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ELECTRICAL ASPECTS OF RAINOUT

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PREFACE

Rainout¹ commonly denotes the aggregate of phenomena associated with precipitation scavenging of radioactivity from a cloud of nuclear debris that is within a natural rain cloud. (In contrast, the term, "washout," is applicable when the nuclear cloud is below the rain cloud and the term, "fallout," commonly denotes the direct gravitational settling of contaminated solid material from a nuclear cloud.) Nuclear debris aerosols may be scavenged within natural clouds by a variety of different physical processes which may involve diffusion, convection, impaction, nucleation, phoresis, turbulence and/or electricity among others. Processes which involve electrical aspects will be scrutinized in this report for their susceptibility to the intimate presence of the radioactive-cloud environment.

This particular choice of electrical processes is not accidental. Nearly all of the listed processes were examined earlier by Williams² among others.³ His rough estimates suggested that electrical effects, and to a lesser extent turbulence, could enhance the scavenging of those submicron aerosols which reside in the size-range that bridges the minimum in the scavenging rate coefficient which is commonly called the "Greenfield gap." This minimum in the scavenging-rate coefficient is created by the simultaneous reduction (with increasing size) of scavenging via diffusion and the reduction (with decreasing size) of scavenging via inertial impaction. However, Williams omitted the specific influence of a radioactive environment. This report aims to remedy this omission.

Electrical aspects of rainout will be described. It will be shown that the intimate presence of very high radioactivity serves to generate enough ionization to effectively "short-out" any mechanisms that would tend to concentrate charge or generate electric fields within natural clouds. The presentation closely follows the outline of the earlier survey by Rosenkilde.⁴ However, much sharper conclusions now can be drawn. Revised estimates will be presented for attachment rates between submicron nuclear-debris aerosols and natural cloud droplets under contrasting electrical situations which are characterized by very high and very low rates of ionization.

INTRODUCTION

The radioactivity of debris particles from a free nuclear air-burst can completely alter the electrical complexion within a natural cloud. There is sufficient residual beta and gamma activity to generate ion pairs in the atmosphere at rates that are many orders of magnitude greater than those produced by natural terrestrial sources and cosmic rays. This production, even when counterbalanced by recombination and attachment, still yields steady-state ion concentrations that are several orders of magnitude greater than those normally present in fair weather. Such concentrations of charge, when coupled with the dominant strength of electrical forces in many situations, suggest that electrical interactions should be included in investigations of the scavenging of nuclear debris.

The physical system of interest consists of a hierarchy of species interacting within a given cloud volume. On the very largest scale are the infrequent (10^{-4} cm^{-3}) and large (1 mm) hydrometeors (raindrops) that hurtle ($6 \text{ m}\cdot\text{s}^{-1}$) through this volume sweeping up within their path the more numerous ($100\text{-}300 \text{ cm}^{-3}$) and smaller scale ($5\text{-}30 \text{ }\mu\text{m}$) cloud droplets which settle much more slowly ($1 \text{ cm}\cdot\text{s}^{-1}$) through the multitude (10^{19} cm^{-3}) of background air molecules. (Some representative values for atmospheric parameters of interest and their variation with tropospheric altitude are tabulated in Appendix A.)

Now contemplate the additional presence here of small particulates with a scale ($0.01\text{-}1.0 \text{ }\mu\text{m}$) and concentration ($10^3\text{-}10^5 \text{ cm}^{-3}$) comparable with

those of aerosols found over an industrial city. Allow for the possibility that these particulates may have sufficient beta and/or gamma activity (10^{-10} to 10^{-5} Bq· μm^{-3}) to produce small ions in the surrounding atmosphere at rates (exceeding 10^9 cm $^{-3}$ ·s $^{-1}$) which greatly exceed the generation rate due to naturally occurring radioactivity in the troposphere (less than 10 cm $^{-3}$ ·s $^{-1}$). Representative magnitudes of interest are listed in Table 1.

Table 1. Constituents of interest and their representative sizes and concentrations.

Species	Radius (μm)	Concentration (cm $^{-3}$)
Raindrops	10^2 - 10^3	10^{-4}
Cloud droplets	5 - 30	100-300
Particulates	0.1-1.0	10^3 - 10^5
Small ions	mean free path ~ $\left\{ \begin{array}{l} 0.01-0.04 \\ 0.06-0.20 \end{array} \right.$	10^2 - 10^7
Air molecules		10^{19}

The simultaneous presence of gas molecules, ions, possibly free electrons, and both neutral and charged droplets and particles introduces the possibility of many kinds of interactions. A partial list of possible interactions can be generated by following a typical sequence of events after the decay of a representative particle. Consider first, the events following a beta decay, which leaves the particle with a positive charge and introduces into the surrounding medium a primary electron with an energy in the range 0.5 - 1.0 MeV. This electron collides with the surrounding gas molecules, occasionally

ejecting an electron that leaves behind a positive ion and costs an average of 34 eV, until it loses all its energy by producing some 3×10^4 electron-ion pairs within a 2-5 m radius sphere. At low altitudes essentially all the secondary electrons very rapidly (~ 20 ns) become attached to neutral O_2 molecules forming negative ions. (A more detailed examination of the fate of free electrons is given in Appendix B.)

Primary gamma decay, on the other hand, does not leave the particle charged, but does contribute to the ionization over a larger region than ascribed to primary beta decay. A typical 1-MeV gamma has a mean free path in air of 350 m and loses an average of 44% of its energy in its first encounter. If the initial flux is sufficiently great there still exists the possibility of influential ionization out to several kilometers.

At this point the ions become available for ion recombination, hydrated cluster formation, attachment to charged or neutral particles or droplets, as well as electron transfer and other chemical reactions with trace contaminants. Several important questions are apparent:

- 1) What is the ambient ion concentration in the presence of particles and cloud droplets?
- 2) What is the magnitude of the effective charge on a decaying particle within such an environment?
- 3) What are the magnitudes of charge that may be expected on cloud droplets and raindrops in the same environment?

Answers to these questions cannot be wholly independent of gross electric fields that may be present. The gravitational and convective forces that are

believed to drive the charge separation processes and generate electric fields in natural clouds still are operative here. However, such processes must adjust to the greatly increased ion concentration, which causes a proportionate increase in the electrical conductivity within the cloud, and thus may serve to limit the growth of electric fields.

Finally, some of the attachment processes, which influence the magnitude of the ion concentration, depend upon the strength of the ambient electric field. Thus, the magnitudes of the ion concentration, the particle and droplet charges, the electric field, and ultimately, even the scavenging rate of the debris particles, which is of primary interest here, are all intimately coupled and ought to be determined simultaneously in a self-consistent manner.

Each of these questions and issues will be examined in the following sections. Technical details and data used are summarized in a series of appendices abstracted from earlier reports.

AMBIENT ION CONCENTRATION

Primary beta and gamma rays from the radioactive particles lose energy through interactions with the neutral atmospheric gas molecules. These interactions produce positive ions and secondary electrons. At tropospheric densities, the electrons rapidly become attached to neutral molecules to form negative ions (see Appendix B). Even in the presence of ionization rates exceeding $10^9 \text{ cm}^{-3} \cdot \text{s}^{-1}$, the tropospheric free electron concentration reaches a steady-state value that is less than 300 cm^{-3} in a characteristic time scale of 30 ns. Because of this, electron interactions with all other constituents (e.g., ions, particles, droplets, etc.) will be suppressed in the subsequent discussion.

The steady-state concentration of positive and negative ions depends upon a dynamic balance between the ion production rate per unit volume, s , and the various recombination and attachment processes among the ions, debris particles, and cloud droplets. When the positive and negative ion concentrations are approximately equal, the steady-state concentration, n , is determined by a quadratic equation of the form

$$\alpha n^2 + 2\sum n - s = 0, \quad (1)$$

where α is the volume recombination coefficient between ions and $2\sum$ represents the total ion depletion rate due to attachment of ions to particles and droplets by whatever means. Estimates for the values of these coefficients in various situations of possible interest and the resulting forms taken by the solution of Eq. (1) are discussed in Appendix C.

The curves displayed in Figure 1 illustrate that when the ion generation-rate exceeds $10^9 \text{ cm}^{-3} \cdot \text{s}^{-1}$, which is a representative value for fresh nuclear debris, the steady-state bipolar ion concentration exceeds 10^7 cm^{-3} . This concentration is determined principally by ion recombination and does not begin to diminish until the ion attachment rate exceeds 10 s^{-1} . Such an attachment rate can be attained only in the presence of large ambient electric fields or highly charged collectors, such as could be present under thunderstorm conditions, or else unreasonably high concentrations of large particles and droplets. In the opposite extreme, when the ion generation-rate is of the order of $10 \text{ cm}^{-3} \cdot \text{s}^{-1}$, which is representative of normal atmospheric conditions, very modest collector concentrations, collector charges, or ambient fields can cause significant reductions in the equilibrium ion concentrations below the nominal value of 10^3 cm^{-3} . In the former case, n is proportional to $s^{1/2}$; in the latter situation, n is inversely proportional to s and directly proportional to the ambient field or collector charge and the collector concentration (see Appendix C).

The ion attachment rate is being treated here as a parameter. However, it depends upon the collector charge and the ambient electric field and these quantities in turn, depend upon the ion concentration itself. Ultimately, all three of these quantities must be determined simultaneously by using self-consistent relationships that include electrostatics and the meteorological driving forces of precipitation and convection. Such a determination will be illustrated subsequently.

Fig. 1. Steady-state ion concentration as a function of the ion depletion-rate due to attachment of ions to aerosol particles and droplets, for various ion generation-rates from 10^1 to $10^9 \text{ cm}^{-3}\cdot\text{s}^{-1}$. Under normal atmospheric conditions, the ion generation rate is of the order of $10 \text{ cm}^{-3}\cdot\text{s}^{-1}$ and very modest collection concentrations, collector charges, or ambient fields can cause significant reductions in the steady-state ion concentration below the nominal value of 10^4 cm^{-3} . In the presence of fresh nuclear debris, the ion generation-rates may exceed 10^9 cm^{-3} . Then the steady-state ion concentration, which exceeds 10^7 cm^{-3} , is determined solely by ion recombination and is unaffected by the presence of large numbers of other sinks for ions.

EFFECTIVE PARTICLE CHARGE

Electrical interactions between radioactive nuclear debris particles and natural cloud constituents would be reduced greatly if the particles remained effectively uncharged. Consequently, it is quite important to establish the existence and also the duration of any particle charge.

Primary interest in the present context centers on radioactive debris particles situated in a bipolarly ionized atmosphere where ion concentrations greatly exceed those which occur naturally. Although it was recognized that such particles could acquire positive charge through beta decay, Greenfield⁵ argued that the half-life of this charge within such an environment would be less than 1/20 of the time between disintegrations. Therefore, he concluded (i) that there was practically no probability of these particles becoming more than singly charged, and (ii) that although they might conceivably possess a small intermittent positive or negative charge, effectively their net charge would be zero. In contrast, Keefe, et al.⁶ argued that charge will very probably be distributed on particles in a bipolar atmosphere according to a Boltzmann distribution law which depends upon the ratio of the particle's electrostatic potential energy (which varies inversely with particle radius) to the mean ionic thermal energy. They predicted that 70% of all particles with a radius of 0.1 μm would be charged with 20% of the total being doubly charged - irrespective of any intrinsic particle radioactivity. The average absolute particle charge should be close to the unit electron charge. These two viewpoints are not necessarily incompatible, but some clarification is useful.

The particle charge distribution can be obtained by solving a set of rate equations (see Appendix E) which involve the particle decay rate, the positive and negative equilibrium concentrations, and the various attachment coefficients between positive or negative ions and charged or neutral particles. Mead⁷ showed that the asymptotic steady-state solution of these rate equations takes the form of a shifted Gaussian distribution with a mean charge and dispersion that depend upon the particle radius and activity, the ambient external electric field, and the ionic mobilities (see Appendix E). His formulae reduce to all previously known special cases (without activity): the low field case studied by Klett², the high field case studied by Gunn⁹, the case with unequal ionic mobilities solved by Gricard¹⁰ (whose method Mead extended) and Sal'm¹¹, and the "Boltzmann" charge-distribution law proposed by Keefe, et al.⁶ which is applicable when the ionic mobilities are equal.

Figures 2 and 3 show the variation in debris particle charge with particle radius for two extreme cases of particle radioactivity. Both figures have been constructed assuming monodisperse particle distributions with the same fixed mass of material (debris) distributed uniformly throughout a spatial volume of air. When the particle activity is low ($10 \text{ Bq} \cdot \mu\text{m}^{-3}$ in Fig. 2), the average number of negative charges per particle slightly exceeds the average number of positive charges per particle (because unequal ionic mobilities were assumed; see Appendix D) and both averages are less than unity over almost the entire range of radii from 0.01 to 1.0 μm . Clearly, the average net charge for the entire distribution is nearly zero and the dispersion shows that relatively few particles have more than one charge in general agreement with Greenfield. The distribution is Gaussian, but shifted

Fig. 2

Fig. 3

Figs. 2-3. Variation in debris particle charge with particle radius for two extreme values of material radioactivity: $10 \text{ Bq} \cdot \mu\text{m}^{-3}$ in Fig. 2; $10^{-5} \text{ Bq} \cdot \mu\text{m}^{-3}$ in Fig. 3. Both figures have been constructed by assuming monodisperse particle distributions with the same fixed mass of material (debris) distributed throughout a spatial volume of air. Unequal ionic mobilities were assumed. Curves labeled P and N respectively represent the mean numbers of positive and of negative charges per particle for monodisperse distributions at each radius. Curves labeled D represent the dispersion about the mean value of the respective charge distributions. Only the larger particles have sufficient material (and hence radioactivity) to acquire a mean positive charge by beta decay, even in the bipolar ionic environment. This effect is proportional to the particle volume and commences at a smaller radius for the higher activity case.

ever so slightly toward the negative side, with the shape predicted by Keefe, et al. However, as the particle size is increased, the activity per particle is increased and eventually dominates over the rate of neutralization by negative ions. Under these conditions, the mean charge of the distribution can be driven back through zero toward positive values. When the particle activity is large ($10^5 \text{ Bq} \cdot \mu\text{m}^{-3}$ in Fig. 3) this phenomenon is manifest for radii exceeding $0.1 \mu\text{m}$. Quite large numbers of charges (approaching 10^3 for $1\text{-}\mu\text{m}$ radii particles) can be sustained under these conditions.

