CHARGE DISTRIBUTIONS FOR RADIOACTIVE AEROSOLS IN A BIPOLAR ATMOSPHERE PERMEATED BY AN ELECTRIC FIELD

by

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CHAPTER I

Introduction

This thesis is an investigation of the electric charge distribution among aerosol particles in a bipolar atmosphere. The aerosol particles considered here have radii greater than 1×10^{-7}[m] but usually less than 1×10^{-5}[m]. The particles may be radioactive emitting beta rays. This radiation both ionizes atmospheric molecules and affects the charge on the particle. Furthermore, the atmosphere may be permeated by an electric field whose magnitude, in absence of significant ionization, may range up to, or be on the order of, 10^{6}[Vm^{-1}].

The particle charge distributions have been calculated previously under various conditions by different methods. These methods differ in the calculation of the ion-particle attachment coefficients. Bricard[1] has calculated the attachment coefficients for stationary particles with radii greater than 1×10^{-7}[m] in the absence of external electric fields. Fuchs[2] and Bricard[1] have extended these calculations to approximate the attachment coefficients for stationary particles with radii on the order of 1×10^{-8}[m]. These theories include effects due to the free flow of the ions one mean-free path length from the surface of the particle. Gunn[3] has calculated the attachment coefficients for the case involving large external electric fields, where the diffusion of ions is negligible. Klett[4] has approximated the attachment coefficients for the case where a weak external electric field is added to the continuum equations used by Bricard[1]. Precise definitions concerning the regions where the above calculations are valid will be presented in Chapter III.

Another approach to the calculation of the particle charge distribution has been presented by Keefe, Nolan, and Rich.[5] They argue that the aerosol particles may be considered in charge equilibrium according to the Boltzmann law. It will be illustrated in Chapter V that the more recent experiments agree with this formula.

The study of the particle charge distributions involves first examining the rate equations for the charge transfer interactions among the radioactive particles, ions, electrons, cloud droplets, and neutral gas molecules. These rate equations are examined in Chapter II. Chapter III contains an examination of the ion-particle attachment coefficients which govern the particle charge distributions. The method used to calculate the steady-state charge distributions is explained in Chapter IV. Asymptotic formulae for the particle charge distribution in various cases are then developed along with the mean charge and variance. Comparisons with the numerical and experimental results follow in Chapter V.

CHAPTER II

Rate Equations for Charge Transfer Interactions Among Radioactive Particles, Ions, Electrons, Cloud Droplets, and Neutral Gas Molecules

An overview of relevant interactions may be obtained by considering one radioactive particle and the physical phenomena in its immediate vicinity. The particle is considered at rest relative to the atmosphere and may emit beta rays with a typical energy of 1[MeV]. The beta rays collide with gas molecules, ejecting secondary electrons and creating positive ions, with an average energy loss of 33.7[eV][6] per ion pair. The electrons then combine with gas molecules to form negative ions.

The above species all interact with themselves and the cloud droplets. The rate equations governing the concentration of the various species can most easily be divided into three categories: 1) the rate equation governing the concentration of free electrons in the atmosphere, 2) the rate equations governing the concentrations of singly-charged ions, 3) the rate equations governing the concentrations of the charged radioactive particles.

The concentration of free electrons in the atmosphere is determined both by their production rate and by their attachment rate to other atmospheric species. The production rate depends upon both the primary beta decay and the secondary electron production. Secondary electrons are occasionally released in collisions between a primary electron and a gas molecule. These collisions leave behind a positive ion-electron pair and cost the primary an average energy of 33.7[eV][6]. The free electrons may then attach themselves to neutral gas molecules, aerosol particles, cloud droplets, and other atmospheric ions. The rate equation for the concentration of free electrons can be written symbolically as

$$\dot{n}_e = q_e - (\alpha_o n_o + \alpha_o' n + \beta N + \gamma C)n_e \quad , \tag{2.1}$$

where $q_e[m^{-3}s^{-1}]$ is the volumetric production rate of electrons, $\alpha_o[m^3s^{-1}]$ is the electron attachment coefficient to neutral molecules of concentration $n_o[m^{-3}]$, $\alpha_o'[m^3s^{-1}]$ is the electron attachment coefficient to atmospheric ions of concentration $n[m^{-3}]$, $\beta[m^3s^{-1}]$ is an effective electron attachment coefficient to particles of concentration $N[m^{-3}]$ (summation over charge states has been suppressed), and $\gamma[m^3s^{-1}]$ is an effective electron attachment coefficient to cloud droplets of concentration $C[m^{-3}]$ (summation over charge states has been suppressed). The steady state solution is obtained by setting $\dot{n}_e = 0$, it follows that

$$n_e = q_e(\alpha_o n_o + \alpha_o' n + \beta N + \gamma C)^{-1} \quad . \tag{2.2}$$

By examing the relevant magnitudes of attachment rates (see appendix A), we find $n_e \cong q_e(\alpha_o n_o)^{-1}$. This concentration is attained over a characteristic time scale $\tau \cong n_e q_e^{-1}$. Based upon the reactions enumerated by Phelps[7], the effective attainment rate at a temperature of 275°[K] and pressure 7.95×10^4[Pa] is $\alpha_o n_o \cong 5 \times 10^7[s^{-1}]$. Now if we contemplate values of $q_e \leq 10^{16}[m^{-3}s^{-1}]$, we find that $n_e \leq 2 \times 10^8[m^{-3}]$, which is several orders of magnitude less than the ion concentration (see fig. 1) Hence free electrons may be ignored under these conditions.

