

# Scavenging of aerosol particles by large water drops

## 2. The effect of electrical forces

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**Abstract.** The effect of electrical forces on the collection efficiency of millimeter-sized water drops collecting micron-sized aerosol particles has been investigated in a laboratory experiment. The observations show higher collection efficiencies for drops of 3.6- to 4.8-mm diameters than reported in some of the earlier studies for smaller drops. The limited and sparse data obtained in our experiments show that the collection efficiency of a drop is higher when it is charged or interacts with the aerosol in the presence of an electric field. The collection efficiency shows a maximum when the drop charge of either polarity is in the range of  $10^{-12}$  to  $10^{-11}$  C. The data show that the drop surface charge density required for this maximum decreases with the increase in drop size but is independent of the particle size. However, the peak value of collection efficiency is higher for larger particles. Moreover, the total charge on the drop required for this maximum remains almost constant at about  $2\text{--}3 \times 10^{-12}$  C. The collection efficiency increases with the increase in the electric field, and the effect of the electric field is stronger for larger drops. In high fields, the drop collection efficiency shows a maximum for particles of diameter between 3.5 and 5  $\mu\text{m}$ . The change in collection efficiency for the same change in particle size is larger for higher electric fields. Distortion of large drops and the consequent charge accumulation on the rim of the drop has been proposed to explain the results. The decrease in collection efficiency for large values of drop charge and electric field support the drop-to-particle charge transfer during their interactions.

### 1. Introduction

The collection of aerosol particles by neutral water drops is mainly determined by the processes of Brownian diffusion, thermophoresis, diffusiophoresis, and inertial impaction. However, when the drops are electrically charged and/or interacting with aerosol particles in an electric field, as is the case for a rainfall from an electrically active cloud, the electrical forces play an important role in the collection of particles. Therefore, to assess the efficiency of a rainfall in removing the atmospheric aerosol particles, the role of electric forces in drop-particle interactions needs to be understood. Earlier authors have reported the results of a laboratory simulation experiment to determine the collection efficiency of millimeter-sized neutral water drops collecting micron-sized neutral aerosol particles [Pranesha and Kamra, 1996] (hereinafter referred to as PK). In this paper we extend our measurements to study the effect of electrical forces on the collection efficiency of large drops.

The effect of electrical forces on the collection efficiency of drops has been studied earlier, both theoretically [e.g., Beard, 1974; Wang *et al.*, 1978] and experimentally [e.g., Adam and Semonin, 1970; Lai *et al.*, 1978; Barlow and Latham, 1983], by several investigators. Theoretical studies of collision efficiency,

even though covering a wide range of particle sizes and charges, are limited to drop diameters less than 1 mm. The theoretical computation of collision efficiencies for drops larger than 1-mm diameter is complicated because of their deformation, shape oscillations, and the unsteady flow fields around them. Previous experimental studies have used either high drop charges [Adam and Semonin, 1970; Lai *et al.*, 1978] or submicron polydisperse aerosol particles [Barlow and Latham, 1983]. Moreover, the drops used by Beard [1974] are mechanically suspended in a wind tunnel, and the drops in Lai *et al.*'s [1978] experiments are not falling at their terminal velocities. The effect of electric charge on the collection efficiency of water drops of diameter  $>3$  mm collecting aerosol particles of diameter  $>1$   $\mu\text{m}$  is not investigated in previous theoretical and experimental studies. Moreover, the effect of electric field on the scavenging of particles has not been thoroughly studied so far. In the experiment reported herein, we measure the collection efficiencies for (1) electrically charged large drops falling at terminal velocity through a cloud of neutral aerosol particles and (2) neutral drops falling through a cloud of neutral aerosol particles in the presence of a horizontal electric field.

### 2. Experimental Setup and Procedure

The experimental arrangement, which is described in detail by PK, consists of a drop generator, a vertical free-fall tube, an