Thus, we conclude that some of the larger radioactive debris particles could develop large numbers of positive charge under certain conditions. This possibility must be taken into account in studies of rainout.

CLOUD DROPLET CHARGES

All cloud constituents situated in an ionized atmosphere are potential carriers (and sinks) of charge. Large droplet charges can increase the droplet's collection rate of ions and thus reduce the steady-state ion concentration (see Appendix C). Large droplet charges also enhance the droplet's collection rate of charged debris particles and thus increase the chances for in-cloud scavenging. Therefore, it is of interest to examine the magnitude of charge that can be expected on cloud droplets (and raindrops) in the presence of ionization.

Many observations have been made of the mean electric charge on cloud droplets and raindrops under natural conditions in fair weather and in thunderstorms. (Takahashi¹² has summarized a number of these measurements.) Various fits have been made to this type of data. The most commonly used charge-radius relationships have the form

$$(Q/e)_0 = KR^2 \quad (2)$$

and show that the number of droplet charges, Q/e , is proportional to the square of the droplet radius, R . If R is given in μm , then $K \approx 40 \mu\text{m}^{-2}$. This suggests that 10- μm radius cloud droplets may possess several thousand charges under normal conditions.

A relationship with this dependence on radius is known to be a consequence of field charging in a unipolar ionic atmosphere.¹³ The collecting drop, which is assumed to be a conductor for the sake of simplicity, perturbs the otherwise uniform field in its vicinity allowing ions to reach its surface

along the electric field lines which connect the surface with infinity. The accumulated number of charges quickly approach the saturation value,

$$(Q/e)_s = 12\pi\epsilon_0 e^{-1} ER^2 \approx (0.002 \text{ m}\cdot\text{V}^{-1}\cdot\text{m}^{-2}) ER^2, \quad (3)$$

where E is the electric field, R is the collector radius, $\epsilon_0 = 8.85 \times 10^{12} \text{ F}\cdot\text{m}^{-1}$, and $e = 1.6 \times 10^{-19} \text{ C}$. The characteristic charging time scale is $t_s = 4\epsilon_0(e\mu n)^{-1}$, where μ is the ionic mobility ($\sim 1 \text{ cm}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$) and n is the steady-state ion concentration. If $n \approx 10^7 \text{ cm}^{-3}$, then $t_s \approx 0.2 \text{ s}$ and if $n \approx 5 \times 10^5 \text{ cm}^{-3}$, $t_s \approx 4 \text{ s}$. An electric field of about $20 \text{ kV}\cdot\text{m}^{-1}$ brings Eq. (3) into general agreement with the empirical relationship, Eq. (2).

Droplet and raindrop charges observed under natural conditions generally fall within the band predicted by using the limits of $10^2 \text{ V}\cdot\text{m}^{-1}$ and $10^6 \text{ V}\cdot\text{m}^{-1}$ in Eq. (3). This steeply sloping band, centered on the line obtained with $10 \text{ kV}\cdot\text{m}^{-1}$, is displayed in Fig. 4. This band lies below the Rayleigh limit for disruption, $(Q/e)_R = e^{-1}(32\pi^2\epsilon_0 T_g R^3)^{0.5}$, where $T_g = 0.076 \text{ N}\cdot\text{m}^{-1}$ is the surface tension of water.

Contrast these observations and predictions with the situation to be expected under steady-state conditions in a bipolar ionic atmosphere similar to those described earlier. The number of charges per droplet ought to be distributed according to the Boltzmann distribution proposed by Keefe, et al.⁶ provided that the effects of gravitational settling and external fields are suppressed. The more general case for which the ionic mobilities are unequal and electric fields are present is described by Mead's general formula⁷ but

Fig. 4. Mean number of droplet charges vs droplet radius in a unipolar or a bipolar ionic environment. Lines L, M, and U represent the saturation charge acquired by unipolar charging with electric fields of 10^2 , 10^4 , and $10^6 \text{ V}\cdot\text{m}^{-1}$, respectively. Line M resembles the mean charge vs radius relationship which has been empirically determined for natural thunderclouds. Curves N and P represent the mean number of negative and of positive charges per droplet to be expected in a bipolar ionic atmosphere permeated by an electric field of $10^2 \text{ V}\cdot\text{m}^{-1}$. All the curves are below the Rayleigh disruption limit, line R.

with the intrinsic "particle" activity set to zero (see Appendix E). These relationships predict the number of steady-state droplet and raindrop charges shown by the more gently rising line in Fig. 4. Notice that a 10 μ m-radius droplet is predicted to have an average about 10 (negative) charges in such an atmosphere. This value is more than two orders of magnitude less than that predicted for the natural cloud environment. The principal physical reason for this reduction is the high bipolar ionization which provides such a high conductivity that larger charges cannot be sustained for any appreciable length of time.

Actual droplet charges in the nuclear debris-cloud environment probably lie somewhere between the unipolar and bipolar limits. Such distributions can be determined only after the various dynamical charge transfer and charge transport mechanisms in clouds have been identified and specified. Then the droplet charge distribution, the ion concentrations, and the steady-state electric field all must be determined simultaneously and self-consistently in accordance with the Maxwell equations. An attempt in this direction will be described subsequently.

ELECTRIC FIELD INTENSITIES

Spectacular lightning discharges vividly attest to the occurrence of extremely large environmental electric fields. In principal, such fields could greatly enhance the attachment even of slightly charged aerosols to cloud droplets. Consequently, it is very important to establish the magnitudes of any electric fields that could be present in an environment containing nuclear debris and natural clouds.

Electric fields have been observed in the neighborhood of nuclear explosions. Perturbations in the Earth's electric field following low-altitude (< 1 km) nuclear detonations (15-30 kT) were observed in 1952 at the Nevada Test Site and discussed by Holzer.¹⁴ From a distance of about 8 km, he observed a very sharp initial decrease of 80-100% in the downward-directed surface electric field at the time of detonation. The field recovered its ambient value after 2-3 minutes, but then continued to increase 20% more passing through a maximum value at about 5 minutes, and finally gradually decayed over the next 10-20 minutes back to the preshot ambient value. An example of Holzer's data is displayed in Fig. 5.

Holzer analyzed his field data in terms of an "electric-dipole model" in which the perturbations in the ambient field were attributed to a downward-directed electric dipole of magnitude 1 C $\cdot$ km, which was centered on the drifting debris cloud. This dipole was presumed to be caused by asymmetries in the displacement of charge, which was generated by Compton scattering of delayed gamma rays from fission fragments. Unfortunately, Hill's¹⁵ and Holzer's theoretical estimates of the dipole moment generated by this mechanism are several orders of magnitude smaller than the observed

Electrostatic Field Measurement at NTS (Holzer)

Fig. 5 Perturbations in the earth's electric field vs time as recorded by Holzer at a station 7.8 km from a nuclear explosion at the Nevada Test Site in 1952. The sharp drop at the time of detonation is followed by a steady recovery over the next several minutes which, however, surpasses the preshot ambient value, passes through a maximum value at about 5 minutes, and then gradually decays back to the ambient value.

value, and furthermore, decrease with time (in contradiction with the observations). Both Holzer and Hill mention, but then dismiss the possibility that the dipole could have been induced upon the large, nearly hemispherical, highly ionized region which surrounds the debris cloud, by the small ($\sim 40 \text{ V}\cdot\text{m}^{-1}$) ambient preshot electric field. Initially such a configuration would resemble an idealized conducting, hemispherical "boss" superimposed on infinite ground plane.

Holland¹⁶ has revived a variant of this "boss model". He argues that the spatial extent and temporal development of this highly conducting region can account for the qualitative features displayed in Fig. 5. Holland defines the conducting region to be that volume within which the ion generation rate is sufficiently greater (100X) than the ambient background value (of $10 \text{ cm}^{-3}\cdot\text{s}^{-1}$) in the absence of nuclear debris and, simultaneously, within which the electrical relaxation time (which is an inverse measure of the medium's conductivity) is shorter than the time scale ($\sim 10 \text{ s}$) over which there are appreciable changes in the debris-cloud geometry. Needless to say, this does not define a sharp boundary. At early times ($\sim 1 \text{ s}$) the region extends almost out to Holzer's nearest observation station ($\sim 8 \text{ km}$). The conducting, slightly (prolate) hemispheroidal boss concentrates electric field lines on its surface and causes a deficiency of field lines (which implies a reduction in the electric intensity) nearby along the conducting plane.¹⁷ As time progresses, the boss increases its height as it follows the rising debris cloud. After 2-3 minutes, this region becomes detached from the ground plane and its top expands horizontally because of the rapid increase in gamma-ray mean-free-path with altitude. After 5 minutes this large, extended region has improved its electrical connection with the more

conducting upper atmosphere (above 20 km) and may be represented by a large inverted (oblate) hemispheroidal boss extending downwards from this "conducting plane." By this time Holzer's observation stations were beneath and well encircled by this new boss. The reduction in distance between the "plates" of this giant capacitor (composed of the inverted boss and the ground plane) in comparison with the preshot separation, accounts for the enhancement in the potential gradient at these late times. Figures 6a and 6b depict these two limiting situations.

The boss model can be given additional credence by exhibiting isochrones of constant electrical relaxation time for a sequence of times after detonation. Figure 7 contains such a sequence of snapshots. Notice the time evolution of the various isochrones from closed to open contours as the center of the debris cloud rises. The contours in these snapshots have been calculated by using an elementary absorption theory (with a simple exponential build-up factor) from a monoenergetic point source, but with temperature and density variations based upon the standard atmosphere. The procedure which was used is described in Appendix F.

This "boss-model" provides a qualitative picture which does account for the early-time depression and the late-time enhancement of the original ambient field. A more quantitative comparison is difficult to achieve because, as Holland points out, the conductivity varies with altitude and the electric potential gradient is not uniform. Indeed, the ambient electric field rapidly (almost exponentially) decreases from its surface value to zero beyond approximately 20 km. Nevertheless, within the same spirit as earlier estimates of dipole moments, it is possible to apply the well-known expressions¹⁷ for the electrostatic potential outside an isolated,

Figs. 6a, b Schematic diagrams showing how the time-evaluation of Holland's boss model can be used to explain Holzer's observations. The large, conducting region, which surrounds the debris cloud, contains ionization created by the long-range (low probability) penetration of delayed gammas from fission fragments. At 1 s after detonation, there is a hemispheroidal boss on the ground plane. The charge induced on this boss by the ambient field concentrates electric field lines on the boss's surface and causes a reduction in field intensity at the neighboring observation stations. After 5 minutes (Fig. 6b), the conducting region no longer makes contact with the ground and resembles a large inverted boss extending below the "conducting upper atmospheric plane." Its radial extent is much larger because the gammas can penetrate farther in this rarefied atmosphere. The effective distance between this boss and the ground plane is less than normal causing the potential gradient at the observation stations to exceed the ambient value.

Boss Model

(Holland)

Fig. 6a

Fig. 6b

Fig. 7a-f Sequence of snapshots showing the evolution of the isochronal surfaces of constant electrical relaxation time. The altitude of the debris cloud, which is denoted by an X on the ordinate, varies from 0.3 km to 11.2 km as time evolves from 0 to 600 s. The positions of Holzer's observation stations, relative to the location of the drifting debris cloud, are denoted by X's along the abscissa. Isochrons of 1, 3, 10, 30, and 100 s are shown. The 10 s isochron is taken as representative of the extent of the conducting region which surrounds the debris cloud. Initially, its shape approximates a prolate hemispheroidal boss on the ground plane. After 120 s, its top has opened; after 180 s the bottom ceases to contact the ground and continues to rise thereafter.

Fig. 7a-f

hemispheroidal boss on an infinite conducting plane with a uniform field at infinity to estimate the electric field at Holzer's observation stations. Even such crude estimates are within a factor of 2 of the observed values. These estimates are described in Appendix G.

The presence of extremely high conductivity in the neighborhood of a debris cloud is an essential ingredient in this explanation of Holzer's observations. The observed effective dipole moment is a consequence of the charge distribution that has been induced on the "surface" of a conductor by an external field rather than as the result of gross charge separation within the debris cloud. Since electric fields cannot be sustained without conductors, there should not be any compulsion to infer the existence of a strong static internal electric field within a debris cloud. Nevertheless, one may still ask whether or not the field generation mechanisms within natural thunderclouds are sufficiently strong to overcome the effects of high ionization.

Naturally, occurring atmospheric electric field intensities generally are expected to range from the Earth-directed fair-weather field of $120 \text{ V}\cdot\text{m}^{-1}$ up to the breakdown potential gradient of $3 \text{ MV}\cdot\text{m}^{-1}$ for dry air. Typical cumulus and shower clouds develop fields of 1 to $10 \text{ kV}\cdot\text{m}^{-1}$, whereas those observed in thunderclouds reach $400 \text{ kV}\cdot\text{m}^{-1}$ and may exceed $1 \text{ MV}\cdot\text{m}^{-1}$ within small, fluctuating pockets. There is an abrupt, several-fold increase in field strength upon flying into an electrified cloud. This is attributed to the formation of a screening layer at the interface between the cloud and the surrounding clear air. Ions, being drawn toward the electrified cloud, become attached to the much more massive cloud droplets. These droplets move much more slowly, so the electrical conductivity within the cloud is reduced greatly.