The concentrations of the singly charged ions $n_i[m^{-3}]$ are governed by the production rates, ion-ion recombinations, and ion attachment to both particles and droplets. The production rate of the negative ions is governed by attachment of free electrons onto the gas molecules, $\alpha_o n_o n[s^{-1}]$. However, the production rate of the positive ions will be given by the particle beta-decay rate $q[s^{-1}]$. This is due to creation of a positive ion and an electron by the primary beta particle. The accepted value for the ion-ion recombination coefficient, $\alpha_i[m^3 s^{-1}]$, is $1.6 \times 10^{-12}[m^3 s^{-1}]$[8]. The ion-ion recombination rate per unit volume is represented by $\alpha_i n_1 n_2 [m^{-3} s^{-1}]$. If we let $\beta_{1,j}[m^{-3} s^{-1}]$ and $\beta_{2,j}[m^{-3} s^{-1}]$ represent the coefficients for attachment of positive and negative ions, respectively, to particles with j electronic charges, then the total attachment rate of small ions to particles with total concentration $N[m^{-3}]$ may be written symbolically as

$$\beta N \equiv \sum_{j=-\infty}^{\infty} \beta_{1,j} N_j, \text{ and } \beta' N \equiv \sum_{j=-\infty}^{\infty} \beta_{2,j} N_j \quad . \tag{2.3}$$

A detailed discussion of the calculation of these coefficients $\beta_{i,j}$ is given in Chapter III. In the same spirit the attachment of small atmospheric ions to cloud droplets with total concentration $C[m^{-3}]$ and charge states j, each with concentration C_j, can be represented by

$$\gamma C \equiv \sum_{j=-\infty}^{\infty} \gamma_{1,j} C_j \quad , \text{ and } \gamma' C \equiv \sum_{j=-\infty}^{\infty} \gamma_{2,j} C_j \quad . \tag{2.4}$$

The rate equations can now be expressed as

$$\dot{n}_1 = q - (\alpha_i n_2 + \beta N + \gamma C) n_1 \quad , \tag{2.5}$$

and

$$\dot{n}_2 = \alpha_o n_o n_e - (\alpha_i n_1 + \beta' N + \gamma' C) n_2 \quad . \tag{2.6}$$

The steady-state ion concentrations are determined by setting $\dot{n}_1 = \dot{n}_2 = 0$, and by introducing the approximations $q \cong \alpha_e n_o n_e$, $\beta \cong \beta'$, and $\gamma \cong \gamma'$. The only physical solutions occur when $n_1 = n_2$ (Appendix B) with

$$n_1 = \chi(2\alpha_i)^{-1}[(1 + 4\alpha_i q\chi^{-2})^{1/2} - 1] \quad , \tag{2.7}$$

where $\chi \equiv \beta N + \gamma C$ is an overall ion attachment rate. It is instructive to note that when the ion production rate is large (i.e. $4\alpha_i q\chi^{-2} >> 1$) the ionic recombination dominates over attachment and $n \cong (q\alpha_i^{-1})^{1/2}$. In the opposite limit the attachment dominates over recombination and $n \cong q\chi^{-1}$. The discussion here will be restricted to cases for which $4\alpha_i q\chi^{-2} >> 1$, that is $n \cong (q\alpha_i^{-1})^{1/2}$. Equilibrium ion concentrations are shown in Figure 1.

The rate equations, which govern the concentrations of charged radioactive particles, are developed by considering the activity per particle, A_p, and the ion-particle attachment rate. The concentration of particles with charge state j, $N_j[m^{-3}]$, can be affected in six ways. A particle with (j-1) charges may beta decay, increasing the charge on the particle by one electron and become a j particle. A particle with j charges may decay and become a (j+1) particle. The ion-particle attachment also changes particles in and out of a j state to and from a (j-1) and a (j+1) state. Hence we need to solve coupled rate equations of the form

$$\dot{N}_j = A_p(N_{j-1}-N_j) + \beta_{1,j-1}n_1N_{j-1} - \beta_{1,j}n_1N_j + \beta_{2,j+1}n_2N_{j+1} - \beta_{2,j}n_2N_j \ , \tag{2.8}$$

along with the conservation of the total concentration of particles $N = \sum_{j=-\infty}^{\infty} N_j$. We expect that for a given physical system we can find a fixed $\ell >> 1$, such that for all $j > \ell$, $N_j \cong 0$. This enables us to delete all equations for $\dot{N}_{j+1}$, and $\dot{N}_{-j-1}$, where $j \geq \ell$. Furthermore, we should delete all terms involving transitions to and from $(\ell + 1)$ and $(\ell - 1)$ states. Hence we delete the term $(-A_pN_\ell - \beta_{1,\ell}n_1N_\ell + \beta_{2,\ell+1}n_2N_{\ell+1})$

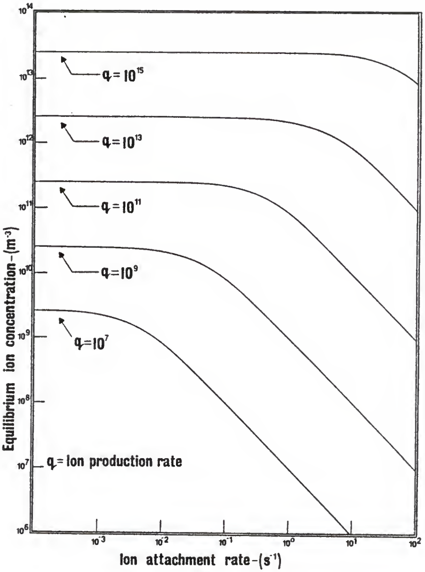
in the equation for $\dot{N}_\ell$, and the term $(A_p N_{-\ell-1} + \beta_{1,-\ell-1} n_1 N_{-\ell-1} - \beta_{2,-\ell} n_2 N_{-\ell})$ in the equation for $\dot{N}_{-\ell}$. The remaining system of $(2\ell + 1)$ equations, along with the relation $N = \sum_{j=-\ell}^{\ell} N_j$, can be solved analytically in the steady-state to obtain a recursion relation of the form

$$\beta_{2,j+1} n_2 N_{j+1} = (\beta_{1,j} n_1 + A_p) N_j \quad . \tag{2.9}$$

A detailed discussion of the coefficients $\beta_{i,j}$ will be given in Chapter III. The recursion relation (2.9), which expresses the principle of detailed balance, is the fundamental starting point for calculating the particle charge distributions in Chapter IV.

Figure 1.

