R is the rainfall rate in millimeters per hour.

The half-lives and rainfall depths corresponding to washout coefficients calculated for the raindrop size distribution from 3.6- to 5-mm diameter (Figure 1) and for the raindrop size distribution from 0.1- to 5-mm diameter (Figure 2) are presented in Figures 3-6. Figure 5 shows that the half-life for a 1-μm-diameter particle is of the order of a few hours, indicating the lower efficiency of raindrops in collecting particles of diameter around 1 μm. The observation is in accordance with the well-recognized fact that the particles in the "Greenfield gap" are least affected by the scavenging process and have long residence times in the atmosphere. Our results show that the above conclusion is not much affected by the presence of large drops in the rainfall. Similar conclusions can be drawn from Figures 4 and 6 in terms of rainfall depth, namely, the rainfall depths required for scavenging of about 1-μm-diameter particles are much higher than those required for the larger particles. Figure 6 shows an interesting feature. For particles smaller than ~2.2-μm diameter, the rainfall depths are higher for lower rainfall rates. On the contrary, for particles greater than ~2.2-μm diameter, the rainfall depths are higher for higher rainfall rates. We will further discuss this point in section 5.

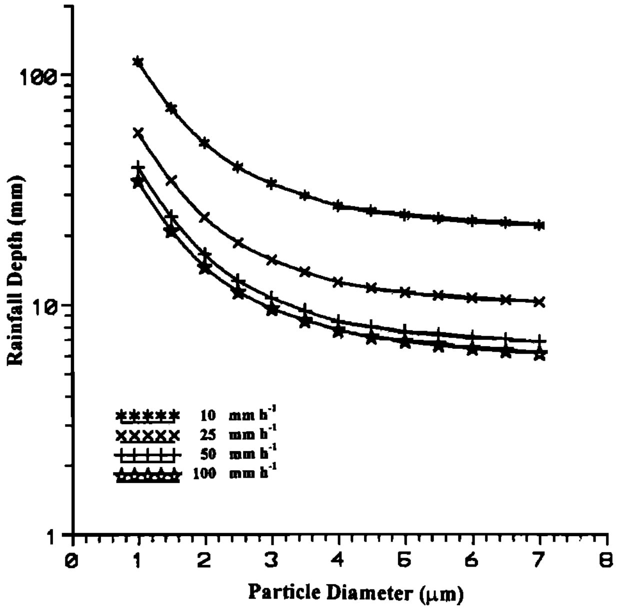

Figure 4. Rainfall depth versus particle size for scavenging by 3.6 ≤ D ≤ 5.0 mm drops as a function of rainfall rate.

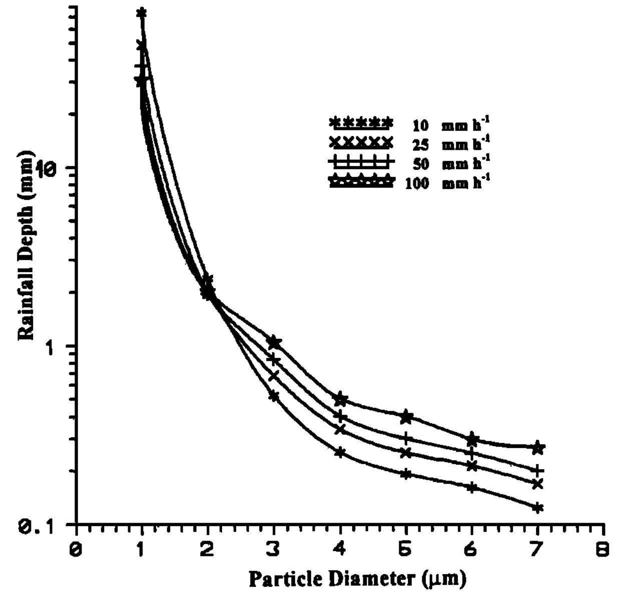

Figure 6. Rainfall depth verses particle size for scavenging by 0.1 ≤ D ≤ 5.0 mm drops as a function of rainfall rate.