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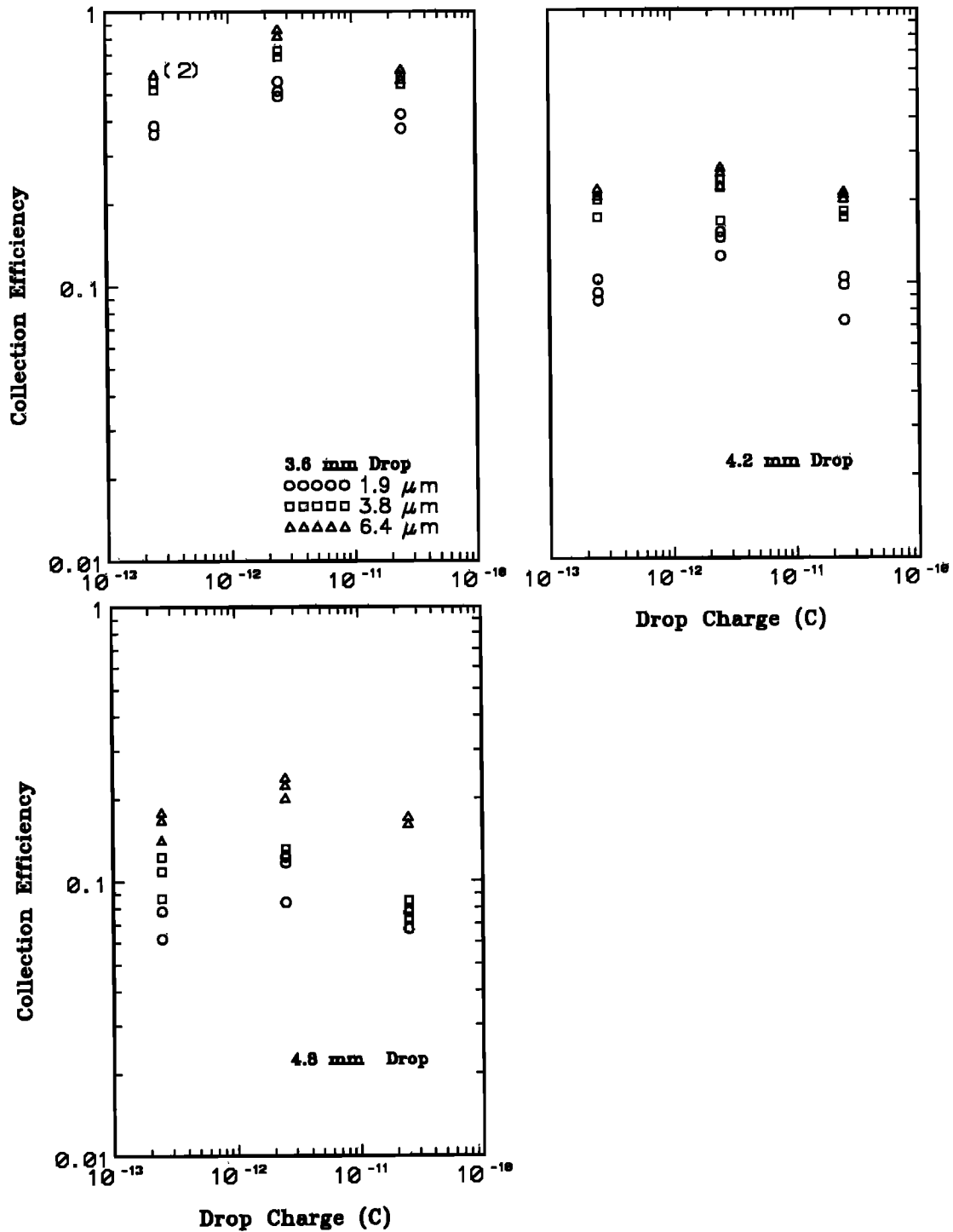
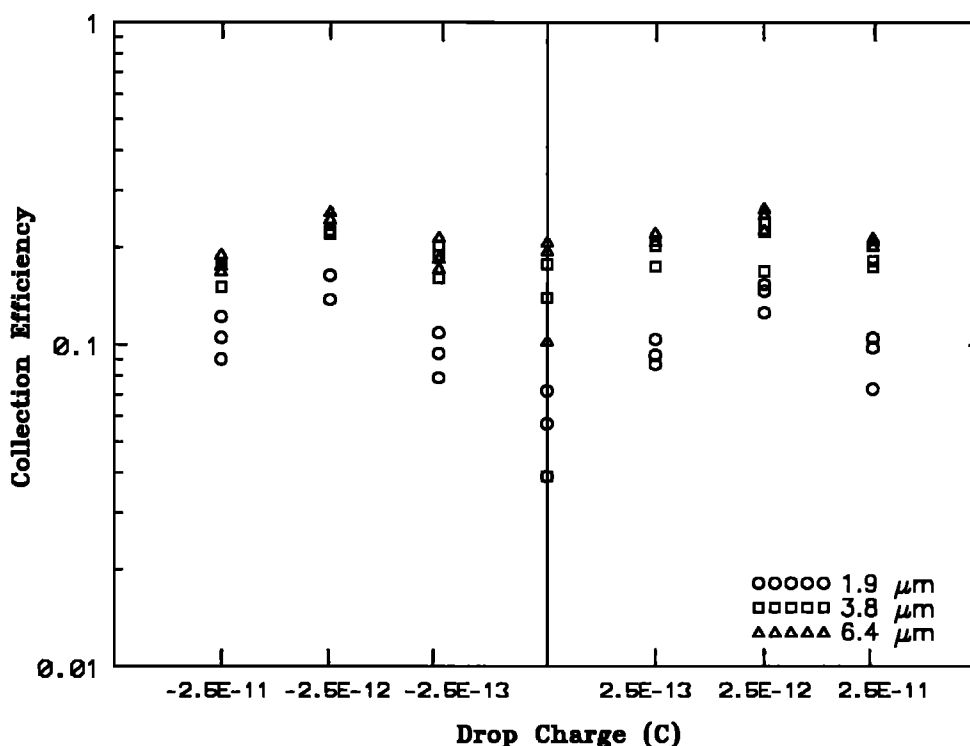


Figure 1. Collection efficiency as a function of drop charge for three drop sizes.

aerosol generator, an aerosol chamber, and the sample collection assembly. Water drops of known size were produced with calibrated capillaries connected to the water reservoir. A wire dipped in the water reservoir was connected to Earth to get neutral drops and to an appropriate potential to get charged drops. To avoid spurious charges on the drops, the capillary tip was located well inside two Earth-connected electrodes of the free-fall system. After the attainment of terminal velocity during their fall in the free-fall tube, the drops were allowed to pass through the aerosol chamber containing a cloud of

monodisperse aerosol particles of known size. The aerosol particles were generated with a TSI model 3054 vibrating orifice aerosol generator and were neutralized to Boltzmann equilibrium level by passing them through a Kr-85 radioactive neutralizer. The drops were then collected in a clean polypropylene bottle at the bottom of the aerosol chamber. One hundred to 200 drops were collected for each data point. The collected samples were stored in a refrigerator for later chemical analysis by atomic absorption spectroscopy in the Atmospheric Chemistry Laboratory of the Institute.



**Figure 2.** Collection efficiency as a function of positive and negative charge on a 4.2-mm drop. For comparison, values of collection efficiency for neutral drops are plotted on the y axis where the value of drop charge is equal to  $2 \times 10^{-13}$  C.

Occasionally, the magnitude of charge on a drop was confirmed by allowing it to pass through an induction ring placed at the bottom of the aerosol chamber.

To make collection efficiency measurements for neutral drops in the presence of a horizontal electric field, one of the electrodes inside the aerosol chamber was connected to Earth while the other one was raised to a desired potential. Two or three data points were obtained for each drop-particle pair in the cases of both the charged and neutral drops in the presence of an electric field.

The performance of the aerosol generator was continuously monitored. If the monodispersity or the continuous supply of aerosol was doubted because of the excessive divergence of the aerosol jet or the occasional plugging-in of the orifice in the generator, the set of observations was discarded, as it generally gave inconsistent and unrepeatable data. However, the number of such sets was less than about 10% of the total number of sets. Total error, due to the chemical analysis, the drop size change due to change in the reservoir's water level, and the nonvolatile impurities present in 2-propanol, was estimated to be less than  $\pm 16\%$ . However, the individual experimental values of collection efficiency for drops under electrical forces are found to deviate not more than  $\pm 5\%$  from the mean value.