Equilibrium ion concentrations as a function of the ion production rate and ion attachment rate.



Equilibrium ion concentration-(m^{-3})
Ion attachment rate-(s^{-1})
q = ion production rate
q = 10^{15}
q = 10^{13}
q = 10^{11}
q = 10^{9}
q = 10^{7}
10^{14}
10^{13}
10^{12}
10^{11}
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{-3}
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}

fig. 1

CHAPTER III

Ion-Particle Attachment Coefficients

The scheme used to calculate the ion-particle attachment coefficients, $\beta_{i,j}[m^{-3}s^{-1}]$, can be motivated as follows. We first define the $\beta_{i,j}$ through the equation

$$\dot{N}_j = \beta_{i,j}\bar{n}_i N_j \quad , \tag{3.1}$$

where $N_j[m^{-3}]$ is the concentration of particles with j electronic charges, and $\bar{n}_i[m^{-3}]$ is the uniform background concentration of the ionic species "i". Then, if $I_{i,j}[s^{-1}]$ is the total current of an ion of type "i" to a particle with j charges, we must also have $\dot{N}_j = I_{i,j}N_j$. Hence, the ion-particle attachment coefficient must be given by $\beta_{i,j} = I_{i,j}(\bar{n}_i)^{-1}$. The problem reduces to one of calculating $I_{i,j}$. To do this we assume a stationary particle with j electronic charges immersed in a sea of ions with overall background concentration $\bar{n}_1$ and $\bar{n}_2$. In the steady-state the ion concentrations will vary as a function of distance from the particle. Hence the current is given by

$$I_{i,j} = \int_S \vec{J}_i(\vec{r},j)\cdot d\vec{A} \quad , \tag{3.2}$$

where $\vec{J}_i[m^{-2}s^{-1}]$ is the current density of ions of type i, and S is any closed surface enclosing the entire particle. The calculation of $\vec{J}_i$ has been done for three cases of interest: 1) no externally applied electric field $\vec{E}_o[Vm^{-1}]$, 2) a weak external electric field $(eE_oa(kT)^{-1} << 1)$, and 3) a strong external electric field $(eE_oa(kT)^{-1} >> 1)$. In all cases the space charge in the vicinity of the droplet will be considered so disperse as to be negligible.

The case with no externally applied electric field has been done by Bricard[1]. The calculation of $I_{i,j}$ is obtained by solving the steady-state source-free continuity equation $\vec{\nabla} \cdot \vec{J}_i = 0$, with

$$\vec{J}_i = -D_i \vec{\nabla} n_i + s_i e \mu_i n_i \vec{E} \quad , \tag{3.3}$$

where $D_i[m^2 s^{-1}]$ is the diffusion coefficient for ions of type i, $n_i(\vec{r},j)[m^{-3}]$ is the concentration of ions of type i, s_i is +1 (-1) for positive (negative) ions, e[C] is the unit electronic charge, $\mu_i[m^2V^{-1}s^{-1}]$ is the electrical mobility of ions of type i, and $\vec{E}$ is the radial electric field due to the charge on the spherical particle. The accepted macroscopic boundary conditions are $n_i(a,j) = 0$ and $n_i(\infty,j) = \bar{n}_i$, where a[m] is the particle radius. For particles with radii larger than 1×10^{-8}[m] the image forces between the ion and perfect-conductor particle can be neglected[9,10]. In this case the ion-particle attachment coefficients are given by

$$\beta_{i,j} = s_i 4\pi a D_i I_i'(j) \quad , \tag{3.4}$$

where

$$I_i'(j) \equiv 2bj \, \{\exp(2jbs_i) - 1\}^{-1} \quad , \tag{3.5}$$

and 2b is defined by

$$2b \equiv e^2(4\pi\varepsilon_o akT)^{-1} \quad , \tag{3.6}$$

where ε_o is the permitivity of free space. It is instructive to note that 2bj is a ratio of a characteristic electrostatic potential energy of a spherical conductor of radius a and with charge ej, to a characteristic thermal energy kT.

The case involving a weak $(eE_o a(kT)^{-1} << 1)$ externally applied electric field has been investigated by Klett[4]. The source-free steady-state continuity equation is used to calculate $n_i(\vec{r},j)$. The form for $\vec{J}_i$ is identical to (3.3). However in this case $\vec{E}$ is the electric field surrounding a charged spherical conductor in an external field $\vec{E}_o$. Due to the analytic complexity of the problem, one must resort to the method of matched asymptotic approximations. The solution depends on two naturally arising nondimensional physical quantities $\alpha \equiv eE_o a(kT)^{-1}$, and $\tau \equiv 2bj$, where 2b is defined as in (3.6). The parameter α is the ratio of a characteristic electrostatic potential energy due to the external field $\vec{E}_o$, to the characteristic thermal energy kT. Both the "inner" solution, where diffusion dominates over conduction due to the external electric field, and the "outer" solution, where conduction due to the external electric field dominates over diffusion, are calculated to first order in α. The matched inner solution is used to calculate the ion-particle attachment coefficient. To first order in α the result is

$$\beta_{i,j} = s_i 4\pi a D_i \tau I_i''(j) \quad , \tag{3.7}$$

where

$$I_i''(j) \equiv \left(1 + \frac{s_i \alpha \tau}{2(1-\exp(-s_i\tau))}\right) \left(\frac{\exp(-s_i\tau)}{1-\exp(-s_i\tau)}\right) \quad . \tag{3.8}$$

Klett further argues that these formulae are valid for $\alpha \leq 4$.