4. The Effect of Electrical Forces on Washout Coefficient, Half-Life, and Rainfall Depth

To understand the role of electrical forces in the removal of aerosol particles by scavenging due to large drops, we have computed the washout coefficients using average values of our experimentally measured collection efficiency for charged drops and for the neutral drops falling in presence of an electric field, reported in PK 2. Since these estimates are only for a small range of drop sizes, they may be useful only in understanding the physics of the scavenging process. In the atmosphere, the raindrop sizes vary over more than an order of magnitude, and the electrical charge on them varies from $\sim 10^{-14}$ to 5×10^{-10} C [e. g., *Marshall and Winn*, 1982]. Therefore, to have a correct estimate of the effect of electrical forces on the efficiency of the scavenging process in the atmosphere, one needs to better understand such electrical effects for the entire range of sizes and charges on drops observed in the atmosphere. For example, the collection efficiency data for drops of diameter 1-3 mm collecting particles of diameter > 1 μm in the presence of electrical forces is needed.

Figures 7 and 8 show washout coefficients for drops of $3.6 \le D \le 5$ mm carrying a uniform charge of 2.5×10^{-13}, 2.5×10^{-12}, or 2.5×10^{-11} C, and for the neutral water drops of the same size falling under a uniform horizontal electric field of strength 3.75, 7.5, or 11.25 kV m^{-1} for the four rainfall rates mentioned above. When compared with the case of neutral drops (Figure 1), the results in Figure 7 show that the washout coefficients are 1-3 times higher for the corresponding drop sizes if the drops are charged. This increase in washout coefficient is greater for the smaller particles, especially for ~1-μm-diameter particles. It can be concluded therefore that the effect of drop charge on the scavenging process is more pronounced for smaller particles and progressively decreases with the increasing particle size.

As for neutral and charged drops, the washout coefficients in the case of raindrops falling in an electric field also increase with the increasing particle size. Similar to the case of charged drops, the increase is greater for the smaller aerosol particles, especially for ~1-μm-diameter particles (Figure 8). With the increase in particle size, the washout coefficient increases almost linearly when the electric field is 3.75 kV m^{-1}. However, when the field strength is increased to 7.5 or 11.25 kV m^{-1}, the washout coefficient first increases, reaches a maximum for d_p = ~4 μm, and then decreases with further increase in d_p. In all three cases, however, the values of washout coefficient for very large particles, say, of diameter ~7 μm and above, are almost the same in the absence or presence of the electric field. The values of half-lives and rainfall depths, computed in both the electrical cases, are found to always be less than that for the neutral case.

5. Discussion

On the basis of their measured collection efficiencies, *Lai et al.* [1978] have estimated the washout coefficient for submicron particles. Their washout coefficients decrease with increasing particle size. This trend in washout coefficient is expected, as the collection efficiencies of submicron particles on the left side of the Greenfield gap decrease with increasing particle size. Also, the contribution due to the diffusion process decreases with increasing particle size. Previous theoretical [*Garcia et al.*,1994] and experimental [*Nicholson et al.*, 1991] studies show that the washout coefficients increase with increasing particle size for particles of diameter > 1 μm. Our results agree with this trend.