### 3. Collection Efficiency of Charged Drops

In our experiments, water drops of three sizes, namely, 3.6-, 4.2-, and 4.8-mm diameter, each carrying a charge of  $2.5 \times 10^{-13}$ ,  $2.5 \times 10^{-12}$ , or  $2.5 \times 10^{-11}$  C, were used for collection of particles of 1.9-, 3.8-, and 6.4- $\mu\text{m}$  diameters. In all, experiments were

performed for nine drop-particle pairs for each value of charge on the drop.

Figure 1 shows the collection efficiency as a function of drop charge for three different drop sizes. In this and all other figures, all data points obtained in our experiment are plotted. When compared with collection efficiencies for neutral drops, reported by PK, the results show a distinct increase in collection efficiency when drops are charged. Further, the plotted values of collection efficiency suggest a small maximum when drop charges are in the range of  $10^{-12}$  to  $10^{-11}$  C. The collection efficiencies seem to decrease with further increases in drop charge. Some experiments performed with negatively charged drops show that the charge of either polarity on the drop has almost similar effects on the collection efficiency (Figure 2). Figure 3 shows the collection efficiency as a function of particle size for three different values of charge and size of the drop. For comparison, it also shows the variation of collection efficiency with particle size for neutral drops. Both Figures 1 and 3 show that the enhancement of collection efficiency due to the presence of charge on the drop is higher for the smallest 1.9- $\mu\text{m}$  particles used in this experiment in the case of all three drop sizes. Comparatively lower values of the collection efficiency for drops carrying a larger charge of  $2.5 \times 10^{-11}$  C are perhaps due to drop-to-particle charge transfer and will be further discussed in section 4.

#### 3.1. Maximum Collection Efficiency of a Charged Drop

Figure 4 presents the collection efficiency as a function of drop surface charge density for all three drop sizes and also suggests a maxima to occur in all cases similar to those in

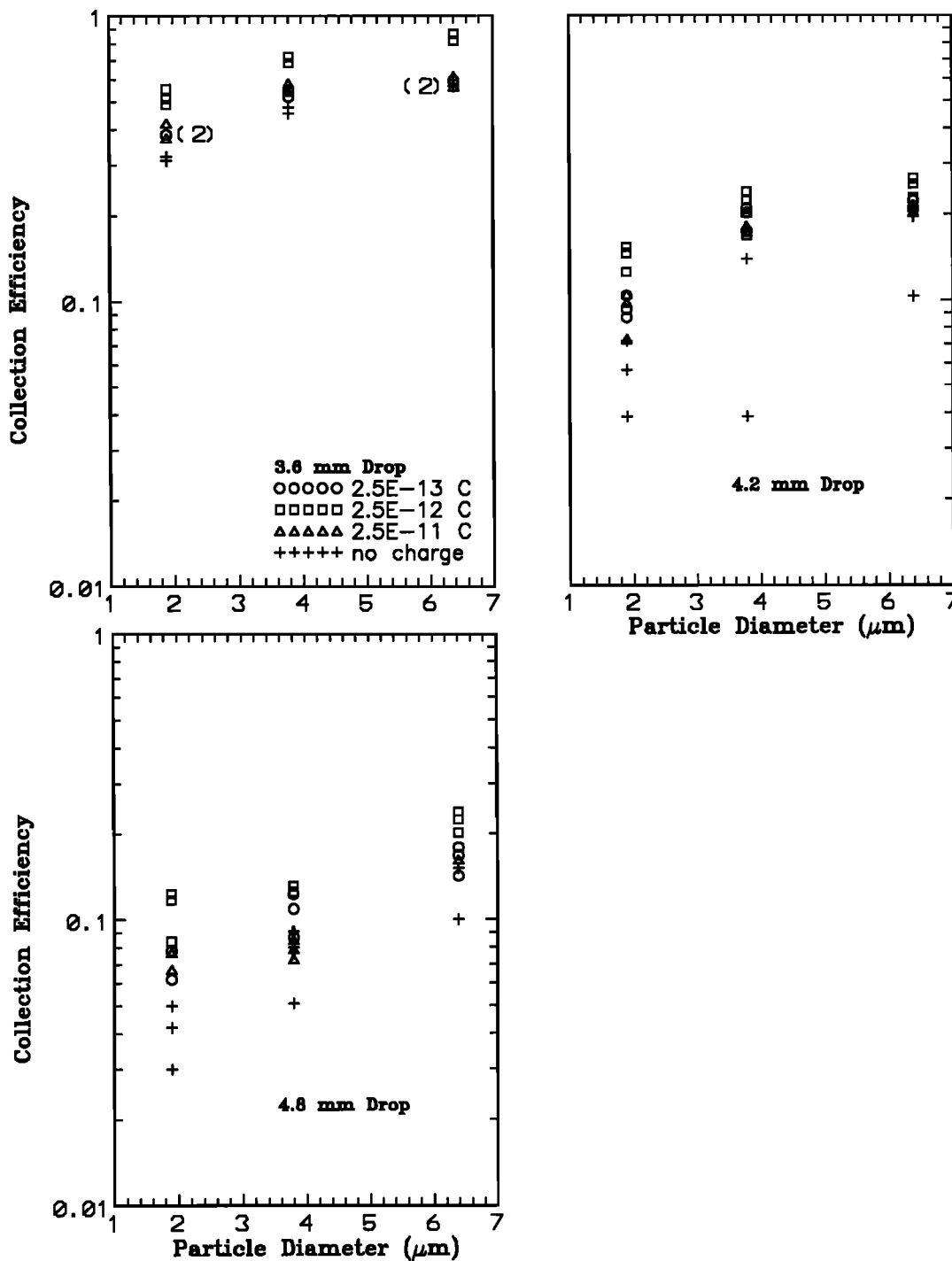


Figure 3. Collection efficiency as a function of particle size for different drop charges.