The magnitude of any electric field within a cloud must be inversely proportional to the internal electrical conductivity. When the ion production-rate is low ($\sim 10 \text{ cm}^{-3} \cdot \text{s}^{-1}$ from natural radioactivity) then the steady-state ion concentration, and hence also the conductivity will be low (see Fig. 1). This low conductivity permits the growth of strong fields (by mechanisms to be discussed subsequently). When onset fields ($> 400 \text{ kV} \cdot \text{m}^{-1}$) required for corona discharge from ice crystals have been attained, the ion production-rate rises to $\sim 5 \times 10^5 \text{ cm}^{-3} \cdot \text{s}^{-1}$ with a corresponding, perhaps sudden, increase in the conductivity and reduction in the field. However, notice (see Fig. 1) that the ion production-rates expected from radioactive nuclear debris may greatly exceed this limit and the resulting steady-state ion concentrations generally are unaffected by the presence even of rather large numbers of collecting droplets (also see Appendix C). These considerations appear to place severe restrictions on the growth of any electric fields in the presence of such large volume ionization rates.

There is not agreement on the mechanism(s) by which large fields are generated in thunderstorms. Favored theories in recent years involve the charge separation produced by charged solid precipitation falling away from oppositely charged cloud droplets that move with the updraft. The charging of these two types of objects with opposite signs arises from collisions of cloud droplets with precipitation hydrometeors. Mechanisms proposed for this collision charging include an inductive mechanism involving polarization of the hydrometeor by the ambient electric field and several noninductive

mechanisms involving differential charge transport along a thermal gradient or differences in the work functions of ice surfaces.

The details of various simple models based on precipitation theories of electrification have been worked out. (A general survey describing a number of these models have been given by Mason¹⁸ and by Moore and Vonnegut.¹⁹) Starting from a fair weather field of the order of $10^2 \text{ V}\cdot\text{m}^{-1}$, these models yield fields greater than $10^5 \text{ V}\cdot\text{m}^{-1}$ in times of from 50 to 2000 s depending on the particular values of various parameters that are used. With few exceptions, most precipitation-theory models of thunderstorm electrification have in common the assumption of an infinite cloud with neither horizontal nor vertical spatial dependence of its parameters. The electric field and the droplet and hydrometer velocities are assumed to have only vertical components. The evolution of the electric field in time is obtained by solving

$$dE/dt = - J/\epsilon_0, \quad (4)$$

where the total current density, J , has contributions from the motions of charged droplets, charged hydrometeors, and in some cases ions.

Spangler and Rosenkilde²⁰ have assessed the effect of high ion production rates on electric field growth. Their estimate of the ion concentration that might be expected to prevent field growth is described in Appendix H. This estimate is based upon a comparison between the generator current inferred to be in actual thunderstorms and the ion conduction current anticipated to be present in a debris cloud. It suggests that even under the most favorable conditions, the natural field generation mechanisms, whatever

they may be, will be suppressed (indeed, shorted-out!) if the steady-state ion concentration should exceed a value of the order of 10^4 cm^{-3} . An examination of Fig. 1 shows that this value is roughly three orders of magnitude less than that to be expected in a nuclear debris cloud and inferred from the foregoing explanation of Holzer's observations.

The associated ion generation-rate can be inferred most easily from Fig. 8, which is a different representation of the information contained in Fig. 1. It shows how the steady-state ion-concentration increases with generation rate along curves of constant ion attachment rate. Estimates in Appendix C show that it is difficult to drive this attachment rate much above 10 s^{-1} , even under the most favorable conditions in the strongest possible electric fields [see Eq. (C8)]. This implies that an ion generation rate exceeding $10^6 \text{ cm}^{-3} \cdot \text{s}^{-1}$ certainly ought to prevent the growth of electric fields in thunderclouds since this value corresponds to a steady-state ion concentration of $5 \times 10^4 \text{ cm}^{-3}$ under these conditions. Typical generation rates in debris clouds are expected to be several orders of magnitude greater than this.

Spangler²¹ showed that these estimates were consistent with the predictions of a more detailed cloud electrification model. He extended a simple, infinite cloud, precipitation theory model to include effects due to the presence of radioactive aerosol particles. The cloud was assumed to be spatially homogeneous. The electric field development was governed by Eq. (4) together with an additional set of eleven coupled first-order ordinary differential equations which govern the time development of positive and negative ion concentrations and charged and neutral hydrometeor and droplet concentrations. Both inductive and noninductive charging mechanisms were examined. Figure 9 shows examples of the field growth with time when the

Fig. 8. The steady-state ion concentration in a debris-cloud environment plotted as a function of the ion production rate for various values of the ion attachment rate (in s^{-1}) of ions to cloud droplets or particles. Electric field growth is inhibited (shorted out) for debris cloud environments outside the dashed square.

Fig. 9 Development of the electric-field strength with time when only the inductive charge-separation mechanism is operative.

The ion-generation rates and hydrometeor number densities for the various curves are given in the following table.

Curve	Ion-Generation Rate $s \text{ (cm}^{-3} \text{ s}^{-1})$	Number Density of Hydrometeors (cm^{-3})
1	0	10^{-3}
2	9.99×10^2	10^{-3}
3	9.99×10^3	10^{-3}
4	9.99×10^3	10^{-4}
5	3.17×10^4	10^{-3}
6	5.64×10^4	10^{-3}
7	9.99×10^4	10^{-4}
8	9.99×10^4	10^{-3}

inductive charging mechanism was operative. In these examples, growth is prohibited if the ion generation rate, s , exceeds $10^5 \text{ cm}^{-3} \cdot \text{s}^{-1}$.

Other quantities of interest, such as ion and droplet concentrations and droplet charges, are computed self-consistently within Spangler's charging model. Figures 10-12 display the time development of these quantities for three separate cases: (1) $s = 0$, (2) s near but less than the maximum value consistent with field growth, and (3) s near but greater than the minimum value that prevents field growth. These figures show that field growth is associated with increase in the magnitudes of the hydrometeor and collected-droplet charges, that field growth terminates when the hydrometeor is levitated, and the field growth is prohibited in cases where the ion density remains at consistently high values. In this third case, note that the droplets have only 10 to 20 charges rather than numbers exceeding 10^4 as suggested in the first case.

Some noninductive charging mechanisms also were investigated and found to be slightly more effective than the inductive mechanisms, but in all cases field growth could be prohibited if s exceeded $10^6 \text{ cm}^{-3} \cdot \text{s}^{-1}$.

In "short," strong electric fields are not expected to be present within fresh nuclear debris clouds because the high radioactive decay rate generates so many ions that the interior remains a very good conductor within which electric fields can neither be generated nor sustained. Electric field growth in the presence of nuclear debris appears to be possible only if the material can be diluted sufficiently to reduce the steady-state ion concentration below the effective limits estimated herein.

A postscript needs to be appended here. The use of nuclear debris is not being advocated here under any circumstances, as a method, practical or

otherwise, to "short-out" a thunderstorm. However, there have been other proposals to modify the conductivity within thunderstorms. Kasemir and Weickmann²² suggested the release of large quantities of thin, conducting fibers into the updrafts near the bottom of a thundercloud. The electric fields induced near the fine tips of these fibers should be strong enough to ionize the air. Vonnegut et al.²³ showed that convective clouds may be primed by releasing artificially produced point-discharge ions into the subcloud air.

Figs. 10-12 These figures illustrate the time development of various quantities when an inductive charge-separation mechanism is operative. The quantities shown are (1) electric field strength, (2) hydrometeor velocity, (3) droplet number density, (4) droplet charge, (5) hydrometeor charge, and (6) the sum and the difference of the positive and negative ion number densities. All curves are for hydrometeor concentrations of 10^{-3} cm^3 . The ion-generation rates, which are given in the following table, are for the cases $s = 0$, s near but less than the maximum value consistent with field growth, and s near but greater than the minimum value that prevents field growth. In the droplet number density and droplet charge graphs, solid lines are for virgin droplets, lines of short dashes are for once-collided droplets, and lines of long dashes are for twice-collided droplets.

Figure	Ion-Generation Rate $s \text{ (cm}^{-3} \cdot \text{s}^{-1})$	Charge-Separation Mechanism
10	0	inductive
11	3.14×10^4	inductive
12	9.99×10^4	inductive

Fig. 10

Fig. 11

Fig. 12

ELECTRICAL SCAVENGING OF NUCLEAR DEBRIS

Precipitation scavenging of nuclear debris aerosols ultimately requires their incorporation into falling raindrops. This can be accomplished by direct interaction of particles with raindrops, as must be the case for below-cloud washout. Within a cloud, several other less direct, but overall possibly more efficient, multi-stage processes also are available. The overall removal rate for such a multi-stage process is determined principally by the slowest single-process rate in the entire sequence.²⁴

Of particular interest here are two-stage removal processes in which submicron particles are first incorporated into cloud droplets with a rate coefficient, Λ_1 , and then the cloud droplets are accreted by raindrops with a rate coefficient, Λ_2 . The total concentration of particles suspended in the cloud (i.e., those freely suspended plus those already attached to droplets) decreases with time at the rate

$$dN/dt = -N_0(\Lambda_1^{-1} - \Lambda_2^{-1})^{-1}[\exp(-\Lambda_1 t) - \exp(-\Lambda_2 t)], \quad (5)$$

where N_0 is the initial particle concentration. The accretion rate coefficient, Λ_2 , for typical cloud droplet populations is normally in the range 10^{-3} to 10^{-4} s^{-1} with the variation being primarily dependent on the rain rate. Unless Λ_1 is also in (or exceeds) this range, the rate of incorporation of aerosol particles into cloud droplets will determine the overall removal rate.

There are essentially two classes of possible interactions between aerosol particles and cloud water: nucleation and attachment. Interest centers here

on attachment processes, and in particular, those which involve direct electrical interactions with cloud droplets. Several other processes including inertial capture, Brownian-, convective-, and turbulent-diffusion, diffusio- and thermo-phoresis, as well as some electrical interactions, were compared by Williams.² Of these, the electrical processes appeared to contribute the largest rate coefficient for the particular parameters which were chosen. However, uncertainties in the values of the various parameters, particularly those values to be expected in a highly radioactive environment, have prompted the foregoing examination of the electrical environment in which rainout may occur.

On the basis of the foregoing examination of the electrical aspects of a highly radioactive environment, it is now possible to give answers to the first four questions, which were posed initially. (1) The extremely high, early-time ($< H + 1$ hr) radioactivity of nuclear debris creates so much ionization that the resulting steady-state ion concentration essentially is independent of the presence of any natural cloud constituents (see Fig. 1). Ambient ion concentrations in the range 10^5 to 10^7 cm^{-3} are expected. (2) The effective charge on nuclear debris with radii from 0.01 to 0.1 μm follows a Boltzmann-like distribution with a slightly negative, but nearly zero mean value; the effective charge on debris with radii from 0.1 to 1.0 μm is driven positive by the beta decay with magnitudes, which are proportional to particle volume and which approach 10^3 charges for the largest particles (see Fig. 3). (3) Cloud droplets with radii around 10 μm acquire 10 to 20 charges on the average, but cannot accumulate typical thunderstorm charges in excess of 10^3 because the high ion concentration is

bipolar (see Fig. 4). (4) The electric field perturbations, which were observed near nuclear explosions, are consistent with the existence of a very large diameter (> 5 km) region of high electrical conductivity, which is 10 to 10^4 times the natural value (see Figs. 7a). This electrical conductivity is sufficient to hold thunderstorm electric fields below $10^2 \text{ V}\cdot\text{m}^{-1}$ by effectively short-circuiting the natural charge-separation mechanisms, which otherwise could generate fields in excess of $10^5 \text{ V}\cdot\text{m}^{-1}$ (see Fig. 9).

Now it is possible to re-examine the final issue which concerns the scavenging rate of nuclear debris by cloud droplets in this electrical environment. Two distinct physical models commonly are used to describe the scavenging of aerosols by droplets: (1) a discrete-collision model and (2) a continuum-flux model. The collision model has been applied most successfully to the collection of particles of the order of $1 \text{ }\mu\text{m}$ or larger. In this range, the particle mass is sufficiently large that the mediating air represents a relatively small perturbation on the particle trajectory and it is sensible to follow the trajectories of individual particles as they move relative to a droplet in accordance with the laws of Newtonian mechanics. The flux model is used widely to describe scavenging of an ensemble of particles of the order of $0.1 \text{ }\mu\text{m}$ or smaller. In this range, the particle mass is sufficiently small that the many random collisions with air molecules, produce large angle deflections on a microscopic scale and make it impractical to follow individual trajectories. However, on a macroscopic scale the mediating air can be regarded as a continuum through which the ensemble drifts with both random and deterministic velocity components which behave in accordance with the laws continuum mechanics. Both models, commonly are extrapolated beyond

their regimes of successful applicability into the intermediate regime and probably represent reasonable first approximations in that domain. These models, as well as many other aspects of scavenging, are discussed, both generally and in considerable detail, in the recent book by Pruppacher and Klett.²⁵

A continuum-flux model will be adopted for the present discussion. The thermal motion of debris aerosol is independent of electrical effects so Brownian diffusion still is operative. Gravitational settling of 10- μ m radius cloud droplets still dominates over electrical levitation if the droplet possesses < 20 charges and is subjected to electric fields $< 10^3$ V $\cdot$ m⁻¹. Consequently, the Brownian diffusion rate should be augmented in order to account for the effective thickness of the diffusion boundary-layer which is caused by the convection of air past the settling droplet. Electrical interactions should include: (1) the direct, spherically symmetric, attraction between the charged debris and charged droplets and (2) the "convective-like" drift of charged particles along those externally-applied electric-field lines which enter the polarized droplet. (Additional complications caused by image forces, polarization, and dielectrical phenomena will be omitted.) Diffusio- and thermo-phoretic forces should be included if the relative humidity is below 100 percent, but also will be omitted here for the sake of simplicity.