The case involving a large ($\alpha >> 1$) externally applied electric field has been investigated by Gunn[3]. In this case ion diffusion to the droplet is negligible and the current is calculated from Ohm's law,

$$I_{i,j} = \sigma_i \int_S \vec{E}(j) \cdot d\vec{A} \quad , \tag{3.9}$$

where $\sigma_i[V^{-1}m^{-1}s^{-1}]$ is the electrical conductivity of the ion through the medium, $\vec{E}(j)[Vm^{-1}]$ is the electric field surrounding a charged spherical conductor in an external field $\vec{E}_o$, and the integration over

S is carried out over only that part of the particle for which the current is into the particle. Gunn's result is

$$\beta_{i,j} = 3\pi a^2 \sigma_i E_o (en_i)^{-1} \{1-(1/3)s_i E_j E_o^{-1}\}^2 \quad , \tag{3.10}$$

where

$$E_j \equiv je(4\pi\varepsilon_o a^2)^{-1} \ . \tag{3.11}$$

It should be emphasized that Gunn's result is valid only where the linear form of Ohm's law is applicable.

We have developed all of the formulae for the $\beta_{i,j}$'s by using the macroscopic laws for diffusion and conduction of ions. Particles which have dimensions smaller than, or on the order of, a mean free path length for ion-molecular collisions do not obey the continuum or macroscopic laws for diffusion and conduction. Hence, the $\beta_{i,j}$'s for these particles may be quite different. This aspect is discussed in greater detail in Chapter V. However, it should be noted that once the $\beta_{i,j}$'s have been specified, the particle charge distributions still may be calculated according to the prescription to be given in Chapter IV.

CHAPTER IV

Particle Charge Distributions In the Steady-State

The information for determining the particle charge distribution is contained in the rate equations for the charge transfer interactions among the ions, electrons, neutral gas molecules, and radioactive particles. Steady-state particle charge distributions can be calculated from the recursion relation (2.9).

$$\beta_{2,j+1} n_2 N_{j+1} = (\beta_{1,j} n_1 + A_p) N_j \quad . \tag{2.9}$$

Given the $\beta_{i,j}$, which were developed in Chapter III, the first step is to solve (2.9) for $N_{j+1} N_j^{-1}$. By using this ratio in a telescoping product we can form:

$$N_j N_0^{-1} = \frac{N_1}{N_0} \cdot \frac{N_2}{N_1} \cdot \quad \cdots \quad \cdot \frac{N_{j-1}}{N_{j-2}} \cdot \frac{N_j}{N_{j-1}} \quad . \tag{4.1}$$

The normalized charge distribution is then given by

$$N_j N^{-1} = (N_j N_0^{-1})(N_0^{-1} N)^{-1} = N_j N_0^{-1} \left\{ \sum_{\ell=-\infty}^{\infty} N_\ell N_0^{-1} \right\}^{-1} \quad . \tag{4.2}$$

This distribution may be evaluated numerically. Several normalized charge distributions have been calculated for various values of activity, electric field, and particle radius. These results will be illustrated in Chapter V.

An alternative to the numerical approach is to study the charge distributions under appropriate asymptotic limits. There are two physical mechanisms, radioactive decay and ion attachment, which can increase the positive charge on the particles. The relative magnitude of the associated rates determines whether the particle activity or the ion diffusion and conduction processes dominate the particle charging. Hence, we distinguish between two main cases of interest:

Case A - low activity $(A_p(\beta_{1,j}n_1)^{-1} << 1)$

Case B - high activity $(A_p(\beta_{1,j}n_1)^{-1} >> 1)$.

The particle charge distributions are developed by rewriting (2.9) for the two cases as:

(Case A)

$$N_j N_{j-1}^{-1} = \beta_{1,j-1}n_1(\beta_{2,j}n_2)^{-1}(1 + A_p(\beta_{1,j-1}n_1)^{-1}) \quad \text{, and} \qquad (4.3)$$

(Case B)

$$N_j N_{j-1}^{-1} = A_p(\beta_{2,j}n_2)^{-1}(1 + \beta_{1,j-1}n_1 A_p^{-1}) \qquad (4.4)$$

$$\cong A_p(\beta_{2,j}n_2)^{-1} \quad . \qquad (4.5)$$

In both cases, A and B, we have two subcases. The first subcase involves a weak external electric field ($\alpha \equiv eE_o a(kT)^{-1} << 1$). The second subcase involves a strong external electric field ($\alpha >> 1$), applicable where the linear form of Ohm's law is valid and ion-diffusion currents to the particle are negligible. One should be careful with Case B, since a strong external electric field is difficult to maintain in a highly ionized atmosphere. The case where $\vec{E}_o = 0$ can be included in the weak external electric field case by taking the limit when α approaches zero.

The first step to derive the particle charge distributions is to form the product (4.1), and then the normalized charge distribution $N_j N^{-1}$, which is given by (4.2). The four formulae for the particle charge distributions are calculated in the following order: 1) low activity and a weak external electric field, 2) low activity and a strong external electric field, 3) high activity and a weak external electric field, 4) high activity and a strong external electric field.

All cases involving low activity ($A_p(\beta_{1,j}n_1)^{-1} \ll 1$) are developed from (4.3). By using (4.3) and (4.1), we may express the general formula for all low activity cases in the form:

$$N_j N_0^{-1} = \rho^j S_j T_j \quad , \tag{4.6}$$

where we have defined $\rho \equiv n_1 n_2^{-1}$, $S_j \equiv \prod_{\ell=1}^{j} (\beta_{1,\ell-1}\beta_{2,\ell}^{-1})$, and $T_j \equiv \prod_{\ell=1}^{j} (1 + A_p(\beta_{1,\ell-1}n_1)^{-1})$. The problem is to evaluate S_j and T_j in an appropriate asymptotic form for the cases involving the weak and strong external electric fields. The case involving the weak external electric field is developed by using the $\beta_{i,j}$ defined by (3.7). By anticipating that $bj \ll 1$, we have to first order in α (Appendix C)

$$S_j \cong \eta^j (bj)^{-1} \exp\{-bj^2(1 - (1/2)\alpha)\} \sinh(bj) \quad , \tag{4.7}$$

and

$$T_j \cong 1 + 2bA_p\varepsilon_o j\sigma_1^{-1} \cong \exp\{2bjA_p\varepsilon_o\sigma_1^{-1}\} \quad , \tag{4.8}$$

where $\sigma_1 \equiv e\mu_i n_i$ is the electrical conductivity, and $\eta \equiv D_1 D_2^{-1}$. By combining all the factors we have

$$N_j N_0 = \eta^j (bj)^{-1} \sinh(bj) \exp\{-bj^2(1-(1/2)\alpha) + 2bjA_p\varepsilon_o\sigma_1^{-1}\} \quad , \tag{4.9}$$

which can be rewritten by letting $bj \ll 1$ and $\sinh(bj) \cong bj$. Hence we have

$$N_j N_0^{-1} = \exp\{j\ell n(\eta) - bj^2(1 - (1/2)\alpha) + 2bjA_p\varepsilon_o\sigma_1^{-1}\} \quad . \tag{4.10}$$