Figure 1. However, the position of the maximum seems to shift toward larger values of charge density as the drop size decreases. Figure 4 also shows that the surface charge density required for maximum collection efficiency of a drop is independent of the particle size. However, the peak value of the collection efficiency is higher for larger particles.

In Figure 5 we have plotted the surface charge density of the drop estimated from Figure 4 at which the maximum in collection efficiency occurs as a function of drop size. The equation of the straight line in Figure 5 can be written as

$$\ln \rho = -mD + \ln a \tag{1}$$

or

$$\rho = a \exp(-mD) \tag{2}$$

where  $\rho$  is the surface charge density and  $D$  is the diameter of the drop. The constants  $a$  and  $m = 41.26$  and  $0.528$ , respectively.

Equation (2) also gives the condition for maximum collection efficiency of a drop. The surface charge density corresponding to the maximum collection efficiency of a drop

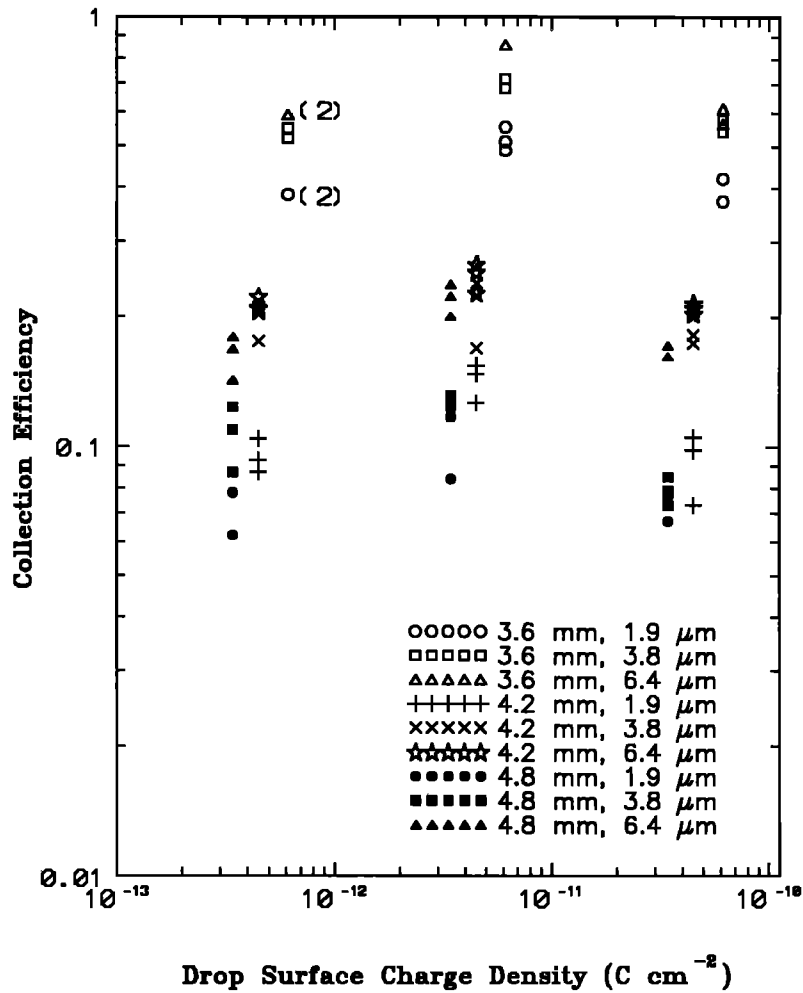
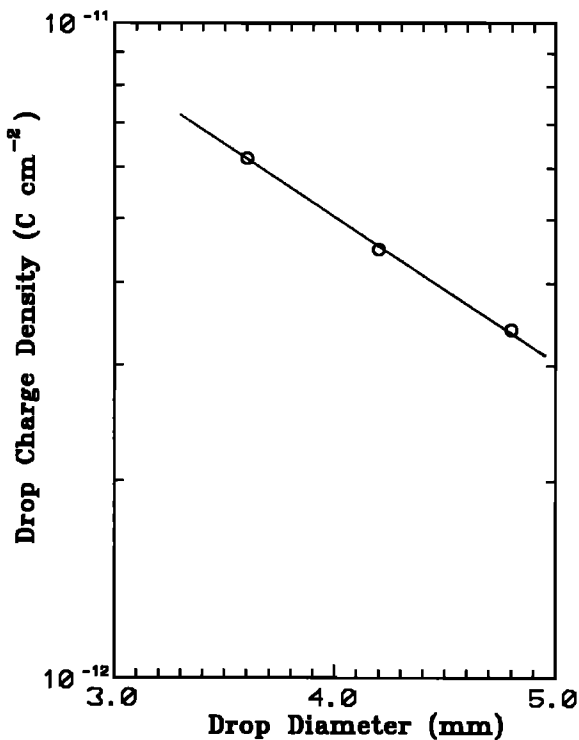


Figure 4. Collection efficiency as a function of surface charge density of the drop.



decreases with increase in its size. However, the total charge on the drop for this maximum collection efficiency remains almost constant at about  $2-3 \times 10^{-12}$  C for the range of drop sizes investigated in this experiment (Figure 6). The dotted portions of the curve in Figure 6 represent the extrapolated values.

#### 4. Comparison With Previous Studies

Since the ranges of parameters used in our experiment and the earlier theoretical studies do not overlap, any difference between the two need not be used for testing the correctness of the theory. Instead, the difference should be helpful in setting guidelines for formulating new theories for this range of parameters. In contrast to neutral cases, the ranges of measured values of collection efficiency for a set in electrical cases were not observed to differ much from each other. However, only two to three measurements were made for each drop-particle pair in our experiment. The following comparison of our results with other studies therefore needs to be considered in view of the uncertainty arising due to this limited number of observations.

Figure 5. Drop surface charge density at which the maximum in collection efficiency occurs as a function of drop size.