These choices lead one to specify a particle flux which consists of two parts: (i) a contribution from Brownian diffusion and (ii) a contribution from conductive drift under the influence of electrical forces. (The precise form of this flux is described in Appendix I.) Steady-state continuity of

this flux leads to a form of the convective-diffusion equation which must be solved in order to determine the particle concentration field in the neighborhood of the droplet. Once this concentration field has been determined, it is possible to integrate the resulting flux over the droplet surface in order to obtain the desired collection (or scavenging) current.

It is customary to express this current as a collection kernel, K , which represents the collection rate per unit collector concentration. The product of this kernel with the collector concentration, C , gives the scavenging-rate coefficient, Λ . This rate coefficient determines the decrease in the particle concentration with time according to the definition:

$$- dN/dt \equiv \Lambda N = (KC)N, \quad (6)$$

in the simple case where the distributions are monodisperse. (A suitable extension of this description covers more general distributions.)

There are two electrical regimes to be compared in the present case: (1) low particle and droplet charges in low electric fields, and (2) low particle, but high droplet charges in high electric fields. On the basis of considerations which are given in Appendix I, the collection kernels in each of these two regimes assume the forms:

$$\text{case (1)} \quad K \sim K_b \cdot f_g \cdot (Sh)_e \quad (7)$$

$$\text{case (2)} \quad K \sim K_g \cdot (Ef)_E \quad (8)$$

The precise expressions for the various factors are given in Appendix I. However, a general description will be given here.

In the first case, where the ionization is so high that particle and droplet charges are low and high electric fields cannot be sustained, the collection kernel in Eq. (7) consists of three factors. The first factor K_b , is the collection kernel for pure thermal (or Brownian) diffusion to a stationary sphere. The remaining two nondimensional factors represent enhancements. The ventilation factor, f_g , accounts for the enhancement due to air flowing past a settling droplet. The Sherwood number, $(Sh)_e$, contains the enhancements, which are due to the electrical interactions between the particle and droplet charges and due to the conductive drift of the charged particle toward the polarized droplet.

In the second case, where the ionization is low enough so that droplet charges and electric fields can grow in magnitude, the collection kernel in Eq. (8) consists of two factors. The first factor, K_g , represents the collection kernel for purely "geometric" (and hence, fictitious) collisions with the settling droplet. (They are fictitious because submicron aerosols are swept easily around the collector with the flowing air and rarely make contact with it.) The second factor is an electrical collection efficiency, $(Ef)_E$, which accounts for the adjustment in the collision cross-section because externally-applied electric field-lines are concentrated by the polarized droplet.

The general effect of these factors may be seen in Fig. 13. Curves labeled B and C represent thermal diffusion and its enhancement by convection of air past the droplet. Curves L and H represent the enhancement obtained in curve C when the particle charges, which were derived for low (L) and high (H) activity from Figs. (2) and (3), are used together with a nominal terrestrial field of $10^2 \text{ V}\cdot\text{m}^{-1}$. Even for the highest activity, the collection

Fig. 13 Various collection kernels as a function of debris particle radius. The collector droplet radius is 10 μm . The curves are labeled according to the following table.

Curve	Collection Mechanisms Operating
B	Thermal (Brownian) diffusion
C	Thermal + convective diffusion
L	Thermal + convective + conductive diffusion in $10^2 \text{ V}\cdot\text{m}^{-1}$ field with low particle activity of $10 \text{ Bq}\cdot\mu\text{m}^{-3}$
H	Thermal + convective + conductive diffusion in $10^2 \text{ V}\cdot\text{m}^{-1}$ field with high particle activity of $10^5 \text{ Bq}\cdot\mu\text{m}^{-3}$
G	Fictitious geometric collection
E	Conductive collection in $300 \text{ kV}\cdot\text{m}^{-1}$ field with saturated droplet charges.

Fig. 13

kernel remains less than $5 \times 10^{-7} \text{ cm}^3 \cdot \text{s}^{-1}$ through most of the particle size-range of interest. Curve G represents the fictitious geometric collection kernel. However, ratios between all the other curves and this curve determine the associated collection efficiencies. Curve E represents the electrical collection by droplets, which have been charged to saturation (see Fig. 4), in a field of $300 \text{ kV} \cdot \text{m}^{-1}$. This situation corresponds to the one considered earlier by Williams.² In this case the collection kernel remains high above $5 \times 10^{-6} \text{ cm}^3 \cdot \text{s}^{-1}$.

This figure clearly illustrates that the high ionization in a nuclear debris cloud ultimately leads to a great reduction in that component of the scavenging which is due to electrical interactions. High radioactivity produces high, bipolar, ionization. The environment, which surrounds the nuclear debris, becomes such a good conductor that large droplet charges and strong electric fields cannot be sustained. So, even though some of the largest debris particles may achieve fairly high positive charge (because of their beta decay), their overall collection by droplets is depressed significantly in comparison with the natural thunderstorm environment.

SUMMARY AND CONCLUSIONS

Rainout - the in-cloud scavenging of airborne contaminants by precipitation - proceeds via a number of physical mechanisms. Those processes which involve electrical aspects are scrutinized in this report for their potential susceptibility to the intimate presence of fresh radioactive nuclear debris. These radioactive particles emit beta- and gamma-rays which copiously ionize the surrounding air. This high ionization rate distinguishes the electrical character of a nuclear-debris cloud environment from that of a natural thundercloud and drives the distinctions being elucidated in this report.

The extremely high, early time ($< H + 1$ hr) radioactivity of the nuclear debris creates so much ionization that the resulting steady-state ion concentration essentially is independent of the presence of any natural cloud constituents. Ambient steady-state bipolar ion concentrations in the range of 10^5 to 10^7 cm^{-3} are expected. The beta decay rate of this nuclear debris is sufficiently rapid that those debris aerosols with radii from 0.1 to $1.0 \text{ }\mu\text{m}$ may be left with a positive effective charge, which is proportional to particle volume and which may approach 10^3 charges for the largest such aerosols. On the other hand, cloud droplets with radii around $10 \text{ }\mu\text{m}$ can acquire only 10 to 20 charges on the average and cannot accumulate typical thunderstorm charges in excess of 10^3 because the high ion concentration is bipolar. The electrical conductivity of this weak plasma, which is 10 to 10^4 times the natural value, is sufficient to hold thunderstorm electric fields below $10^2 \text{ V}\cdot\text{m}^{-1}$ by effectively short-circuiting the natural charge-separation mechanisms, which otherwise could generate fields in excess of $10^5 \text{ V}\cdot\text{m}^{-1}$.

Thus, high radioactivity produces high, bipolar, ionization. The environment, which surrounds the nuclear debris, becomes such a good conductor that large droplet charges and strong electric fields cannot be sustained. Consequently, electrical interactions between the charged debris particles and these droplets in these electric fields are greatly reduced in effectiveness. So, even though some of the largest debris particles may achieve fairly high positive charge (because of their beta decay), their overall collection by droplets is depressed significantly in comparison with the natural thunderstorm environment. In short, the high ionization in a nuclear debris cloud ultimately leads to a great reduction in that component of the scavenging which is due to electrical interactions.

We hasten to add that electrical interactions probably are not the only mechanisms that contribute to scavenging of nuclear debris in a potential rainout situation, although they once were thought² to be the largest factor. Phoretic forces, which depend upon the relative humidity, and turbulence, which is generated around convective cells, still could contribute their share because both of these effects are gas- or fluid-dynamical and, therefore, are expected to be less sensitive to the intimate presence of radioactivity and its attendant ionization. Neither of these mechanisms have been included in the estimates contained in this report.

Nonetheless, electrical interactions do play a major role in the scavenging of nonradioactive aerosols, including the surrogate debris, which was introduced with chemical tracers into dynamical storm systems such as those studied around St. Louis.²⁵ Consequently, great care should be taken in extrapolating to a debris-cloud environment the results of such chemical-tracer experiments conducted in the natural environment. Rapid

scavenging in the natural environment does not necessarily mean that rapid scavenging could be expected in a debris-cloud environment, especially if electrical interactions are the most likely scavenging mechanisms.

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APPENDIX A

ATMOSPHERIC PARAMETERS

Principal interest here centers on the properties of the lower troposphere where the phenomena commonly associated with the weather occur. Various parameters such as temperature, concentration, density, pressure, molecular mean free path, and viscosity are needed for subsequent estimates. Representative values for these quantities and their variation with altitude are listed in Table A-1. These values are based upon the idealized standard atmosphere.^{A1}

A1. U. S. Standard Atmosphere, 1962 (U. S. Government Printing Office, Washington, DC, December, 1962).

Table A-1. Standard atmospheric data

Parameter	Altitude (km)					
	0	2	4	6	8	10
Temperature ($^{\circ}\text{K}$)	288	275	262	249	236	233
Concentration (nm^{-3})	.0255	.0209	.0170	.0137	.0109	.0086
Density ($\text{kg}\cdot\text{m}^{-3}$)	1.23	1.01	0.819	0.660	0.526	0.414
Pressure (kPa)	101.3	79.5	61.6	47.2	35.7	26.5
Saturated vapor pressure (Pa)	1700	706	265	70.4	18.1	3.9
Molecular mean free path (nm)	66.3	80.7	99.2	123	154	196
Viscosity ($\mu\text{Pa}\cdot\text{s}$)	17.9	17.3	16.6	15.9	15.3	14.6
Kinematic viscosity ($\text{nm}^2\cdot\text{s}^{-1}$)	14.6	17.1	20.3	24.1	29.1	35.3

APPENDIX B

FATE OF FREE ELECTRONS

Estimates of the concentration and the transport coefficients of free electrons are obtained here. Primary electrons collide with gas molecules to form positive ions and secondary electrons. Negative ions are formed by attachment of these secondary electrons to neutral gas molecules at low energy in the presence of ions, particles, and cloud droplets. Some of these attached electrons may become detached again because of collisions between the negative ions and neutral molecules.

The concentration, n_e , must satisfy a rate equation of the form

$$\dot{n}_e = s_e + \alpha_e'' n n_0 - (\alpha_e' n_0 + \alpha_e' p + \beta_e N + \gamma_e C) n_e, \quad (B1)$$

where s_e is the electron production rate per unit volume, $\alpha_e'' n n_0$ is the collisional detachment rate per unit volume, $\alpha_e' n_0$ represents the attachment rate to neutral molecules, $\alpha_e' p$ represents the attachment rate to positive ions, $\beta_e N$ represents the attachment rate to both neutral and charged particles, and $\gamma_e C$ represents the attachment rate to cloud droplets. (Summations over individual charge states have been suppressed.)

Electron attachment to gas molecules at tropospheric densities occurs via three-body processes. The principal reactions enumerated by Phelps^{B1} all generate O_2^- . The effective attachment rate at an altitude of 2 km is taken to be $\alpha_e' n_0 \approx 3.7 \times 10^7 \text{ s}^{-1}$. The principal reactions involved in electron-ion combination have been enumerated by Biondi.^{B2} The effective combination coefficient (at 2 km) is taken to be $\alpha_e' \approx 5 \times 10^7 \text{ s}^{-1}$,

which is the appropriate weighted sum of the various two- and three-body contributions. Collisional detachment results principally from interactions between O_2^- and neutral oxygen and nitrogen molecules, with an overall rate, $a_e' n_0 = 1.5 \text{ s}^{-1}$.

The attachment of electrons to particles and to droplets depends upon the electron diffusion coefficient in air. It is known that the drift velocity, v_e , the mobility, $\mu_e = v_e E^{-1}$, and the diffusion coefficient, D_e , all depend upon the strength of any applied electric field, E . Estimates for these quantities can be obtained by first constructing suitable interpolation formulas which fit the data summarized by Huxley and Crompton^{B3} and others.^{B4-B6} The following empirical expressions obtain:

$$v_e = (10 \text{ km} \cdot \text{s}^{-1}) \{4.06 y + 1.33 y^2\}^{0.5} \quad (B2)$$

and

$$D_e \mu_e^{-1} = (0.929 \text{ V}) y^{0.75} \quad (B3)$$

where y is the nondimensional ratio

$$y = (E/10^2 \text{ kV} \cdot \text{m}^{-1}) (10^{19} \text{ cm}^{-3}/n_0) \quad (B4)$$

and n_0 is the neutral gas concentration. Equation (B3) is applicable for $y < 1$. Table B-1 contains some representative values of these quantities together with an estimated electron mean free path, λ_e , at an altitude of 2 km.

An estimate for the attachment coefficient between electrons and a spherical collector of radius R can be made by using the expression, $4\pi R D_e$, provided that $R \gg \lambda_e$. For an atmosphere field of $10^2 \text{ V}\cdot\text{cm}^{-1}$, $D_e \approx 130 \text{ cm}^2\cdot\text{s}^{-1}$ and $\lambda_e \approx 0.7 \text{ }\mu\text{m}$. Thus $\beta_e \approx 0.16 \text{ cm}^3\cdot\text{s}^{-1}$ for $1\text{-}\mu\text{m}$ particles and $\gamma_e \approx 1.6 \text{ cm}^3\cdot\text{s}^{-1}$ for $10\text{-}\mu\text{m}$ droplets.