It should be mentioned that when $A_p = 0$, and $\alpha = 0$, (4.9) is identical to the formula derived by Sal'm[11]. Now, by using (4.10), we can complete the square in the exponential and approximate NN_0^{-1} by

$$NN_0^{-1} \doteq \int_{-\infty}^{\infty} (N_j N_0^{-1}) dj \quad . \tag{4.11}$$

We then obtain

$$N_j N = (N_j N_0^{-1})(NN_0^{-1})^{-1} \doteq (2\pi\overline{(\Delta j)^2})^{-1/2} \exp\{-(j-\bar{j})^2/(2\overline{(\Delta j)^2})\} \quad , \tag{4.12}$$

where

$$\bar{j} = (2b)^{-1}(1 + (1/2)\alpha)[\ln(D_1 D_2^{-1}) + A_p \varepsilon_o \sigma_1^{-1}] \quad , \tag{4.13}$$

and

$$\overline{(\Delta j)^2} = (2b)^{-1}(1 + (1/2)\alpha) \quad . \tag{4.14}$$

It is of interest to note that when $A_p = 0$ the charge distribution is the same one derived by Klett[4], and when $\alpha = 0$, we obtain the same distribution derived by Bricard[1].

The case involving low activity and a strong electric field is developed by using the $\beta_{i,j}$ defined by (3.10). We must approximate S_j and T_j in (4.6). By using the high field approximations $\gamma \equiv e^2(8\pi\varepsilon_o a)^{-1} \cdot (eaE_o)^{-1} << 1$, and $K \equiv A_p(3\pi a^2 \sigma_1 E_o e^{-1})^{-1} << 1$, we can approximate S_j and T_j by (Appendix C)

$$S_j \doteq (n_2 n_1^{-1})^j \exp\{-(4/3)\gamma j^2 + j\ln(\sigma_1 \sigma_2^{-1})\} \quad , \tag{4.15}$$

and

$$T_j \doteq \exp(jK) \quad . \tag{4.16}$$

By combining all terms, S_j and T_j, we have the approximation

$$N_j N_0^{-1} \cong \exp\{-(4/3)\gamma j^2 + j(\ell n[\sigma_1\sigma_2^{-1}] + K)\} \quad . \tag{4.17}$$

Now, by completing the square in the exponential, using the approximation (4.11), we arrive at a charge distribution in the same form as (4.12) with a new $\bar{j}$, and $\overline{(\Delta j)^2}$, given by

$$\bar{j} = 3\pi\varepsilon_o a E_o e^{-1} \ell n(\sigma_1\sigma_2^{-1}) + A_p \varepsilon_o \sigma_1^{-1} \quad , \tag{4.18}$$

and

$$\overline{(\Delta j)^2} = 3\pi\varepsilon_o a E_o e^{-1} \quad . \tag{4.19}$$

Comparisons of the above formulae with actual numerical calculations, involving weak electric fields, can be found in Chapter V. It is interesting to note that when $A_p = 0$, and $\sigma_1\sigma_2^{-1} \cong 1$, the result for $\bar{j}$ agrees with that derived by Gunn[3] (Appendix C).

The cases involving high activity $(A_p^{-1}(\beta_{1,j}n_1) << 1)$ are developed by using (4.5). The case involving a weak electric field ($\alpha << 1$) is developed by using the $\beta_{i,j}$ defined by (3.7). The telescoping product for $N_j N_0^{-1}$ now has an entirely different structure given by

$$N_j N_0^{-1} = (A_p \varepsilon_o \sigma_2^{-1})^j (j!)^{-1} S_j T_j^{-1} \quad , \tag{4.20}$$

where S_j and T_j have entirely new definitions given by

$$S_j \equiv \prod_{\ell=1}^{j} \{1-\exp(-2b\ell)\} \quad , \tag{4.21}$$

and

$$T_j \equiv \prod_{\ell=1}^{j} \{1 - \alpha b\ell[1-\exp(2b\ell)]^{-1}\} \quad . \tag{4.22}$$

Also, to obtain (4.20), the definition of electrical mobility and the Einstein relation were used to obtain the equation $8bn_2\pi a D_2 = \sigma_2\varepsilon_o^{-1}$.

If we note that $2bj >> 1$, then an approximation for $S_jT_j^{-1}$ is given by (Appendix D)

$$S_jT_j^{-1} \cong (1 - e^{-2bj})(\frac{e}{2bj})^{3/2}(\frac{1}{2})\exp(\frac{-\pi^2}{12bj})(1 - \frac{\alpha\pi^2}{24b}) \equiv C' \ . \tag{4.23}$$

The charge distribution is then given by

$$N_jN^{-1} = (j!)^{-1}\xi^j e^{-\xi}C_1 \quad , \tag{4.24}$$

where $C_1 \equiv C'e^{\xi}[1 + C(e^{\xi}-1)]^{-1}$, and $\xi \equiv A_p\varepsilon_o\sigma_2^{-1}$. The mean value of j is

$$\bar{j} = C_1\xi \quad , \tag{4.25}$$

and the variance is

$$\overline{(\Delta j)^2} = C_1\xi + C_1\xi^2(1 - C_1) \quad . \tag{4.26}$$

Values of C_1, as a function of ξ are given in Table 1. It is clear that C_1 may be taken as unity. Hence, we find that for large j

$$N_jN^{-1} = \xi^j(j!)^{-1}\exp(-\xi) \quad , \tag{4.27}$$

where ξ is the mean charge and also the variance of the Poisson distribution. It is important to notice that N_jN^{-1} depends only on ξ and j, neither of which depend on the electric field. Comparisons between the approximate formula and the numerically calculated distributions are given in Chapter V.