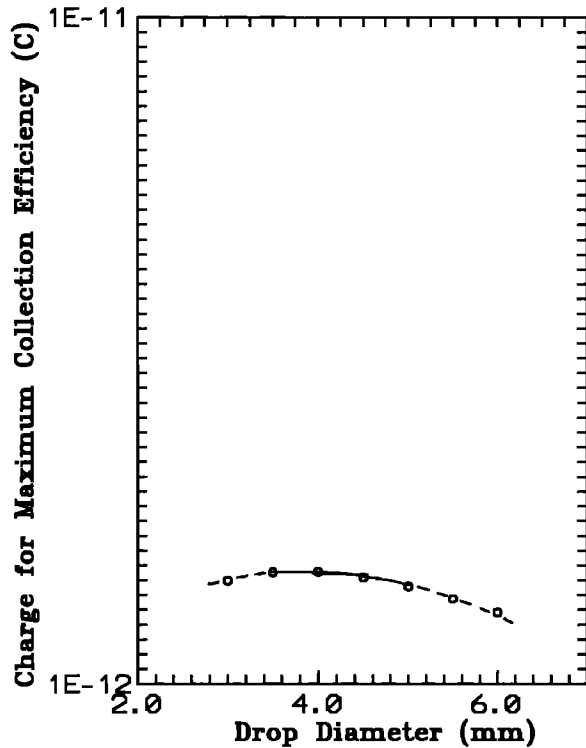


Figure 6. The total charge on the drop at which the maximum in collection efficiency occurs as a function drop size.

Figure 7 shows some of our results with those obtained in some previous experiments. Values of collection efficiencies reported by various investigators range over 3 orders of magnitude. These results can only be understood by considering the largely different values of drop and particle sizes and charges used in different experiments. Comparison of our results with those of the laboratory measurements of *Lai et al.* [1978] brings out some novel aspects of the effect of drop charge on its collection efficiency. In the results of *Lai et al.* collection efficiencies for two drop sizes, 1.24- and 3.64-mm diameters carrying surface charge densities in the range of  $2\text{--}9 \times 10^{-10}$  C  $\text{cm}^{-2}$ , increase quite sharply, reach a maximum, and then decrease for further increases in charge density. Both positive and negative charges are found to have similar effects. Our results indicate very similar trends qualitatively except that the collection efficiency values of larger drops and particles are much higher in our experiment. Further, the maximum in collection efficiency is not as pronounced as in the results of *Lai et al.* but is observed in all cases and is always distinct. The sharp increase in collection efficiency with increasing charge density observed by *Lai et al.* can most likely be attributed to the lower inertia and hence the greater response of their small aerosol particles (maximum 0.72- $\mu\text{m}$  diameter) to the electrical forces.

In the collection efficiency versus charge density curves of *Lai et al.* [1978], the maximum occurs at lower values of charge density for smaller drops. On the contrary, in our results the collection efficiency maximum occurs at higher values of charge density for smaller drops. This reverse trend in the shift of charge density maxima with drop size observed in our results and those of *Lai et al.* can be understood when one considers the

distortion of large drops used in our experiments. This point will be discussed further in the next section.

The combined results of the models of *Grover et al.* [1977] and *Wang et al.* [1978] for the largest drop of diameter 620  $\mu\text{m}$  and of *McGann and Jennings* [1991] for a 1-mm-diameter drop indicate a pronounced increase in the collision efficiency for particles smaller than 2- $\mu\text{m}$  diameter, but almost no change for particles greater than 2  $\mu\text{m}$  with charges of opposite sign on the drop and the particle. In contrast to the above theoretical results for small droplets only, our experimental results for comparatively large drops and particles show a distinct effect of drop charge on the collection efficiency.

The collection efficiency maxima observed by us and *Lai et al.* [1978] have not been observed by *Adam and Semonin* [1970] and *Barlow and Latham* [1983]. It can be understood when one considers the different natures of the particles used in these experiments. *Adam and Semonin* use rod-shaped bacterial spores. Since the charge distribution on such particles is not uniform, a qualitative analysis of their interaction with charged drops is difficult. *Barlow and Latham* use polydisperse particles. Particles of different size respond differently to electrical forces. Since the particles of different size simultaneously interact with the charged drop during its passage through a polydisperse aerosol