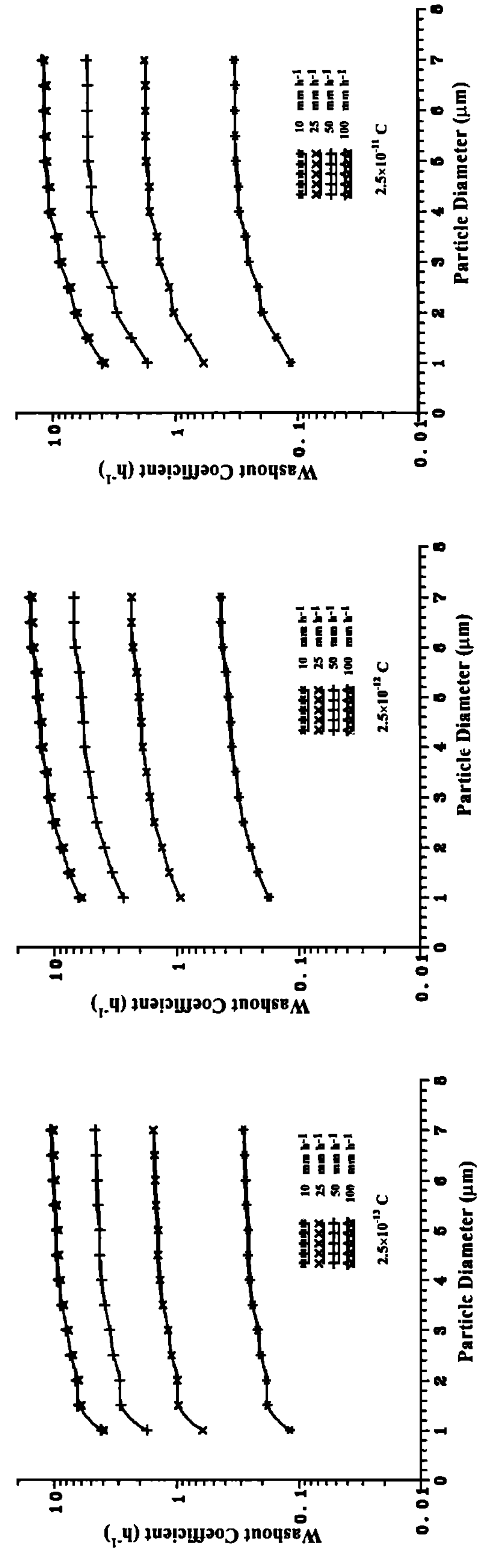

Figure 7. Washout coefficient for charged drops of size $3.6 \le D \le 5.0$ mm as a function of particle size for different rainfall rates.