The case involving the high activity and strong external electric field is calculated by using the formula (4.5), where the $\beta_{i,j}$ are defined by (3.10). To perform this calculation we first note

$$N_jN_{j-1}^{-1} = K(1 + (2/3)j\gamma)^{-2} \tag{4.28}$$

where $K \equiv eAp(3\pi a^2 \sigma_2 E_o)^{-1}$, and $\gamma \equiv e^2(8\pi\varepsilon_o a)^{-1}\{eaE_o\}^{-1}$. It follows that

$$N_j N_0^{-1} = K^j P^{-2} \tag{4.29}$$

where $P \equiv \prod_{\ell=1}^{j} [1 + (2/3)\ell\gamma]$. Now take the logarithm of P and replace the product with a summation of logarithms. By using the strong field approximation, $\gamma << 1$, we can expand each logarithm to first order in γ and then calculate the summation exactly. After exponentiating the result, raising it to the (-2) power, and rewriting the factor K^j as an exponential of a logarithm, we obtain

$$N_j N_0^{-1} = \exp\{-(2/3)\gamma j^2 + j(\ell n(K) - (2/3)\gamma)\} \tag{4.30}$$

We can now complete the square in the exponential, make the same approximation as in (4.11), and obtain the same formula as (4.12). For this case the mean charge is

$$\bar{j} = 3(4\gamma)^{-1}\ell n\ \{A_p(3\pi a^2 \sigma_2 E_o e^{-1})^{-1}\} - (1/2) \tag{4.31}$$

and the variance is

$$\overline{(\Delta j)^2} = \frac{3}{4}\gamma^{-1} \quad . \tag{4.32}$$

Discussion concerning all four cases are found in Chapter V. Comparisons to numerical calculations, experimental results, and discussion of their physical interpretation are included.

TABLE 1.

Comparisons between the nondimensional quantity S_j, given by equation (4.21), and the asymptotic approximation for S_j, $\tilde{S}_j$, equation (D.2). Also included are the values for the nondimensional coefficient C_1, in equation (4.24), as a function of the quantity ξ, where $\xi \equiv A_p \varepsilon_o \sigma_2^{-1}$.

TABLE 1.

j	S_j	$\tilde{S}_j$	ξ	C_1
1	5.8×10^{-2}	7.2×10^{-5}	10	2.3×10^{-1}
5	6.0×10^{-5}	2.5×10^{-6}	20	3.0×10^{-3}
10	4.5×10^{-7}	1.0×10^{-7}	25	4.3×10^{-1}
20	2.5×10^{-9}	1.7×10^{-9}	30	9.9×10^{-1}
50	2.9×10^{-11}	2.5×10^{-11}	40	1.00
100	1.3×10^{-11}	1.1×10^{-11}	50	1.00
150	1.3×10^{-11}	1.1×10^{-11}		

CHAPTER V

Discussion and Conclusions

The particle charge distributions are calculated from the steady-state recursion relations (2.9),

$$\beta_{2,j+1}n_2N_{j+1} = (\beta_{1,j}n_1 + A_p)N_j \quad . \tag{2.9}$$

The ratio $N_{j+1}N_j^{-1}$ is used to calculate the distribution by noting (4.2)

$$N_jN = \left(\prod_{\ell=1}^{j} N_jN_{j-1}^{-1}\right)\left(\sum_{\ell=-\infty}^{\infty}[\prod_{k=1}^{\ell} N_kN_{k-1}^{-1}]\right)^{-1} . \tag{5.1}$$

This calculation can be done to arbitrary precision on a computer. However, several cases of interest can be examined by using the closed asymptotic formulae which were developed in Chapter IV. Application of the Central Limit Theorem allows all asymptotic formulae to be represented approximately in terms of the shifted Gaussian distribution,

$$N_jN^{-1} = \left(2\pi\overline{(\Delta j)^2}\right)^{-\frac{1}{2}}\exp\left(-(j-\bar{j})^2/(2\overline{(\Delta j)^2})\right) \quad , \tag{5.2}$$

with number of charges, $\bar{j}$, and dispersion, $\overline{(\Delta j)^2} \equiv \overline{(j-\bar{j})^2}$. The cases can be summarized as follows:

A. Low Activity $(A_p(\beta_{1,j}n_1)^{-1} << 1)$

a) weak external electric field $(eE_oa(kT)^{-1} << 1)$

$$\bar{j} = (2b)^{-1}\{\ell n(D_1D_2^{-1}) + A_p\varepsilon_o\sigma_1^{-1}\}\{1+(1/2)\alpha\} \quad , \tag{4.13}$$

$$\overline{(\Delta j)^2} = (2b)^{-1}\{1 + (1/2)\alpha\} \quad ; \tag{4.14}$$

b) strong external electric field $(eE_o a(kT)^{-1} \gg 1)$

$$\bar{j} = 3(8\gamma)^{-1}\ln(\sigma_1\sigma_2^{-1}) + A_p\varepsilon_o\sigma_1^{-1} , \tag{4.18}$$

$$\overline{(\Delta j)^2} = 3(8\gamma)^{-1} \tag{4.19}$$

B. High Activity $(A_p^{-1}(\beta_{1,j}n_1) \ll 1)$

a) weak external electric field $(eE_o a(kT)^{-1} \ll 1)$

$$\bar{j} = A_p\varepsilon_o\sigma_2^{-1} , \tag{4.25}$$

$$\overline{(\Delta j)^2} = A_p\varepsilon_o\sigma_2^{-1} ; \tag{4.26}$$

b) strong external electric field $(eE_o a(kT)^{-1} \gg 1)$

$$\bar{j} = 3(4\gamma)^{-1}\ln \{A_p(3\pi a^2\sigma_2 E_o e^{-1})\} - (1/2) \tag{4.31}$$

$$\overline{(\Delta j)^2} = 3(4\gamma)^{-1} . \tag{4.32}$$

The low activity case has two terms which determine the value of $\bar{j}$. The term $A_p(\sigma_1\varepsilon_o)^{-1}$ is the ratio of the activity per particle to the characteristic electrical relaxation rate of the positive ions in the atmosphere. If A_p is increased, then the mean positive charge on the particle increases. Another important term which determines $\bar{j}$ is $\ln(D_1D_2^{-1})$, or equivalently $\ln(\sigma_1\sigma_2^{-1})$. If $D_1 > D_2$ then the mean charge on the particle is driven positive. Similarly, if $D_2 > D_1$ the mean charge on the particle is driven negative.