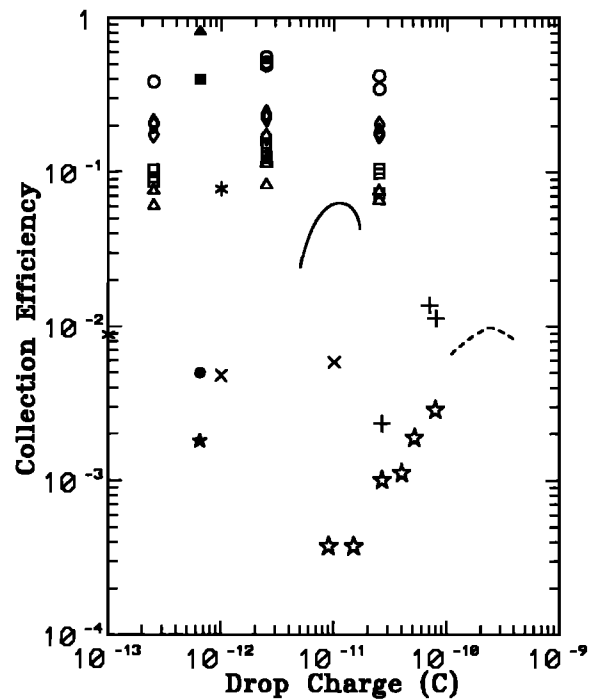


Figure 7. Collection efficiency values of the present experiments as a function of drop charge along with results of some of the previous studies. Legend is as follows: open circles,  $D = 3.6$  mm,  $d = 1.9$   $\mu\text{m}$ ; open squares,  $D = 4.2$  mm,  $d = 1.9$   $\mu\text{m}$ ; open triangles,  $D = 4.8$  mm,  $d = 1.9$   $\mu\text{m}$ ; open diamonds,  $D = 4.8$ ,  $d = 3.8$   $\mu\text{m}$  (this study); solid line,  $D = 1.24$  mm,  $d = 0.3\text{--}0.7$   $\mu\text{m}$ ; dashed line,  $D = 3.6$  mm,  $d = 0.3\text{--}0.7$   $\mu\text{m}$  [*Lai et al.*, 1978]; plus signs,  $D = 2.8$  mm,  $l = 1.2$ ,  $r = 0.7$   $\mu\text{m}$ ; open stars,  $D = 3.7$  mm,  $l = 1.2$ ,  $r = 0.7$   $\mu\text{m}$  [*Adam and Semonin*, 1970]; crosses,  $D = 0.82$ ,  $d = 0.1\text{--}1$   $\mu\text{m}$ ; asterisks,  $D = 1.2$  mm,  $d = 0.1\text{--}1$   $\mu\text{m}$  [*Barlow and Latham*, 1983]; solid star,  $D = 0.62$  mm,  $d = 1$   $\mu\text{m}$ ; solid circle,  $D = 0.62$  mm,  $d = 2$   $\mu\text{m}$ ; solid square,  $D = 0.62$  mm,  $d = 4$   $\mu\text{m}$ ; solid triangle,  $D = 0.62$   $\mu\text{m}$ ,  $d = 10$   $\mu\text{m}$  [*Wang et al.*, 1978].

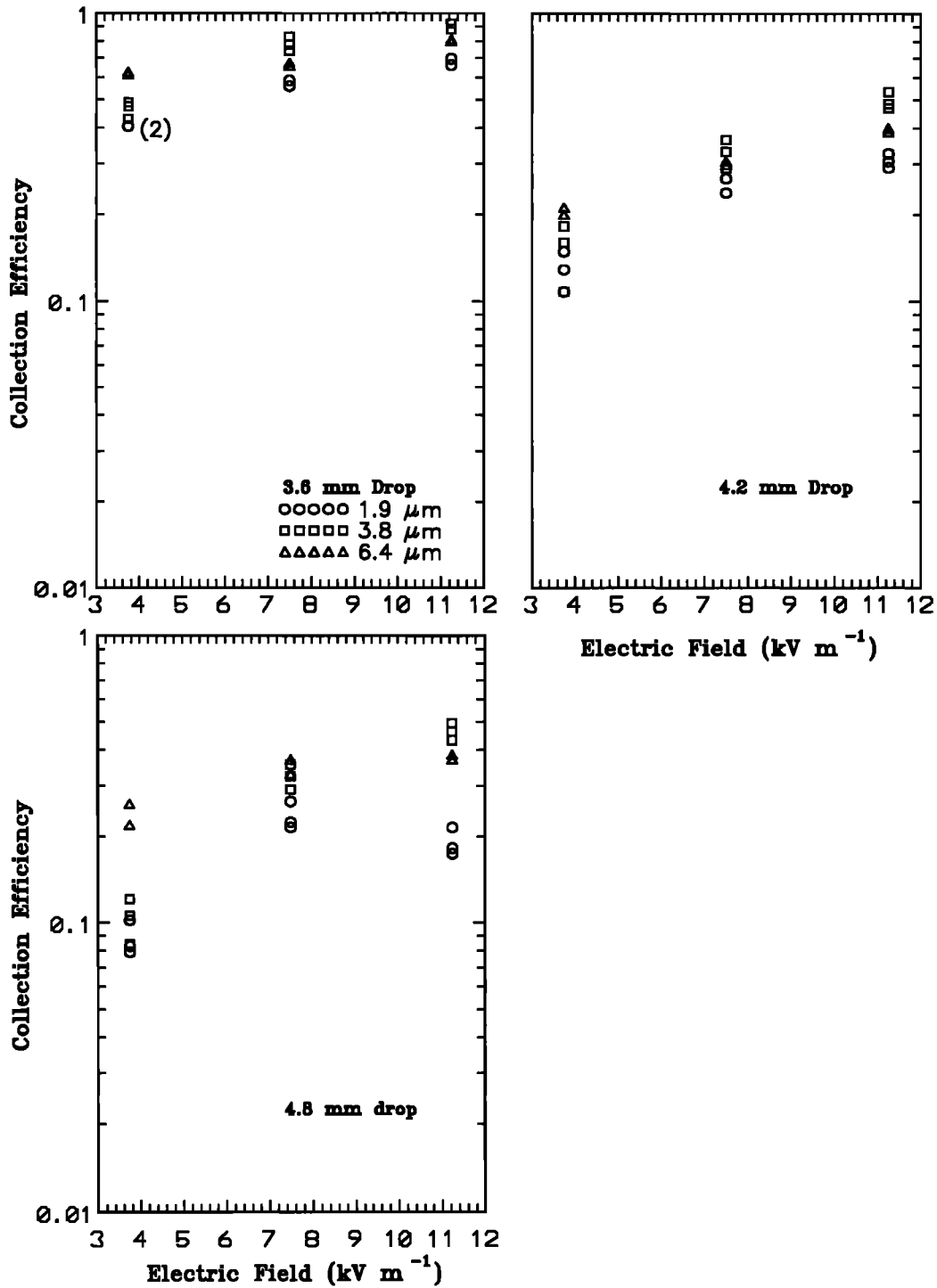


Figure 8. Collection efficiency as a function of electric field for three drop sizes.

cloud any maximum in its collection efficiency with the increase in drop charge may not be of significant magnitude to be observed. Comparatively higher values of collection efficiencies in our results than those of *Barlow and Latham* [1983] are most likely because of the higher inertia of larger particles used in our experiment.

It is evident from Figure 7 that the collection efficiencies observed in our experiments, even though higher, are still comparable to theoretical results for some of the larger particles of *Wang et al.* [1978] and the experimental results of *Lai et al.*

[1978]. It should be noted that the inertia of particles used in our experiments is much higher than the submicron particles used by *Lai et al.* The higher collection efficiencies (even >1) are inferred from the field measurements of *Radke et al.* [1980], who expect such high collection efficiencies to be due to electrical effects. Further, the washout coefficients measured in the field experiments of *Nicholson et al.* [1991] have indicated collection efficiencies greater than unity for particles of diameter >4.3 μm. *Nicholson et al.* found some evidence that such higher collection efficiencies are due to electrical effects.