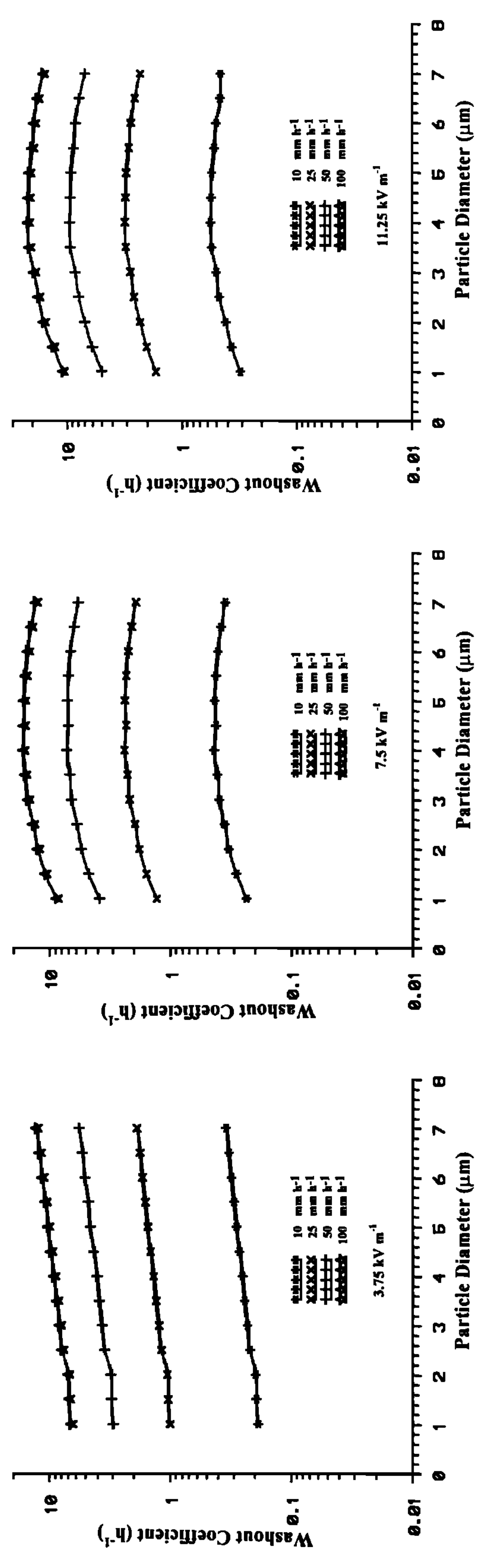

Figure 8. Washout coefficient for neutral water drops of size $3.6 \le D \le 5.0$ mm falling in a horizontal electric field as a function of particle size for different rainfall rates.

However, the washout coefficients measured in field experiments of *Frank et al.* [1991] for particles of diameter > 1 μm decrease with increasing particle size. This trend in washout coefficient is opposite to that generally observed for particle sizes on the right side of Greenfield gap. Frank et al. have attributed this opposite trend to (1) the decreasing importance of the diffusion process with the increasing particle size and (2) the insignificant role of the impaction process in the case of a drizzle type of rainfall because of small raindrop velocities. In our study, since both aerosol particles and the drops are of large size, the impaction process plays a dominant role.

Contribution from the drops in the size range of $0.1 \le D \le 1$ mm to washout coefficient is mainly because of their higher number density and collection efficiency even though their terminal velocity and the cross-sectional areas are small. On the other hand, the drops larger than 1-mm diameter have higher cross-sectional area and terminal velocities, even though their number density is small. Table 1 gives the percentage contributions of three size categories of drops, namely, (1) $0.1 \le D \le 1$ mm, (2) $1.2 \le D \le 3.4$ mm and (3) $3.6 \le D \le 5$ mm, to the total scavenging coefficient. As mentioned earlier, the collision/collection efficiency data for these categories are taken from the theoretically calculated values of *McGann and Jennings* [1991], the graphically extrapolated values of McGann and Jennings, and our experimental measurements, respectively. The large drops in the second and third categories can contribute dominantly to the total washout coefficient, at least for the high rainfall rates considered in our calculations. Exceptionally high contributions of the large drops in these two categories for collection of the 1- or 2-μm-diameter particles are noticeable. Also noticeable is the minimum contribution of the small drops in the first category for collection of 2-μm particles. These results may be attributed to very low collision efficiencies ($\sim 10^{-4}$) for <1- to 2-μm-diameter particles for all drops > 0.5-mm diameter and a sharp increase in collision efficiency for > 2-μm-diameter particles in the results of *McGann and Jennings* [1991]. Because of this sharp change in collision efficiency for 1- to 2-μm-diameter particles, the contributions in Table 1, which are based on combining the two sets of data, may not be quantitatively very accurate. The results in Table 1 should, therefore be used only for qualitative comparison of the contributions by different categories of drops. For particles of diameter > 2 μm, the collision efficiencies of McGann and

Table 1. Percentage Contribution of Drops of Different Size Intervals to the Total Washout Coefficient

Rainfall Rate, mm h^{-1}	Percentage Contribution to Total Λ Particle Diameter, μm							Drop Size Interval, mm
	1	2	3	4	5	6	7	
10	23	2.51	42.9	46.99	45.2	46.79	48.21	$0.1 \le D \le 1$
	13.9	91.52	55.58	52.09	54.02	52.53	51.25	$1.2 \le D \le 3$
	62.8	6.9	1.52	0.914	0.76	0.67	0.54	$3.6 \le D \le 5$
25	8.1	1.23	31.66	35.37	33.53	35.18	36.70	$0.1 \le D \le 1$
	8.1	90.65	64.15	62.00	64.29	62.88	61.94	$1.2 \le D \le 3$
	83.8	8.11	4.19	2.62	2.17	1.94	1.59	$3.6 \le D \le 5$
50	3.73	0.77	24.60	27.90	26.42	28.19	29.25	$0.1 \le D \le 1$
	5.27	87.70	67.83	67.20	70.00	69.44	67.70	$1.2 \le D \le 3$
	91.04	11.46	7.57	4.90	4.04	3.68	3.02	$3.6 \le D \le 5$
100	1.85	0.496	18.71	21.81	20.41	21.86	23.02	$0.1 \le D \le 1$
	3.57	84.38	69.16	69.92	73.09	72.36	71.77	$1.2 \le D \le 3$
	94.64	15.12	12.14	8.03	6.61	6.03	5.08	$3.6 \le D \le 5$