The high activity case depends essentially on the parameter $A_p(\sigma_2\varepsilon_o)^{-1}$, the ratio of the activity per particle to the characteristic electrical relaxation rate associated with negative ions. If the negative ions' electrical conductivity increases, the mean charge on the particles will decrease. It is interesting that for the case involving a strong external electric field, we can rewrite

$$(3/2)\gamma A_p \varepsilon_o \sigma_2{}^{-1} = 3A_p e(16\pi E_o a^2 \sigma_2)^{-1} \quad . \tag{5.3}$$

Hence for large E_o, or σ_2, the mean particle charge may be negative.

The asymptotic formulae have been checked against numerical calculations. Results for several cases are shown in Figures 2 through 8. The histograms are computer generated results, the smooth curves are calculations based on the asymptotic formulae. It should be pointed out, that Figures 2 through 6 were calculated using the low activity approximation. Figure 7 uses the high activity approximation. Figure 8 illustrates the breakdown of the asymptotic formulae when used incorrectly. In Figure 8 the weak electric field, no activity, approximation was used to calculate the smooth curve. The failure occurs since $\alpha \cong .42$ and is not sufficiently small, or less than 1.

Several experiments have been performed to observe the charged aerosols. The experiments measure the relative fraction of neutral particles, in a bipolar atmosphere without an external electric field. A summary of the experiments by Liu and Pui[12] are given in Figure 9. The ion-particle attachment coefficients, which are listed in Chapter III, correspond to the continuum model without electrical image forces. For particle radii greater than, or on the order of, $1x10^{-7}$[m], the ion-particle attachment coefficients agree with the coefficients as calculated by using the model developed by Fuchs[9]. According to Figure 9, the theoretical and experimental results, are in agreement for these particle radii.

Much work is found in the literature involving the theoretical calculation of the ion-particle attachment coefficients for particle radii on the order of 1×10^{-8}[m]. The theory for particles whose radii are on the same order of magnitude as λ, the ion mean free path, is usually developed by what is called the limiting sphere method. In this method the space surrounding the particle is divided into two regions separated by an imaginary sphere of radius $\delta \doteqdot a+\lambda$, where "a" is the particle radius. The ions far from the sphere move by means of macroscopic diffusion. Inside the sphere of radius δ, the ions move by molecular transport or free flow. The method by which the ion currents toward the particle, inside and outside the sphere, are matched and developed give varying theories. Two examples of such theories are Fuchs[2], and Keefe[14] <u>et</u>. <u>al.</u> However, as seen from Figure 9, the formula which best agrees with the experimental results is the Boltzmann distribution as given in the paper by Keefe, Nolan, and Rich.[5] Also the more recent measurements by Liu and Pui[12], and by Servaas and Krider[13], support the conclusion that the Boltzmann distribution,

$$N_j = 2N_o \exp\{-j^2 e^2 (8\pi E_o akT)^{-1}\} \quad , \tag{5.4}$$

gives the best fit to the observed particle-charge distribution, particularly for radii less than 1×10^{-7}[m]. The theoretical justification for this has not yet been found. However, attempts to justify this distribution for all particle sizes have been chiefly done by Keefe, Nolan, and Rich[5]. The fact that the simple Boltzmann distribution agrees with the experiments implies that my asymptotic distributions, which are of the same form, may well predict the actual distributions for the small particle sizes. This extension has no experimental justification, however, the experimental trends suggest that these distributions will give accurate predictions.

FIGURE 2.

The relative concentration of particles as a function of particle charge. The histogram is computer generated and the smooth curve represents the asymptotic formula. The particle radius is 1[μm], the specific activity is $A = 0$, the external electric field is $\vec{E} = 0$, the temperature is 275°[K], and the atmospheric pressure is 7.95×10^4[Pa].

FIGURE 3.

The relative concentration of particles as a function of particle charge. The histogram is computer generated and the smooth curve represents the asymptotic formula. The particle radius is 1[μm], the specific activity is $A = 0$, the external electric field is $E = 10^3[Vm^{-1}]$, the temperature is 275°[K], and the atmospheric pressure is 7.95×10^4[Pa].

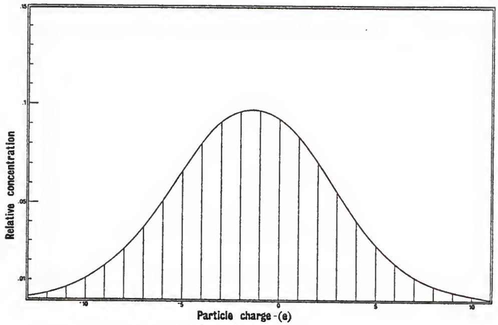

.15
.1
.05
.01
Relative concentration
-10
-5
0
5
10
Particle charge -(e)

fig.2

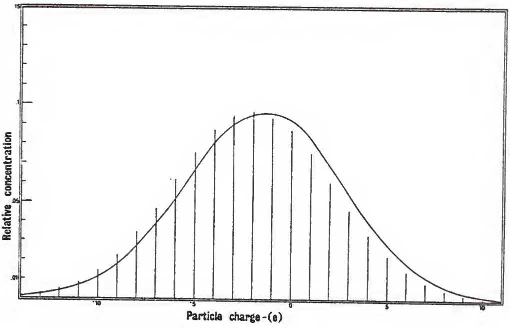

.15
.1
.05
.01
Relative concentration
-10
-5
0
5
10
Particle charge -(e)

fig.3

FIGURE 4.