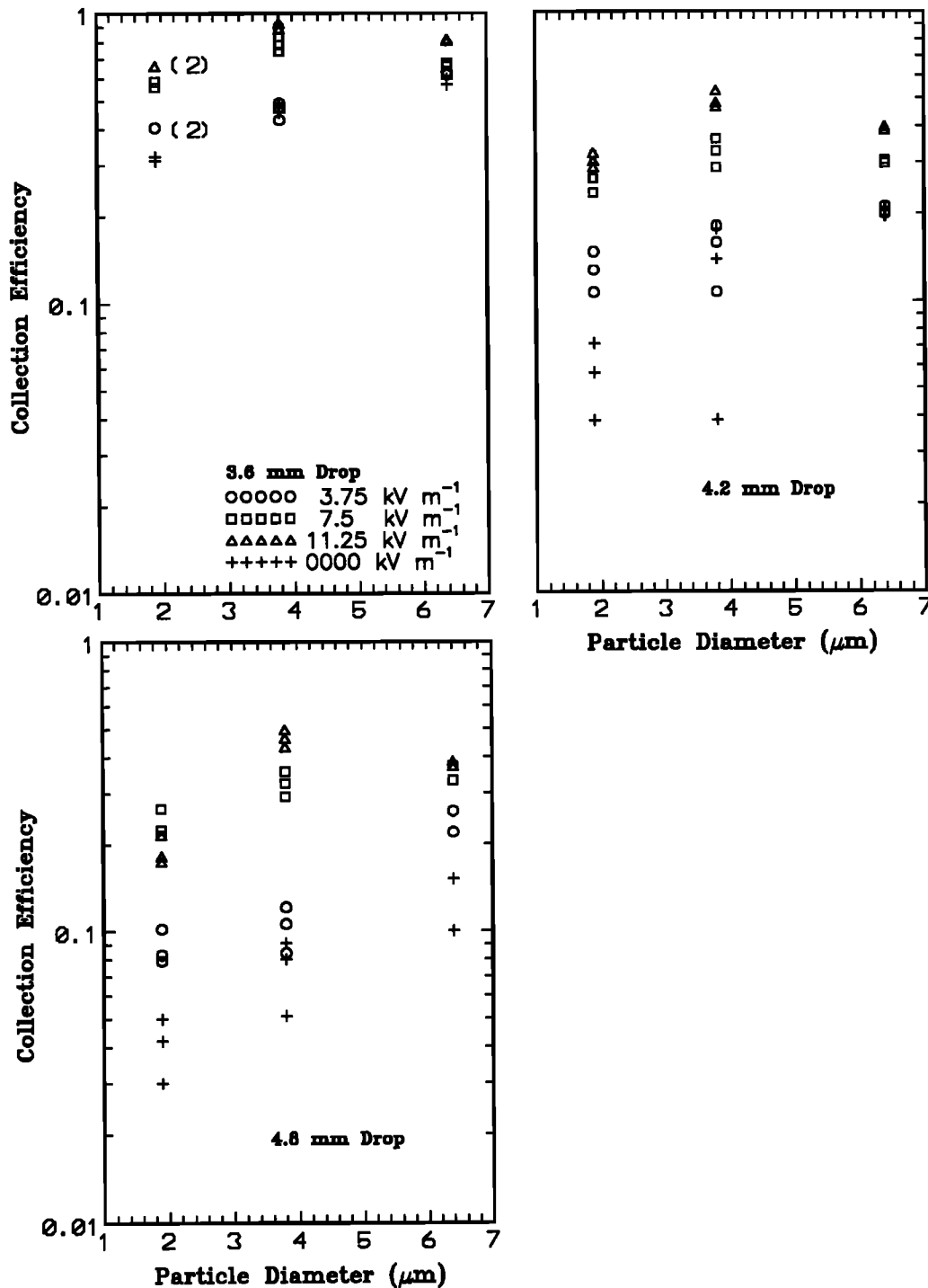


Figure 9. Collection efficiency as a function of particle size for different electric fields.

### 5. Collection Efficiency of Neutral Drops Falling in the Presence of an Electric Field

We also carried out experiments with neutral drops falling in the presence of horizontal electric fields of strengths 3.75, 7.5, and 11.25  $\text{kV m}^{-1}$  for the same three sizes of drops and particles as for the neutral or charged drops. In all, experiments with nine drop-particle pairs were performed for each value of the field strength. Figure 8 shows an increase in collection efficiency with increasing horizontal electric field. A notable

feature for all three drop sizes is that the collection efficiency for 6.4- $\mu\text{m}$  particles is higher at an electric field of 3.75  $\text{kV m}^{-1}$  but lower at 11.25  $\text{kV m}^{-1}$  as compared with that for 3.8- $\mu\text{m}$  particles.

Figure 9 is a plot of collection efficiency as a function of particle size for different field values, including that of the zero-field case. Figures 8 and 9 indicate that the effect of electric field on the collection efficiency is stronger for larger drops. In the case of the zero-field and comparatively weaker electric field of 3.75  $\text{kV m}^{-1}$ , the collection efficiency keeps increasing with the



increase in particle size. However, in the presence of comparatively stronger electric fields of 7.5 and 11.25 kV m<sup>-1</sup>, the collection efficiency seems to first increase, attain a maximum, and then decrease as the particle size increases. For the drop sizes used in the present experiment, the maxima in the collection efficiency occur between 3.5- and 5.0- $\mu\text{m}$  particle diameter. After the maxima, the collection efficiencies for different drops seem to converge toward some lower value of collection efficiency as the particle size further increases. In other words, the increase and decrease of collection efficiency for the same change in particle size are larger for higher electric fields.