A steady-state electron concentration can now be estimated. As an example, suppose that $s_e \approx 10^{10} \text{ cm}^{-3}\cdot\text{s}^{-1}$, $n \approx p \approx 8 \times 10^7 \text{ cm}^{-3}$, $N \approx 10^4 \text{ cm}^{-3}$, and $C \approx 300 \text{ cm}^{-3}$. Then $\alpha'_e p \approx 40 \text{ s}^{-1}$, $\beta_e N \approx 1.6 \times 10^3 \text{ s}^{-1}$, and $\gamma_e C \approx 480 \text{ s}^{-1}$ all of which are small compared with the attachment rate to neutral molecules, $\alpha_e n_0 \approx 3.7 \times 10^7 \text{ s}^{-1}$, and moreover, $\alpha''_e n n_0 \approx 1.8 \times 10^8 \text{ s}^{-1}$ is small compared with s_e . Under these conditions, an equilibrium concentration

$$n_e \sim s_e (\alpha_e n_0)^{-1} \sim 270 \text{ cm}^{-3}$$

is attained over a characteristic time-scale, $\tau_e \sim n_e s_e^{-1} \sim 27 \text{ ns}$.

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Table B-1. Representative values for the drift velocity, v_e , the mobility, μ_e , the diffusion coefficient, D_e , and the mean free path, λ_e , of an electron in air at an altitude of about 2 km for various electric fields, E , of possible interest.

	$E \text{ (V} \cdot \text{m}^{-1}\text{)}$				
	10^2	10^3	10^4	10^5	10^6
$v_e \text{ (km} \cdot \text{s}^{-1}\text{)}$	.44	1.4	4.4	1.5	7.1
$\mu_e \text{ (m}^2 \cdot \text{kV}^{-1} \cdot \text{s}^{-1}\text{)}$	4400	1400	440	150	71
$D_e \text{ (m}^2 \cdot \text{ks}^{-1}\text{)}$	13	24	42	80	210
$\lambda_e \text{ (}\mu\text{m)}$	.38	.69	1.2	2.3	6.2

APPENDIX C AMBIENT ION CONCENTRATION

Factors that determine the steady-state ion concentration are enumerated here. The concentrations of positive and negative ions, p and n , depend upon a dynamic balance between the positive-ion production rate per unit volume, s , and the various recombination and attachment processes among the ions, debris particles, and cloud droplets through rate equations of the form

$$p = s - (\alpha n + B'N + \gamma'C)p \quad , \quad (C1)$$

$$n = \alpha_e n_0 n_e - (\alpha p + B'N + \gamma'C)n \quad , \quad (C2)$$

where α is the ion-ion recombination coefficient, $B'N$ and $B'N$ represent the overall ion depletion rates because of attachment to debris particles, and $\gamma'C$ and $\gamma'C$ represent the overall ion depletion rates because of attachment to cloud droplets. The product, $\alpha_e n_0 n_e$, represents the negative ion production rate per unit volume because of electron attachment to neutral molecules. All other electron interactions have been suppressed here because of the anticipated small concentration of free electrons (see Appendix B).

Estimates described in Appendix B indicate that the tropospheric free-electron concentration is well approximated by $n_e = s_e(\alpha_e n_0)^{-1}$, [see Eq. (B5)]. The electron production rate per unit volume, s_e , is essentially the same as the positive ion production rate per unit volume, s . Let A be the activity of debris-particle material per unit volume and suppose that each disintegration introduces on the average 0.5 MeV into the medium via

either beta or gamma decay. At a cost of 33.7 eV per ion pair,^{C1} this energy is sufficient to produce 1.5×10^4 ion pairs per disintegration. Hence a concentration of N monodisperse debris particles of representative radius, r, will contribute a total ion production rate per unit volume of

$$s = (6 \times 10^4) A N r^3 = s_e = \alpha_e n_o n_e . \quad (C3)$$

For example, if $A \sim 10^3 \text{ Bq} \cdot \mu\text{m}^{-3}$, $r \sim 0.1 \mu\text{m}$, and $N \sim 10^4 \text{ cm}^{-3}$, then $s \sim 10^9 \text{ cm}^{-3} \cdot \text{s}^{-1}$. This value greatly exceeds $10 \text{ cm}^{-3} \cdot \text{s}^{-1}$ which represents the contribution from natural radioactivity and cosmic rays in the troposphere.^{C2}

The product, α_n , represents the volume recombination rate of positive and negative ions. The commonly accepted value^{C3} for the recombination coefficient of small atmosphere ions is $\alpha = 1.6 \times 10^{-6} \text{ cm}^3 \cdot \text{s}^{-1}$. Measured values range from $1.4 \times 10^{-6} \text{ cm}^3 \cdot \text{s}^{-1}$ obtained by Nolan^{C4} to $2.4 \times 10^{-6} \text{ cm}^3 \cdot \text{s}^{-1}$ obtained by Sayers.^{C5} The absolute temperature dependence is

$$\alpha = (1.6 \times 10^{-6} \text{ cm}^3 \cdot \text{s}^{-1}) (273/T)^{1.5} . \quad (C4)$$

The products, βN and $\beta' N$, represent, respectively, the overall depletion rates for positive and negative ions because of attachment to debris particles with a total concentration, N, but comprised of many charge states. The summations over these charge states will be suppressed here and replaced by the estimate

$$\beta N \approx \beta' N \approx 4\pi r D_i N, \quad (C5)$$

where r is a representative particle radius and D_i is the diffusion coefficient for small ions in air. (A more detailed discussion of the suppressed summations is given in Appendix E.) For Brownian diffusion of small ions in air, $D \approx 3 \text{ mm}^2 \cdot \text{s}^{-1}$ (see Appendix C). So if $r \approx 0.1 \text{ } \mu\text{m}$ and $N \approx 10^4 \text{ cm}^{-3}$, then $\beta N \approx 0.04 \text{ s}^{-1}$.

The products, γC and $\gamma' C$, represent, respectively, the overall depletion rates for positive and negative ions because of attachment to cloud droplets with a total concentration, C , but comprised of many charge states. The summations over these charge states will be suppressed here and replaced by the estimate

$$\gamma C \approx \gamma' C \approx 4\pi R D_i C, \quad (C6)$$

where R is a representative droplet radius. Take $D_i \approx 3 \text{ mm}^2 \cdot \text{s}^{-1}$, $R \approx 10 \text{ } \mu\text{m}$, and $C \approx 10^2 \text{ cm}^{-3}$ to obtain $\gamma C \approx 0.04 \text{ s}^{-1}$. Thus, for these choices for the parameters, the debris particles and cloud droplets contribute about equally to the ion depletion rate (in the absence of additional electrical forces).

Both of these estimates [Eqs. (C5) and (C6)] ignore the possibility that the collectors also might carry a charge, Q , and be situated in an ambient electric field, E . An estimate for the magnitude of such an electrical enhancement can be obtained in the case of cloud droplets, for example, by

evaluating $\gamma_E C$ in two limiting cases: (a) low field, high charge for which

$$\gamma_E C \sim 4\pi R^2 \mu_i [Q(4\pi\epsilon_0 R^2)^{-1}] C \sim (3 \times 10^{-4} \text{ s}^{-1})(Qe^{-1}) \quad (C7)$$

and (b) high field, low charge for which

$$\gamma_E C \sim 3\pi R^2 \mu_i E C \sim (1.5 \times 10^{-6} \text{ m} \cdot \text{V}^{-1} \cdot \text{s}^{-1}) E , \quad (C8)$$

where the following values have been assumed: ion mobility, $\mu_i \sim 150 \text{ mm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ (see Appendix D); $\epsilon_0 = 8.85 \text{ pF} \cdot \text{m}^{-1}$; $R = 10 \text{ } \mu\text{m}$; $C = 10^2 \text{ cm}^{-3}$; and the electronic charge, $e = 1.6 \times 10^{-19} \text{ C}$. Under thunderstorm conditions, the number of charges on a $10\text{-}\mu\text{m}$ radius droplet, Qe^{-1} , might exceed 10^3 and electric fields might exceed $0.2 \text{ MV} \cdot \text{m}^{-1}$. Under these conditions, the above contributions are comparable with each other and both are an order of magnitude greater than the estimate based upon diffusion alone. However, in the presence of high ionization, high charges and fields cannot be sustained. More likely values are $Qe^{-1} \sim 10$ and $E < 200 \text{ V} \cdot \text{m}^{-1}$ and then the electrical contributions are much smaller and may be neglected in comparison with diffusion.

The steady-state ion concentration which follows from Eqs. (C1) and (C2) when $n \approx p$ is

$$n \approx -\Sigma\alpha^{-1} + [(\Sigma/\alpha)^2 + \alpha\alpha^{-1}]^{0.5} , \quad (C9)$$

where $2\Sigma = \beta N + \gamma C$ represents the total ion depletion rate due to attachment of ions to particles and droplets by whatever means. The above

estimates show that $(s/\alpha)^{0.5}$ generally exceeds 10^7 cm^{-3} while $\Sigma\alpha^{-1}$, even if electric fields could be present, is 20 times smaller. Thus, for most cases of present interest, the ion production rate is so large that ionic recombination dominates over attachment and $n \sim (s/\alpha)^{1/2}$. In the opposite limit, which is applicable to the natural environment, $s \sim 10 \text{ cm}^{-3} \cdot \text{s}^{-1}$ and $(s/\alpha)^{1/2} \sim 10^3 \text{ cm}^{-3}$, which may be small compared with $\Sigma\alpha^{-1}$. Here, ion attachment dominates over recombination and in this limit, $n \sim 2s\Sigma^{-1}$. Equation (C9) was used to construct Fig. 1.

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APPENDIX D

IONIC CONDUCTIVITY, MOBILITIES, AND DIFFUSION

The electrical conductivity of a medium is directly proportional to the ion concentrations. If σ_p and σ_n are the polar conductivities of positive and negative ions, respectively, then the electrical conductivity, $\sigma = \sigma_p + \sigma_n$. The polar conductivities are proportional to the polar mobilities, μ_p and μ_n , and to the polar concentrations, p and n , so that

$$\sigma_p = e\mu_p p \quad \text{and} \quad \sigma_n = e\mu_n n, \quad (D1)$$

where e is the electron charge.

Mohnen^{D1,D2} has reviewed the formation, nature, and mobility of ions of atmospheric importance. The mobility of natural atmospheric small ions forms a spectrum with all values represented lying roughly between 20 and 250 $\text{mm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$. The most frequent value is then reported as the average mobility. Unfortunately, more than one peak can exist in the frequency distribution depending upon the gaseous composition of the atmosphere. The average mobility value for both negative and positive small ions is smaller in the (humid) atmosphere than it is in (pure) laboratory air with artificially increased ionization introduced just prior to the mobility analysis. This decreased mobility may be due in part to the formation of hydrated clusters. Mass spectrometric analysis suggests that clusters with 4 to 8 water molecules are common. The average mobility of negative ions exceeds that of positive ions. Various explanations have been proposed for this effect.^{D3} Some representative average mobility values are listed in Table D-1.

Table D-1. Representative average positive and negative ionic mobilities of small atmospheric ions (corrected to STP).02

Environment	μ_p ($\text{mm}^2 \cdot \text{v}^{-1} \cdot \text{s}^{-1}$)	μ_n	μ_n/μ_p
Humid air	114	124	1.09
Pure air	133	184	1.39

The Brownian diffusion coefficient is related to the mobility by the Einstein relation

$$eD_i = \mu kT, \quad (D2)$$

where e is the electron charge, k is the Boltzmann constant, and T is the absolute temperature. By using the average values for humid air at STP from Table D-1, one obtains $D_p \approx 2.68$ and $D_n \approx 2.92$ in $\text{mm}^2 \cdot \text{s}^{-1}$ for positive and negative ions, respectively.

Both μ and D_i vary with altitude. Since μ is proportional to the mean free path and inversely proportional to the mean thermal speed, it follows that

$$\mu = \mu_0(P_0/P)(T/T_0) \quad \text{and} \quad D_i = D_0(P_0/P)(T/T_0)^2, \quad (D3)$$

where the reference values of the pressure and temperature are P_0 and T_0 .

The values listed in Table D-2 have been computed from the standard atmospheric data given in Appendix A. The results are somewhat smaller than the values used by Griffiths, et al.,^{D4} but are in general agreement with those computed by Shreve.^{D5}

Table D-2. Average ionic mobilities and diffusion coefficients as a function of altitude in humid air.

Altitude (km)	Mobilities		Diffusion coefficients	
	μ_n	μ_p	D_n	D_p
	$(\text{mm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1})$		$(\text{mm}^2 \cdot \text{s}^{-1})$	
0	131	120	3.26	2.98
2	159	146	3.77	3.47
4	196	180	4.43	4.07
6	243	223	5.22	4.79
8	305	280	6.21	5.70
10	388	356	7.47	6.86

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APPENDIX E

PARTICLE CHARGE DISTRIBUTION

Radioactive nuclear debris aerosols situated in a bipolar atmosphere can become charged by the attachment of positive or negative ions and also by beta decay. The concentration of monodisperse particles with j electronic charges, N_j , may be decreased via transitions to charge state $j+1$ by beta decay or by attachment of another positive ion or via transition to state $j-1$ by attachment of a negative ion. The concentration, N_j , may be increased via attachment of a negative ion to state $j+1$ or by beta decay or positive ion attachment from state $j-1$. In the steady state, the principle of detailed balance requires that per unit volume the total transition rate from charge state j to state $j+1$ must equal the rate from state $j+1$ to j . Thus obtains the relationship

$$(\beta_{p,j}p + \delta) N_j = \beta_{n,j+1}nN_{j+1} \quad , \quad (E1)$$

where δ is the mean decay rate per particle, p and n are the positive and negative ion concentrations, and the $\beta_{i,j}$'s represent attachment coefficients between positive ($i=p$) or negative ($i=n$) ions and particles in charge state j . This relationship, together with the constraint that the sum of N_j over all possible charge states, j , (including $j=0$) must be equal to the ion concentration, N , can be used to determine the steady-state particle charge-distribution. Brizard^{E1} carried out this determination in the absence of radioactivity ($\delta=0$). Mead^{E2} extended the procedure to include the

possibility of beta decay ($\delta \neq 0$) and ambient electric fields ($E \neq 0$). Various limiting cases will be summarized here.