Jennings are comparable to our experimental values. This is reflected in the sharp change in the percentage contribution of small and large drops to the total scavenging coefficient for particles of $d_p > 2$ µm. Further, the percentage contribution of large drops to the total scavenging coefficient increases with increasing rainfall rate, which is expected because of the increased number density of large drops with the increasing rainfall rate.

A comparison of the neutral (Figure 1) and electrical (Figures 7 and 8) cases shows that the washout coefficients for drops in the size range of $3.6 \leq D \leq 5$ mm are 1-3 times higher in the presence of electrical forces. The observations of collection efficiency for some ranges of raindrop and particle size in the presence of electrical forces are reported in the literature [*Adam and Semonin*, 1970; *Lai et al.*, 1978; *Barlow and Latham*, 1983]. However, the data for some size ranges of raindrops and particles (e.g., for raindrops of 1- to 3-mm diameter and particles > 1 µm diameter) are not available. Therefore an estimate of the washout coefficients for raindrop size distribution in the range $0.1 \leq D \leq 5$ mm in the electrical case is not possible. However, on the basis of present results and past data for smaller particles, it may be suggested that the electrical forces enhance the scavenging efficiency of a rainfall. Thus a rainfall from a thunderstorm may be a more efficient scavenger of atmospheric aerosols as compared with that from a weakly electrified cloud.

An important conclusion can be drawn from Figure 6 when one considers that the rainfall depth is essentially the precipitation required for the washout of half the initial concentration of particles. Thus, for a fixed amount of precipitation, the heavier rainfall over a short duration should be more efficient in removing the particles smaller than 2.2-µm diameter, whereas a lower rainfall rate spread over a longer duration may be more efficient in removing bigger particles.

In Figures 9 and 10, we have plotted the washout coefficient for drops of size $3.6 \leq D \leq 5$ mm and for particles of diameter 1, 3, and 7 µm as a function of drop charge and electric field, respectively, for a rainfall rate of 25 mm h^{-1}. Also plotted on the y- axis are the washout coefficients for the neutral case. With the increase in drop charge the values of washout coefficients increase, become maximum for a drop charge of ~2.5 ×10^{-12} C, and then decrease with further increase in drop charge. Results for the case of electric field are more complicated. For 1-µm-diameter particles, the washout coefficient keeps increasing with electric field, whereas for 3-µm-diameter particles, initially it increases and then seems to settle down at some constant value. For 7-µm-diameter particles, washout coefficients show almost a negligible increase with increase in electric field. As discussed in section 4, the values of electric field used in our experiments are observed to significantly contribute to the washout coefficients for particles of only up to ~4-µm diameter. For larger particles, the electrical forces become less important as compared with inertial forces, and hence the washout coefficient curves for these particles show little or no variation with the electric field.