## 6. Discussion

In general, the theoretical studies [e.g., Wang *et al.*, 1978] show that the collision efficiency, particularly for particles with diameters less than 2  $\mu\text{m}$ , increases if the interacting spheres (drop-drop or drop-particle) carry charges of opposite polarity. Slightly beyond 2- $\mu\text{m}$  particle size, these theoretical results for the charged and uncharged drop-particle pairs nearly coincide. Measured collection efficiencies of Barlow and Latham [1983] confirm such a trend, at least for particles with diameters less than 2  $\mu\text{m}$ . Results of the present experiment show that in the case of collector drops larger than 3.6 mm, the effect of drop charge on collection efficiency is clearly distinguishable, even for particles larger than 2  $\mu\text{m}$ . The collection efficiency, of course, tends to attain the neutral-case values as the particle size approaches 10  $\mu\text{m}$ .

With the increase in drop size, the surface charge density required for the maximum collection efficiency of a charged drop decreases in our observations, while it increases in Lai *et al.*'s [1978] observations. We propose the following to explain the opposite trends in these two sets of observations. Water drops smaller than 2.0-mm diameter maintain their approximately spherical shape, and any electrical charge on them is uniformly distributed over the surface. Drops larger than 2.0 mm get deformed into an approximately oblate spheroidal shape and develop a flattened base when they are of 2.8-mm diameter [Pruppacher and Pitter, 1971]. The flattening of the base of the drop becomes increasingly pronounced with the increasing diameter of the drop, and when the diameter is 4.0 mm, a concave depression begins to develop at the base. The charge density on the surface of such deformed drops get redistributed. Since surface charge density is maximum where the surface curvature is maximum, the maximum accumulation of charge results on the rim of the drop. Larger drops accumulate more charges on the rim since they experience more deformation [e.g., Kamra and Ahire, 1989]. Now the collection of aerosol particles by a drop falling through an aerosol cloud, is likely to be most influenced by the charge on the rim of the drop. Supporting this is the observation of Horn *et al.* [1988] that the maximum collection of aerosol particles, in their wind tunnel experiments with solid models of oblate-shaped raindrops, occurs at the rim of the models. Since the surface charge density is generally calculated by assuming the drop to be spherical, the larger drops, being more distorted, will exert greater influence in collecting aerosol particles than the smaller ones for the same values of charge density on them. Consequently, as our observations show, larger drops should show maxima in collection efficiency for lower values of the drop charge density. The drops used in the experiments of Lai *et al.* especially the 1.24-mm drops, are much smaller and are

not likely to experience much deformation. Any accumulation of charge on the rim of such drops may not be sufficiently large to show such difference in the maximum collection efficiency with drop size.

The initial sharp increase in collection efficiency with surface charge density in the results of Lai *et al.* [1978] may be due to the pronounced response of small particles to electrical forces. A possible explanation for the decrease in collection efficiency for higher charge densities on the drop has been attributed by Lai *et al.* to a possible charge transfer from drop to particle such as the one observed by Sartor [1954] for drop-drop collisions. The small particle acquires charge through a spark jumping from drop to the particle on its approach to the collector drop, resulting in drop-particle repulsion instead of charge-induced attraction. This suggests that beyond a certain value of drop surface charge density, the charge transfer may become more significant, resulting in a decrease in collection efficiency. Our results not only support such a drop-to-particle charge transfer hypothesis but also suggest the critical value of drop charge density at which the reversal in variation of collection efficiency with drop charge density occurs. Although this critical value of drop charge density decreases with increase in drop size, the total charge on the drop corresponding to this critical value of charge density is almost constant at  $2\text{-}3 \times 10^{-12}$  C for the whole range of drop sizes investigated in our experiment. The observation strongly suggests the development of a critical electric field between the point charge on the drop and particle for the electric spark to occur. Since the magnitude of this critical electric field will be independent of the particle size, our observation of the occurrence of the maxima in collection efficiency at the same value of the surface charge density of a drop for all particle sizes is expected.

A water drop falling in the presence of a horizontal electric field is polarized, and the induced charges appear near the rim of the drop. Particles flowing past the drop will experience an electrostatic force of attraction toward the drop. Therefore the collection efficiency of the drop should increase. Since the magnitude of induced charges increases with the increase in electric field and is larger for bigger drops, the increase in collection efficiency with the increase in the electric field and drop size as observed by us is expected. Moreover, the sharp increase in collection efficiency with particle size observed for the smaller particles can be explained on the basis of their smaller inertia and therefore their larger response to electric forces. Now while the inertial force of a particle increases as the cube of its diameter, the electrostatic force increases as the square of its diameter. Therefore the decrease in collection efficiency with particle size observed for larger particles in our experiment indicates that the inertial collection becomes more dominant as compared with the electrostatic collection in cases of such large particles. In other words, the position of collection efficiency maxima in Figure 9 indicates the particle size where the inertial and electrostatic collections of particles may just balance each other.

## 7. Conclusions

Results of the present experiment show that the collection efficiencies of millimeter-sized water drops collecting micron-sized aerosol particles are higher in the presence of electrical forces. The collection efficiency is maximum when the drop charge is between  $10^{-12}$  and  $10^{-11}$  C. The decrease in collection

efficiency for higher drop charges is attributed to a charge transfer mechanism of the type observed by Sartor [1954]. In our results the surface charge density required for the maximum collection efficiency decreases with increase in drop size, which is opposite to the trend observed by Lai *et al.* [1978]. This opposite trend is explained as being due to the deformation and the consequent redistribution of charges on the surface of the large drops used in our experiments. In an electric field of  $3.75 \text{ kV m}^{-1}$ , the collection efficiency increases linearly throughout the range of particle sizes investigated by us. However, in higher electric fields of  $7.5$  and  $11.25 \text{ kV m}^{-1}$ , the values of collection efficiency first increase, show a maximum for particles of diameter  $\sim 4 \text{ }\mu\text{m}$ , and then converge to the neutral-case value as the particle diameter approaches  $10 \text{ }\mu\text{m}$ . Further, the effect of the electric field is found to be greater in cases of larger drops. This has been attributed to the greater deformation and consequent appearance of larger induced charges on the rim of the larger drops. Results indicate that the electrical and inertial forces in our experiment may just balance each other when the particle diameter is  $\sim 4 \text{ }\mu\text{m}$ . Collection efficiencies of still larger particles may be mainly governed by inertial forces.

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