Subsequent expressions for the various asymptotic forms of the charge distribution depend upon several nondimensional parameters which represent ratios of various characteristic physical quantities, such as energies, fields, or process rates. They involve the electronic charge e , the permittivity of free space ϵ_0 (in S.I. Units), the Boltzmann constant k , the absolute temperature T , as well as the particle radius r . The ratio

$$b = e^2(4\pi\epsilon_0rkT)^{-1} , \quad (E2)$$

compares the characteristic electrostatic energy, $e^2(4\pi\epsilon_0r)^{-1}$, (in S.I. Units) with the characteristic mean thermal energy, kT . The ratio

$$a = eEr(kT)^{-1} , \quad (E3)$$

compares. The energy gained by charge e in traversing the potential difference, Er , with the mean thermal energy, kT . The ratio

$$c = e(4\pi\epsilon_0r^2E)^{-1} = ba^{-1} , \quad (E4)$$

compares the characteristic electric field, $e(4\pi\epsilon_0r^2)^{-1}$, (in S.I. Units) with the magnitude of the applied electric field, E , or equivalently, compares the two electrical energies involved in the definitions of b and a independently of temperature. Finally, the ratio

$$\xi_i = \delta \epsilon_0 \sigma_i^{-1} , \quad (i = p \text{ or } n) \quad (E5)$$

compares the decay rate per particle, δ , with the characteristic electrical relaxation rate, $\sigma_i \epsilon_0^{-1}$, for a medium with electrical conductivity, σ_i , ($i = p$ for positive or n for negative ions) (in S.I. Units). The product

$$c\xi_i = e\delta(4\pi r^2 \sigma_i E)^{-1} , \quad (E6)$$

compares the outward flowing decay current $e\delta$ with an inward flowing conduction current.

Expressions are required for the various ion-particle attachment coefficients, $B_{i,j}$. In the absence of applied fields, Bricard^{E3} shows that

$$B_{i,j} = 4\pi r D_i F_i(j) , \quad (E7)$$

where

$$F_i(j) = bjs_i [\exp(bjs_i) - 1]^{-1} . \quad (E8)$$

For weak applied electric fields ($a \ll 1$), Klett^{E4} shows that

$$B_{i,j} = 4\pi r D_i F_i(j) G_i(j) , \quad (E9)$$

where

$$G_i(j) = 1 + 0.5a F_i(j) \exp(bjs_i) \quad . \quad (E10)$$

Klett further argues that this expression is valid for $a < 4$. For strong externally applied fields ($a \gg 1$), Gunn^{E5} shows that

$$B_{i,j} = 3\pi r^2 \mu_i E (1 - cjs_i/3)^2 \quad , \quad (E11)$$

where μ_i is the appropriate ionic mobility. (The more general case applicable to falling droplets in a strong electric field was examined by Whipple and Chalmers.^{E6}) In all of the definitions (E7)-(E11), the subscript i takes on the label p for positive and n for negative ions and quantities, $s_p = +1$ and $s_n = -1$. These expressions were developed by using the macroscopic laws for diffusion and conduction of ions. They are appropriate as long as the debris particles have dimensions greater than a mean free path for ion-molecular collisions, which is of the order of 0.01-0.04 μm , in the present case. Image forces, which become important below approximately 0.05 μm , have been neglected.^{E7}

Mead^{E2} used the recursion relationship (E1) together with the ion-particle attachment coefficients (E7)-(E11) to obtain asymptotic formulae for the nuclear debris particle charge distribution in several regimes of interest here. Application of The Central Limit Theorem allows all the asymptotic formulae to be represented approximately in terms of the shifted Gaussian distribution

$$N_j N^{-1} = (2\pi w^2)^{-0.5} \exp[-(j-\bar{j})^2 / (2w^2)] \quad (E12)$$

where $\bar{j}$ is the mean number of charges and $w^2 = \overline{(j-\bar{j})^2}$ is the dispersion. The cases can be summarized as follows:

A. Low Activity ($\delta(B_{1,jp}) < 1$)

a) Weak external electric field ($a \ll 1$)

$$\bar{j} = (1 + 0.5a)[\xi_p + b^{-1} \ln(\sigma_p \sigma_n^{-1})] , \quad (E13)$$

$$w^2 = (1 + 0.5a)b^{-1} , \quad (E14)$$

b) Strong external electric field ($a \gg 1$)

$$\bar{j} = \xi_p + 3(4c)^{-1} \ln(\sigma_p \sigma_n^{-1}) , \quad (E15)$$

$$w^2 = 3(4c)^{-1} , \quad (E16)$$

B. High Activity ($\sigma(B_{1,jp}) \gg 1$)

a) Weak external electric field ($a \ll 1$)

$$\bar{j} = \xi_n , \quad (E17)$$

$$w^2 = \xi_n , \quad (E18)$$

b) Strong external electric field ($a \gg 1$)

$$\bar{J} \sim 3(2c)^{-1} \ln(4c\xi_n/3) \quad , \quad (E19)$$

$$w^2 = 3(2c)^{-1} \quad . \quad (E20)$$

These formulae reduce to all the previously known special cases when $\delta \rightarrow 0$ and/or $a \rightarrow 0$. In particular, when both the radioactivity and external field are absent ($\delta = a = 0$), Eqs. (13) and (14) reduce to the well-known "Boltzmann" charge distribution proposed by Keefe, Nolan, and Rich.^{E8} This particular form seems to give the best fit to the observed^{E9, E10} particle-charge distribution for particle radii less than 0.1 μm . Allowance has been made in these formulae for slightly differing ionic mobilities (see Appendix D) which permit the mean charge to be shifted away from zero in the direction of the sign of the more mobile ion.^{F11} The form of Eqs. (E17) and (E18) in the case of high activity and low field reveals that the charge actually is Poisson-distributed, as can be verified by more elementary methods.

The strength of the externally applied electric field, E , is measured by the parameter, a . When $a \gg 1$, $E \gg (2.4 \times 10^4 \text{ V}\cdot\text{m}^{-1}\cdot\mu\text{m})r^{-1}$.

It will be argued that such fields are difficult to sustain in the highly ionized nuclear-debris environment. Consequently, the relationships for weak ($a \ll 1$) external electric fields will be of principal interest here. In this case useful formulae, which interpolate between the low and high activity limits, are

$$\bar{J} = (1 + 0.5a)[\xi + b^{-1} \ln(\sigma_p \sigma_n^{-1})] \quad (E21)$$

and

$$w = [(1 + 0.5a)b^{-1} + \xi]^{0.5} , \quad (E22)$$

where $\xi = 6\epsilon_0(\mu_p\mu_n)^{-1/2}$ contains the geometric mean ionic conductivity.

The shifted Gaussian distribution with this mean value and dispersion can be used to estimate the mean number of positive charges per particle

$$\bar{J}_+ = N^{-1} \int_0^{\infty} j N_j dj = 0.5\bar{J}[1 + \text{erf}(u)] + w(2\pi)^{-0.5} \exp(-u^2) \quad (E23)$$

and the mean number of negative charges per particle

$$\bar{J}_- = N^{-1} \int_0^{\infty} j N_j dj = 0.5\bar{J}[-1 + \text{erf}(u)] + w(2\pi)^{-0.5} \exp(-u^2) , \quad (E24)$$

where $u = \bar{J}(2w^2)^{-0.5}$. These relationships have been used to construct Fig. 2. Values of $\bar{J}_{\pm} < 1$ simply reflect the fact that most of the particles either are neutral or have the respective opposite charge. These two situations can be distinguished by examining the value of the mean charge, $\bar{J} = \bar{J}_+ - \bar{J}_-$, which is represented graphically by the separation between the respective curves in Fig. 2.

Practical estimates for ξ can be obtained by fixing the amount of material present. It is convenient to fix the product, $r^3N = (.1 \mu\text{m})^3(10^4 \text{ cm}^{-3}) = 10^{-11}$. Then

$$\xi = \frac{\epsilon_0}{e} \left[\frac{4\pi\alpha E_2}{3\mu_p\mu_n E_Y} \right]^{1/2} \left(\frac{Ar^3}{N} \right)^{1/2} \sim 2.5 \left(\frac{A}{1 \text{ Bq} \cdot \mu\text{m}^{-3}} \right)^{1/2} \left(\frac{r}{1 \mu\text{m}} \right)^3 , \quad (E25)$$

where $\epsilon_0 = 8.85 \text{ pF} \cdot \text{m}^{-1}$, $e = .16 \text{ aC}$, $\alpha = 1.6 \text{ mm}^3 \cdot \text{ks}^{-1}$, $E_2 = 33.7 \text{ eV}$, $E_Y = 0.5 \text{ MeV}$, and $(\mu_p\mu_n)^{0.5} \sim .15 \text{ m}^2 \cdot \text{V}^{-1} \cdot \text{ks}^{-1}$. Consider the ranges: $10 < A < 10^5 \text{ Bq} \cdot \mu\text{m}^{-3}$ and $0.01 < r < 1 \mu\text{m}$.

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APPENDIX F

GAMMA-INDUCED IONIZATION

Gamma rays penetrate much farther through air than do beta rays. Consequently, they generate ionization over a much larger region of space. It is this ionization that comprises the conducting boss in the Holland model. Formulae (and their physical units), used to generate the isochrones of constant electrical relaxation time, which are displayed in Fig. 7, will be recorded here.

i) Source Details:^{F1} [Units]

t = time elapsed after burst [s]

$N(t)$ = gamma emission rate at time t [s^{-1}]

$$= \dot{N}(0) \begin{cases} 1 & t \leq 1 \text{ s} \\ t^{-1.2} & t \geq 1 \text{ s} \end{cases}$$

Y = nuclear yield [kt]

$N(0)$ = initial gamma emission rate. [s^{-1}]

$$= \frac{1}{6 \text{ s}} \left(\frac{7.2 \text{ 1 MeV } \gamma/\text{fission}}{200 \text{ MeV/fission}} \right) (1.61 \times 10^{25} \frac{\text{MeV}}{\text{kt}}) Y$$

$$= (1.57 \times 10^{23} \text{ s}^{-1} \cdot \text{kt}^{-1}) Y$$

ii) Atmospheric ^{F2} and Material Properties:^{F3}

z = altitude above sea level [m]

$T(z)$ = atmospheric temperature altitude z [$^{\circ}\text{K}$]

$\rho(z)$ = atmospheric mass density at altitude z [$\text{kg} \cdot \text{m}^{-3}$]

$P(z)$ = atmospheric pressure at altitude z [Pa]

$\mu_a(z)$ = linear energy absorption coefficient for [m^{-1}]

gamma's of some specified energy at altitude z

$\mu_s(z)$ = linear scattering coefficient for gamma's [m⁻¹]
of some specified energy at altitude z

$\mu_t(z)$ = total linear attenuation coefficient for [m⁻¹]
gamma's of some specified energy at altitude z

$\mu_t(z) = \mu_a(z) + \mu_s(z)$ [m⁻¹]

$\mu_j(z) = \mu_j(0) \frac{\rho(z)}{\rho(0)}$ j = a, s, t [m⁻¹]

Data: F³ (T = 288° K, 1 atm, 1 MeV)

$\mu_a(0) = 3.43 \text{ km}^{-1}$; $\mu_s(0) = 4.29 \text{ km}^{-1}$; $\mu_t(0) = 7.72 \text{ km}^{-1}$

iii) Gamma-Ray Flux

H = source altitude above sea level [m]

r = (cylindrical) radial distance to detector [m]
= (x²+y²)^{0.5}

R(r,z) = distance from source to detector [m]
= [r²+(z-H)²]^{0.5}

g = gravitation acceleration [m•s⁻²]

K_j(r,z,H) = attenuation integrals for j = a, s, t [1]

$$= \int_H^z \mu_j(\zeta) \frac{R(r,\zeta) d\zeta}{\zeta-H} = \frac{\mu_j(0)}{\rho(0)g} \frac{[P(H)-P(z)]}{(z-H)} R(r,z)$$