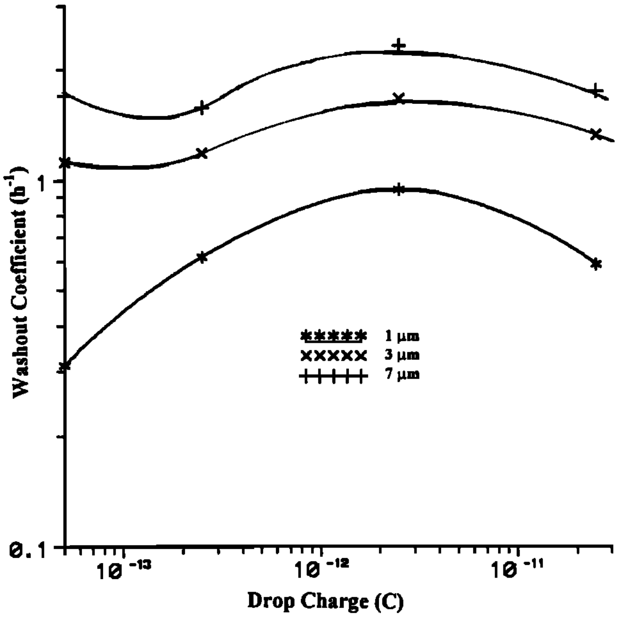

Figure 9. Washout coefficient as a function of drop charge for 1-, 3-, and 7-µm particles for rainfall rate of 25 mm h^{-1}.

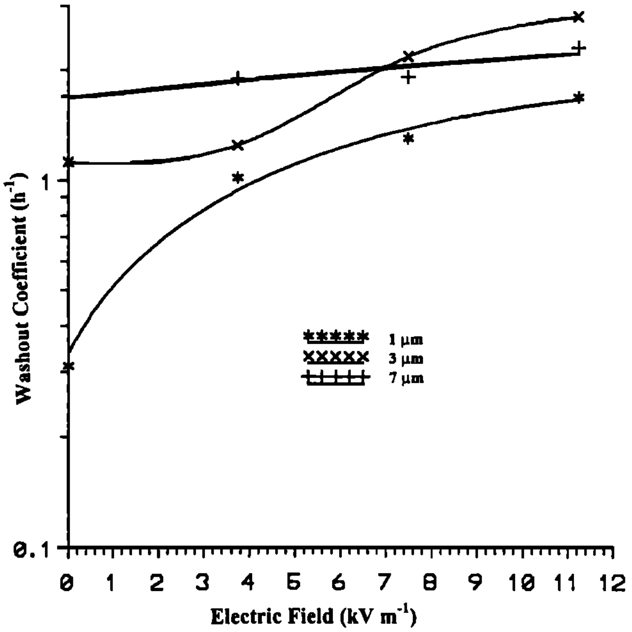

Figure 10. Washout coefficient as a function of electric field for 1-, 3-, and 7-µm particles for rainfall rate of 25 mm h^{-1}.

6. Conclusions

The present results of washout coefficients show that the scavenging of particles is dominated by drops in the 1- to 3-mm-diameter range. It is also evident that the experimental results presented by PK 1 and PK 2 for drops $3.6 \leq D \leq 5$ mm and particles of diameter >1 µm may not be critically needed in the computation of washout coefficients.

Our results demonstrate the relative roles of the inertial and electrical forces in determining the drop-aerosol interaction characteristics for the sizes of drops and particles used in this study. The results suggest that the effect of electrical forces is outstanding for collection of aerosol particles of ~1-µm diameter by 3.6- to 5.0-mm-diameter drops. As the particle diameter increases to ~4 µm, the effect of inertial forces begins to dominate over that of electrical forces. Consequently, the increase in collection of particles of this size under electrical forces is less dramatic. As the particle diameter increases to ~7 µm, the inertial forces increase and become more dominant than the electrical forces. In such cases, the inertial forces mainly govern the aerosol collection in this size range, and the effect of electrical forces becomes almost negligible.

For the ranges of large drops and particles considered in this investigation, the effect of electrical forces is observed to enhance the aerosol collection and thus increase the scavenging efficiency of a rainfall. The effect is likely to be even stronger for smaller particles, as the past and present studies indicate. A rainfall from a thunderstorm may therefore remove the atmospheric aerosol particles more efficiently than one from a weakly electrified cloud.

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