J_p(r,z,H) = primary gamma-ray flux at (r,z) [m⁻²•s⁻¹]
from point source at (0,H)

$$= \frac{\dot{N}(t)}{4\pi[R(r,z)]^2} \exp[-K_t(r,z,H)]$$

$$B_s(r,z,H) = \text{build-up factor due to scattering} \quad [1]$$

between (0,H) and (r,z)^{F4,F5}

$$= \frac{\mu_s(0)}{\mu_t(0)} \exp[K_s(z,H)]$$

$$J(r,z,H) = \text{total gamma-ray flux at (r,z)} \quad [m^{-2} \cdot s^{-1}]$$

from point source at (0,H)

$$= B_s(r,z,H) \cdot J_p(r,z,H)$$

$$E_Y = \text{mean gamma-ray energy} \quad [eV]$$

$= 10^6 \text{ eV}$

$$E_2 = \text{mean energy needed to produce} \quad [eV]$$

one ion-pair

$= 33.7 \text{ eV}$

$$I(r,z,H) = \text{ion-pair production rate per unit} \quad [m^{-3} \cdot s^{-1}]$$

volume at (r,z)

$$= \frac{E_Y}{E_2} \mu_a(z) J(r,z,H)$$

iv) Electrical Relaxation Time

$$e = \text{electron charge} \quad [C]$$

$= 1.6 \times 10^{-19} \text{ C}$

$$\epsilon_0 = \text{permittivity of free space} \quad [F \cdot m^{-1}]$$

$= 8.85 \times 10^{-12} \text{ F} \cdot m^{-1}$

$$\alpha(z) = \text{ion recombination coefficient at altitude } z \quad [m^3 \cdot s^{-1}]$$

$= \alpha(0) [T(0)/T(z)]^{1.5}$

Data: (T = 288°K, 1 atm)

$$\alpha(0) = 1.49 \times 10^{-12} \text{ m}^3 \cdot s^{-1}$$

$$\begin{aligned}
 n(r,z,H) &= \text{equilibrium ion concentration at altitude } z & [\text{cm}^{-3}] \\
 &= \{I(r,z,H)/\lambda(z)\}^{0.4} \\
 u(z) &= \text{ionic mobility at altitude } z & [\text{cm}^2/\text{V}\cdot\text{sec}] \\
 &= u(0)u(z)/u(z) \\
 \lambda(r,z,H) &= \text{ionic conductivity at altitude } z & [\text{sec}\cdot\text{cm}^{-1}] \\
 &= en(r,z,H) \cdot u(z) \\
 \tau(r,z,H) &= \text{electrical relaxation time at altitude } z & [\text{sec}] \\
 &= \epsilon_0/\lambda(r,z,H)
 \end{aligned}$$

Procedure:

Compute ion production rate per unit volume, $I(r,z,H)$, using the values of $\rho(z)$, $P(z)$, and τ from the standard atmosphere^{F1} and the formulae in i)-iii). Then evaluate $\lambda(r,z,H)$ and plot contours of constant $\tau(r,z,H)$.

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APPENDIX G SOME ELECTROSTATICS

Estimates for the electric field in the neighborhood of a conducting, hemispheroidal boss on a conducting plane with a uniform electric field, F , at infinity can be obtained by using the following formulae:^{G1,G2}

$$E_z = F \cdot \left\{ 1 - \frac{A(\lambda)}{A(0)} + \frac{2z^2(a^2+\lambda)(c^2+\lambda)^{1.5}}{A(0)[r^2(c^2+\lambda)^2 + z^2(a^2+\lambda)^2]} \right\}$$

where

$$\lambda = \frac{1}{2}(r^2 + z^2 - a^2 - c^2) + \frac{1}{2}[(r^2 + z^2 - a^2 - c^2)^2 + 4(a^2 z^2 + r^2 c^2 - a^2 c^2)]^{0.5}$$

The quantity λ is the spheroidal coordinate of the point, (r, z) (in cylindrical coordinates), outside the spheroid with symmetry axis c along z (perpendicular to the conducting plane) and whose surface coincides with $\lambda=0$. For oblate spheroids: $(c < a)$ $(1 - c^2/a^2)^{0.5} \equiv \sin \zeta$,

$$A(\lambda) = \frac{2 \cos \zeta}{a^2 c \sin^3 \zeta} [\beta \tan \zeta - \tan^{-1}(\beta \tan \zeta)]$$

For prolate spheroids: $(c > a)$ $(1 - a^2/c^2)^{0.5} \equiv \sin \eta$

$$A(\lambda) = \frac{2 \cos^2 \eta}{a^2 c \sin^3 \eta} \left[\ln \left(\frac{1 + \beta \sin \eta}{1 - \beta \sin \eta} \right)^{0.5} - \beta \sin \eta \right]$$

where $\beta \equiv (1 + \lambda/c^2)^{-0.5}$ in both oblate and prolate cases.

Table G-1 contains a comparison between Holzer's observations^{G3} and the electric fields which have been estimated by using Holland's boss model^{G11} and the formulae for a hemispheroidal boss in a uniform field.

Table G-1. Comparison of boss model with observations at two of Holzer's stations.

Time [s]	Observed fields ⁶³ [V·m ⁻¹]		Model fields [V·m ⁻¹]	
	station 1	station 2	station 1	station 2
1	5	20	4.2	20
600	57	26	68	40

The agreement between the observations and the model is extremely good at very early time because the isochrone for the 10 s electrical relaxation time (in Fig. 7a) is nearly a prolate hemispheroid. The agreement near the maximum (at about 5 min) is within a factor of two, even though: (i) a representative oblate spheroid is more difficult to prescribe from the curves in Fig. 7e, (ii) the infinite series of "image bosses," which would be mirrored through the ground plane, have been omitted, and (iii) the nearly exponential decrease in conductivity with altitude has been neglected.

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APPENDIX H

SUPPRESSION OF ELECTRIC FIELD GROWTH

The temporal evolution of a thunderstorm electric field, E , is governed by the Maxwell equation

$$\frac{dE}{dt} + \epsilon_0^{-1} J = 0, \quad (H1)$$

where $\epsilon_0 = 8.85 \text{ pF} \cdot \text{m}^{-1}$ is the vacuum permittivity and J is the total current density. This current has contributions from the motions of charged droplets, charged hydrometers, and in some cases ions.

As a first attempt to assess the effect of high ion production rates on electric field growth, Spangler and Rosenkilde^{H1} presented the following rough estimate of the ion concentration that might be expected to prevent field growth. The estimate is based upon a comparison between the generator current inferred to be in actual thunderstorms and the ion conduction current anticipated to be present in a debris cloud.

The electrification processes, whatever they may be, must be capable of starting from a fair-weather field the order of $10^2 \text{ V} \cdot \text{m}^{-1}$ and generating fields in excess of $400 \text{ kV} \cdot \text{m}^{-1}$ according to measurements by Winn et al.^{H3} The initial electrification of a developing cloud often increases exponentially with an e-folding time of two minutes or so, according to observations of Reynolds and Brook.^{H2} So, suppose that during its growth, in the absence of significant ionization, the electric field strength is roughly an exponential function of time, $E = E_0 \exp(\xi t)$. The growth rate, ξ , can be estimated from the relationship

$$\xi = \Delta(\ln E) / \Delta t. \quad (H2)$$

For growth from $10^2 \text{ V}\cdot\text{m}^{-1}$ to $400 \text{ kV}\cdot\text{m}^{-1}$ over 10^2 s , this relationship gives $\xi < 0.08 \text{ s}^{-1}$. Even under the most favorable conditions, it appears to be difficult to achieve growth rates much greater than 0.1 s^{-1} . Then Eq. (H1) implies a generator current density,

$$-J_g = \epsilon_0 dE/dt = \epsilon_0 \xi E \approx (7.1 \times 10^{-4} \text{ nA}\cdot\text{m}^{-1}\cdot\text{V}^{-1})E \quad (\text{H3})$$

must be operating in a thunderstorm. For $E \sim 10^2 \text{ kV}\cdot\text{m}^{-1}$ this gives $J_g \sim 70 \text{ nA}\cdot\text{m}^{-2}$, which is consistent with the value ($>60 \text{ nA}\cdot\text{m}^{-2}$) found by Rust and Moore^{H4} who allowed instrumented free balloons to rise through active thunderstorms.

Now estimate the conduction current that would be present in a debris cloud. If the positive and negative ion concentrations, p and n , are about equal and μ_p and μ_n are the respective positive and negative ion mobilities, then the conduction current density due to ions is

$$J_c = ne(\mu_p + \mu_n)E, \quad (\text{H4})$$

where $e = 1.6 \times 10^{-19} \text{ C}$ is the magnitude of the electron charge. According to the data in Appendix D, $\mu_p + \mu_n \sim 4 \times 10^{-4} \text{ m}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$. It follows that $J_c \approx (6.4 \times 10^{-23} \text{ Am}^2\cdot\text{V}^{-1})nE$.

Electric field growth will be prohibitive if the conduction current density, J_c , exceeds the current density, J_g , of the generator, whatever its mechanism. It follows that the steady-state ion concentration

$$n > \frac{7.1 \times 10^{-13} \text{ A}\cdot\text{m}^{-1}\cdot\text{V}^{-1}}{6.4 \times 10^{-23} \text{ Am}^2\cdot\text{V}^{-1}} \sim 1.1 \times 10^4 \text{ cm}^{-3} \quad (\text{H5})$$

This is comparable to the maximum space-charge concentrations

$(8 \times 10^{-9} \text{ C} \cdot \text{m}^{-3} \sim 5 \times 10^{-4} \text{ ions} \cdot \text{cm}^{-3})$ that have been observed^{H4}

aloft. However, an examination of Fig. 1 will show that this value is three orders of magnitude less than that to be expected in a nuclear debris cloud.

Electric field growth in the presence of nuclear debris appears to be possible only if the material can be diluted sufficiently to reduce the steady-state ion concentration below a limit of this order of magnitude.

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APPENDIX I

THE CONTINUUM-FLUX MODEL

The flux model is used widely to describe scavenging of aerosols of the order of $0.1 \mu\text{m}$ or smaller. It is designed to describe the behavior at an ensemble of particles as it drifts, with both random and deterministic velocity components, through the mediating air accordance with the laws of continuum mechanics.

Physical interactions are introduced into the model by specifying the form of the particle flux, $\vec{J}$, to a single collector. The continuity of the steady-state flux requires that

$$\nabla \cdot \vec{J} = 0 \quad (11)$$

This condition usually reduces to some form of the convective-diffusion equation which must be solved in order to determine the particle concentration, N , as a function of position. Once this function has been determined, it is possible to evaluate the desired particle current by integrating the flux over the collector surface.

Unfortunately, the convective-diffusion equation is difficult to solve in general because of the presence of various boundary layers. Only a few particular solutions are known for simple geometries. Such special solutions commonly are applied to obtain estimations in situations where more appropriate exact solutions are unavailable.

It is useful to record here some of these particular solutions which are applicable to scavenging by a spherical collector of radius, R . Solutions,

which include diffusion, have been obtained by using the two boundary conditions:

$$\lim_{r \rightarrow R} N = 0 \quad \text{and} \quad \lim_{r \rightarrow \infty} N = N_{\infty} \quad (I2a,b)$$

Solutions, which include only conduction or convection (and ignore diffusion), have been obtained by using only the second boundary condition [Eq. (I2b)]; since diffusion is being ignored, there is no compulsion to require $N(R)$ to vanish. For each of the solutions to be listed, the form assumed for J_x (with subscript label, x , to be specified) and the collection current (to a single collector), I_x , or an associated quantity, will be given.

1. Brownian diffusion in spherical symmetry to a stationary collector:

$$J_b = -D_b(dN(r)/dr)\hat{r}, \quad (I3a)$$

$$I_b = 4\pi R^2(D_b N_{\infty}/R) = 4\pi R D_b N_{\infty}, \quad (I3b)$$

where D_b is the constant Brownian diffusion coefficient for aerosol particles in air (see Appendix J).

It often is convenient to compare expressions for the current per collected particle, I_x/N_{∞} , which is known as a collection kernel, K_x . It also is common to express the kernels, which involve diffusion, relative to the kernel for Brownian diffusion,

$$K_b = 4\pi R D_b. \quad (I4)$$

The ratio, K_x/K_b , may be regarded as a Sherwood number, $(Sh)_x$, which is associated with the particular mass-transfer process labeled by the subscript, x. Clearly, $(Sh)_b \equiv 1$.

2. Conductive-diffusion in spherical symmetry to a stationary collector:

$$J_F = [NBF(R/r)^2 - D_b(dN(r)/dr)]r^2, \quad (15a)$$

$$(Sh)_F = \frac{w}{\exp(w)-1} \sim \begin{cases} w \exp(-w) & +1 \ll w \\ 1 - 0.5 w & 0 < w \ll 1 \\ 1 + 0.5 w & -1 \ll w < 0 \\ -w & w \ll -1 \end{cases} \quad (15b)$$

where B is the constant particle mobility in a force field, F, (see Appendix J) and

$$w \equiv (Pe)_F = BFR/D_b \quad (15c)$$

is a signed Péclet number whose magnitude measures the importance of convection (or conduction) relative to diffusion. This quantity contains the ratio, D_b/B , which equals kT ($k=1.38 \times 10^{-23} \text{ J} \cdot \text{°K}^{-1}$ is Boltzmann's constant and T is the absolute temperature) when local thermodynamic equilibrium prevails. Because of the existence of this Einstein relationship, it is not permissible to seek a limiting form for the above function, $(Sh)_F(w)$, for which D_b approaches zero as B remains finite. This prevents one from using a simple limiting process to obtain a solution, which would be appropriate for pure conduction (or convection) with diffusion being ignored, directly from a solution, which explicitly includes diffusion.

In the present case of electrical scavenging,

$$F = qQ(4\pi\epsilon_0 R^2)^{-1} , \quad (16)$$

where the particle charge, q , and the droplet charge, Q , (both in S.I. Units) may have either sign in order to account for repulsion (positive w) or attraction (negative w). In the particular case for which $q = e$ and $Q = ej$, the earlier expressions in Eqs. (E2) and (E8) are recovered with $w = bj$.

Wang, Grover, and Pruppacher¹¹ have shown that diffusiophoresis and thermophoresis also can be treated within this framework because the vapor fields are proportional to r^{-2} .

3. Pure conduction (with diffusion ignored) in spherical symmetry to a stationary collector:

$$\vec{J}_c = NBF(R/r)^2 \hat{r} \quad (17a)$$

$$I_c = 4\pi R^2 BFN_\infty . \quad (17b)$$

In the particular case that $F = qE_Q$, where $E_Q = Q(4\pi\epsilon_0 R^2)^{-1}$, this current is independent of the collector radius.

Small, charged, aerosol particles with little inertia, which are being collected here via purely conductive (or convective) means, drift along trajectories that closely follow the electric field lines. The trajectories (or field lines), which intersect the collector surface, are used to determine an effective "collision" cross-section for scavenging. In such cases, it is convenient to measure the effectiveness of a scavenging process relative to

the collection that would have been obtained if the purely geometrical collision cross-section, πR^2 , had been applicable and the objects were assumed to come together with the relative gravitation settling velocity, U_g . In such a case, the collection kernel would have been

$$K_g = \pi R^2 U_g \quad (18)$$

The efficiency, $(Ef)_x$, of a given scavenging process then can be expressed as the ratio, K_x/K_g . For the above example

$$(Ef)_c = I_c (N_\omega K_g)^{-1} = 4 U_Q U_g^{-1} \quad (19)$$

where $U_Q = B|qE_Q|$ is the drift velocity of a particle with charge q in the spherically symmetric field, E_Q , at the collector surface. In cases, where the particle radius is not negligible compared to the collector radius, the geometrical cross-section should involve the sum of these radii.

4. Pure conduction (with diffusion ignored) in axial symmetry to a stationary collector:

$$\vec{J}_E = NqB\vec{E} \quad (110a)$$

$$\begin{aligned} \vec{E} = & [E_\infty(1 + 2R^3/r^3)\cos\theta + Q(4\pi\epsilon_0 r^2)^{-1}]\hat{r} \\ & - E_\infty(1 - R^3/r^3)\sin\theta \hat{\theta} \end{aligned} \quad (110b)$$

$$(Ef)_E = \frac{U_E}{U_g} \cdot \begin{cases} 3[1 - E_Q/(3E_\infty)]^2 & U_E > U_g ; -3E_\infty < E_Q \leq 0 \\ 4|E_Q|E_Q^{-1} & U_E < U_g ; -3E_\infty < E_Q \leq 0 \\ \text{all } U_E ; E_Q < -3E_\infty & \end{cases} \quad (110c)$$

where $q > 0$, $Q < 0$, and $U_E = qBE_\infty$ is the charged particle's electrical drift velocity in the uniform field, E_∞ . Whipple and Chalmers¹² originally derived efficiencies of this form for ion collection by a freely falling raindrop. In extending their formulae to the case of particle collection, several cases must be considered, which depend upon the relative magnitudes of U_E and U_g . Only the cases for which $q > 0$, $Q < 0$, with a downward-directed electric field have been listed here. Spangler¹³ has listed all the other possible combinations for positive or negative particles, for falling or rising droplets, and for downward or upward directed electric fields.

5. Conductive-diffusion in axial symmetry to a stationary collector:

$$\vec{J}_e = NqBE - D_b[\hat{r}(\partial N/\partial r) + \hat{\theta}r^{-1}(\partial N/\partial \theta)], \quad (111a)$$

where E is given by Eq. (110b). Klett¹⁴ argued that the conduction term grows (relative to the diffusion term) with increasing distance from the droplet until conduction eventually dominates over diffusion for any chosen value of the parameter, $a|q|E_\infty R(kT)^{-1}$ [see Eq. (E3)]. Under these conditions, a regular perturbation expansion in powers of a must break down at large distances. In order to obtain an approximate solution, which is uniformly valid at all distances, it is necessary to use the technique of

matched asymptotic expansions.¹⁵ For values of the expansion parameter, $a \leq 4$, Klett derived the expression

$$(Sh)_e = \frac{w}{\exp(w)-1} \left[1 + \frac{a w \exp(w)}{2(\exp(w)-1)} \right], \quad (111b)$$

where $a = \{q|BE_\infty R D_b^{-1}\}$ and $w = qBE_Q R D_b^{-1}$ [cf. Eqs. (E2), (E3), and (15c)]. Note that the parameters, a and w , represent Péclet numbers, $(Pe)_E$ and $(Pe)_Q$, which compare the charged particle's conductive drift in fields, E_∞ and E_Q , with the process of thermal diffusion in the air. The leading term in Eq. (111b) agrees with Eq. (15b) in the limit that E_∞ approaches zero and w is regarded as a signed Péclet number for which the negative sign corresponds to attraction.

Simple analytical solutions of the conductive (or convective)-diffusion equation, which take into account the hydrodynamic flow fields surrounding a settling droplet, are more difficult to obtain.¹⁶ Several elementary flow fields are available. Of these, Stokes flow seems most appropriate for the settling of small cloud droplets. It is common to account for the boundary layer's effect on diffusion by introducing a ventilation factor.¹⁷ Several elementary functional forms have been proposed for this factor.¹⁸ These forms are based upon boundary layer arguments and some asymptotic calculations. The expressions depend upon the appropriate Reynolds number, $(Re) \equiv 2RU_g v^{-1}$, Schmidt number, $(Sc) \equiv \omega D_b^{-1}$, and/or Péclet numbers $(Pe) \equiv RU_g D_b^{-1}$, where v is the kinematic viscosity of air (see Appendices A and J). (The Péclet numbers, which are being defined here, depend upon radii instead of diameters in contrast to common practice.)

In the case of low Reynolds numbers, $Re \ll 1$, but very high Péclet numbers, $Pe \gg 1$, Levich¹⁹ and others^{110,111} have shown that this

ventilation factor should be proportional to $(Pe)^{1/3}$. The interpolation formula

$$f_g = 1 + 0.625 (Pe)^{1/3} \quad (112)$$

often is used.¹⁹ In the case of large Reynolds numbers, $Re \gg 1$, Acrivos¹¹² and others have shown that this factor should be proportional to $(Re)^{1/2}(Sc)^{1/3}$. Experiments by Beard and Pruppacher¹¹³ and numerical results by Woo and Hamielec,¹¹⁴ both suggest that this second form is appropriate even for $(Re) > 0.1$. Their expression for the ventilation factor is

$$f_g = \begin{cases} 1.00 + 0.108 x^2 & x < 1.4 \\ 0.78 + 0.308 x & x > 1.4 \end{cases} \quad (113)$$

where $x \equiv (Re)^{1/2}(Sc)^{1/3}$. (The subscript g on f_g signifies that the gravitational settling velocity, U_g , has been used in the definition.)

According to Table J-1, when $R = 10 \mu m$, $U_g = 1.27 \text{ cm} \cdot \text{s}^{-1}$, and $v = 0.17 \text{ cm}^2 \cdot \text{s}^{-1}$, the collector's Reynolds number, $(Re) \sim 7.4 \times 10^{-3}$. The Péclet number for scavenging is the product of the collector's Reynolds number and the particle's Schmidt number. For particles with radii from 0.01 to $1.0 \mu m$, Schmidt numbers range from 10^3 up to 1.3×10^6 and hence this Péclet number ranges from 7.4 up to 10^4 . Consequently, $(Pe)^{1/3}$ ranges from 2 up to 21.5 , while the parameter, x , ranges from 2 up to 9.5 . Two observations can be made from these comparisons. (1) Both functional forms of Eq. (113) will be needed.

(2) Eq. (I12), if it were to be used, would overestimate the ventilation factor, as given by Eq. (I13), by as much as a factor of 5 (for a 1.0- μ m radius particle).

The electrical scavenging of charged aerosols by charged droplets in an electric field can be estimated by combining appropriate formulas from the above list of elementary solutions. For low electric fields, where $a < 4$, the overall collection kernel ought to behave like the product of factors defined in Eqs. (I4), (I11b), and (I13), so

$$K_{el} \sim K_b \cdot f_g \cdot (Sh)_e \quad (I14)$$

For high electric fields, where $a \gg 4$, the overall collection kernel ought to behave like the product of factors defined in Eqs. (I4) and (I10c), so

$$K_{el} \sim K_g \cdot (Ef)_E \quad (I15)$$

An expression like Eq. (I15) was used by Williams^{I15} in his earlier discussion of electrical effects in rainout. Care should be exercised in using the sum of these two kernels to interpolate between the two limiting cases because $(Sh)_e$ in Eq. (I14) involves a series expansion in powers of a and this limits the range of its validity. Recently, Wang and Pruppacher^{I16}

proposed an interpolation function, which contains only the leading terms in each of these expressions and has the form

$$K_{e1} \sim 3K_g U_g^{-1} + K_b \cdot f_g \cdot (Sh)_E \quad (116)$$

in the present notation. The replacement of $(Sh)_e$ by $(Sh)_E$ from Eq. (15b) eliminates the dependence on E_∞ from the second term.

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APPENDIX J TRANSPORT COEFFICIENTS AND TERMINAL VELOCITIES

It is necessary to have values available for various transport coefficients and terminal velocities for both aerosols and cloud droplets in air. These quantities are discussed and tabulated here.

A sphere of mass, m , which is subjected to the gravitational acceleration, g , while in air whose dynamic viscosity is η , eventually will achieve a terminal velocity, U_g , over a characteristic time scale, τ_i . Suppose that the objects are sufficiently small, so that the air flow around them can be represented by Stokes flow. Then the particles must obey the equation

$$m d\vec{v}/dt + 6\pi\eta r\vec{v} = m\vec{g} \quad (J1)$$

In this case, the particle mobility is $B = (6\pi\eta r)^{-1}$, the characteristic entrainment time is $\tau_i = mB$, and the terminal velocity is $U_g = g\tau_i$. Deviations from these relationships are observed for very small ($r < 0.1\mu m$) particles whose size approaches the molecular mean free path, λ_a . In such cases, where the Knutsen number, $K_n \equiv \lambda_a/r$, approaches unity, a slip-flow correction factor, α_c , is applied to the mobility. This factor generally takes the form, $1 + \alpha_c(Kn)$, where

$$\alpha_c = A_1 + A_2 \exp[A_3/(Kn)] \quad (J2)$$

and the coefficients have the values: $A_1 = 1.257$, $A_2 = 0.400$, and $C = -1.10$, according to Davies.^{J1} It follows that the corrected expressions are:

$$B = [1 + \alpha_c(K_n)] (6\pi\eta r)^{-1}, \quad (J3)$$

$$\tau_i = \frac{2(\frac{\rho}{\rho_a})r^2}{9\nu} [1 + \alpha_c(K_n)] , \quad (J4)$$

$$U_g = \frac{2(\frac{\rho}{\rho_a})r^2g}{9\nu} [1 + \alpha_c(K_n)] , \quad (J5)$$

where ρ is the particle's mass density and $\nu = \eta/\rho_a$ is the kinematic viscosity of air. These expressions give adequate approximations for solid spheres with radii $< 25 \mu\text{m}$. Some representative values are tabulated in Table J-1. The diffusion coefficient is obtained by using the Einstein relationship, $D_b = BkT$, where $k = 1.38 \times 10^{-23} \text{ J}^\circ\text{K}^{-1}$ is Boltzmann's constant and T is the absolute temperature (275°K at 2 km). Then the particle Schmidt number, $Sc = \nu/D_b$, and the particle Reynolds number, $Re = rU_g/\nu$. The last column in Table J-1 lists the rms thermal velocity, $v_{\text{rms}} = (1.5 kT/m)^{0.5}$. A particle mass density of $1 \text{ gm}\cdot\text{cm}^{-3}$ has been assumed when necessary.

Table J-1. Various aerosol parameters as a function of radius, r ,
at 275° K and a mass density of $1 \text{ gm}\cdot\text{cm}^{-3}$.

r [m]	K_n [1]	B [$\text{m}\cdot\text{s}^{-1}\cdot\text{N}^{-1}$]	D [$\text{m}^2\cdot\text{s}^{-1}$]	τ_i [s]	U_g [$\text{m}\cdot\text{s}^{-1}$]	Sc [1]	Re [1]	v_{rms} $\text{m}\cdot\text{s}^{-1}$
1E-8	8.07E0	4.28E12	1.63E-8	1.79E-8	1.76E-7	1.05E3	1.03E-10	1.66E0
2E-8	4.04E0	1.12E12	4.26E-9	3.75E-8	3.68E-6	4.02E3	4.30E-10	5.85E-1
5E-8	1.61E0	2.06E11	7.82E-10	1.08E-7	1.06E-6	2.19E4	3.09E-9	1.48E-1
1E-7	8.07E-1	6.43E10	2.44E-10	2.70E-7	2.64E-6	7.01E4	1.54E-8	5.24E-2
2E-7	4.07E-1	2.33E10	8.85E-11	7.80E-7	7.65E-6	1.93E5	8.94E-8	1.85E-2
5E-7	1.61E-1	7.38E9	2.80E-11	3.87E-6	3.79E-5	6.10E5	1.11E-6	4.69E-3
1E-6	8.07E-2	3.38E9	1.28E-11	1.42E-5	1.39E-4	1.33E6	8.11E-6	1.66E-3
2E-6	4.07E-2	1.61E9	6.12E-12	5.40E-5	5.29E-4	2.79E6	6.19E-5	5.85E-5
5E-6	1.61E-2	6.26E8	2.38E-12	3.28E-4	3.21E-3	7.19E6	9.39E-4	1.48E-4
1E-5	8.07E-3	3.10E8	1.18E-12	1.30E-3	1.27E-2	1.45E7	7.44E-3	5.24E-5
2E-5	4.04E-3	1.54E8	5.86E-13	5.17E-3	5.06E-2	2.92E7	5.92E-2	1.85E-5

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