The relative concentration of particles as a function of particle charge. The histogram is computer generated and the smooth curve represents the asymptotic formula. The particle radius is .1[μm], the specific activity is A = 0, the external electric field is $\vec{E} = 0$, the temperature is 275°[K], and the atmospheric pressure is 7.95 x 10^4[Pa].

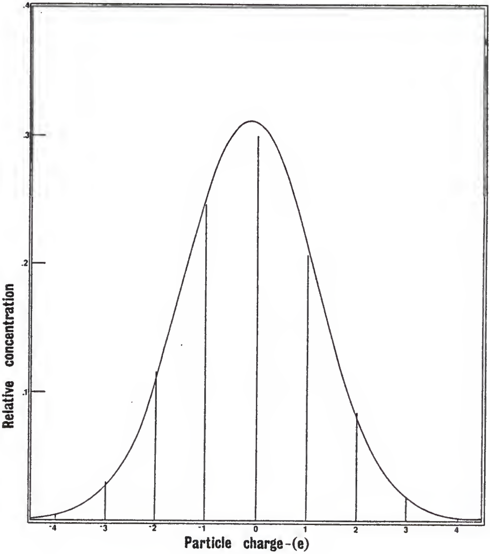


fig.4

FIGURE 5.

The relative concentration of particles as a function of particle charge. The histogram is computer generated and the smooth curve represents the asymptotic formula. The particle radius is .1[μm], the specific activity is $A = 10^6$, the external electric field is $\vec{E} = 0$, the temperature is 275°[K], and the atmospheric pressure is 7.95×10^4[Pa].

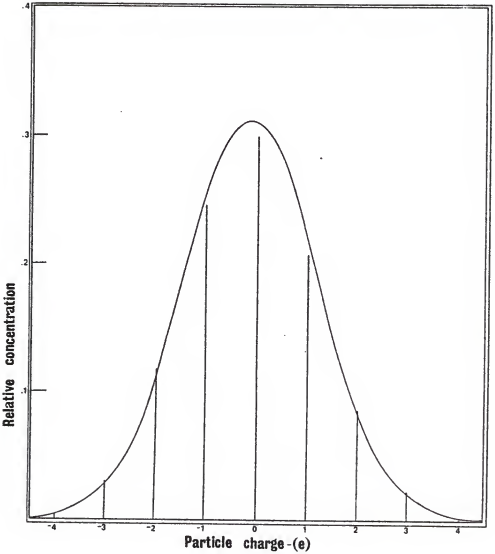


fig.5

FIGURE 6.

The relative concentration of particles as a function of particle charge. The histogram is computer generated and the smooth curve represents the asymptotic formula. The particle radius is .1[μm], the specific activity is $A = 10^{16}$, the external electric field is $E = 10^{4}[Vm^{-1}]$, the temperature is 275°[K], and the atmospheric pressure is 7.95×10^{4}[Pa].

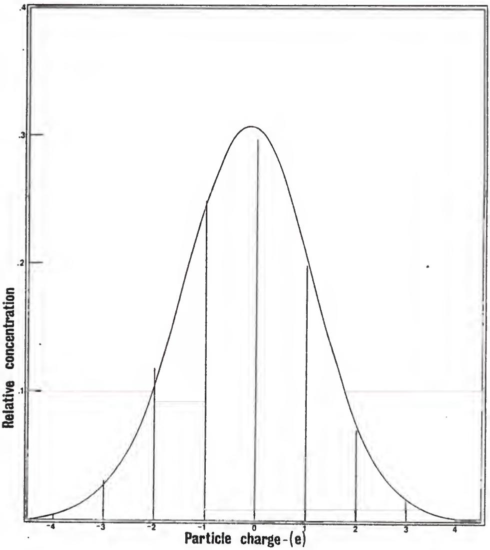


fig.6

FIGURE 7.

The relative concentration of particles as a function of particle charge. The histogram is computer generated and the smooth curve represents the asymptotic formula. The particle radius is .2[μm], the specific activity is $A = 10^{20}$, the external electric field is $\vec{E} = 0$, the temperature is 275°[K], and the atmospheric pressure is 7.95×10^4[Pa].

FIGURE 8.

The relative concentration of particles as a function of particle charge. The histogram is computer generated and the smooth curve represents the asymptotic formula. The particle radius is 1[μm], the specific activity is $A = 0$, the external electric field is $E = 10^4[Vm^{-1}]$, the temperature is 275°[K], and the atmospheric pressure is 7.95×10^4[Pa].

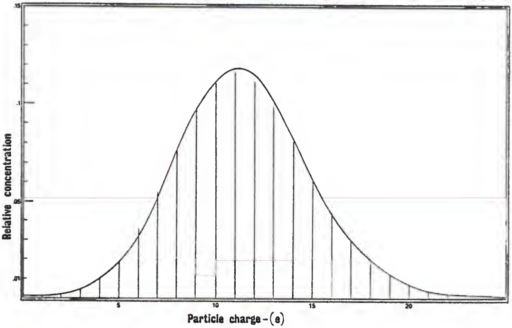

.15
.1
.05
.01
Relative concentration
5
10
15
20
Particle charge-(e)

fig.7

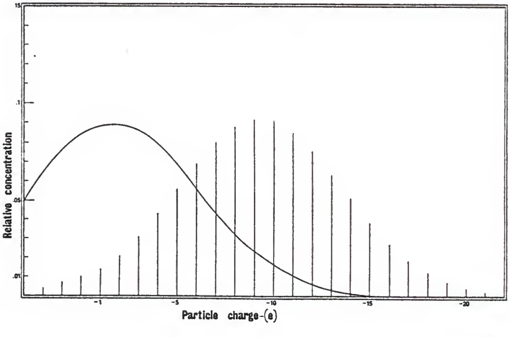
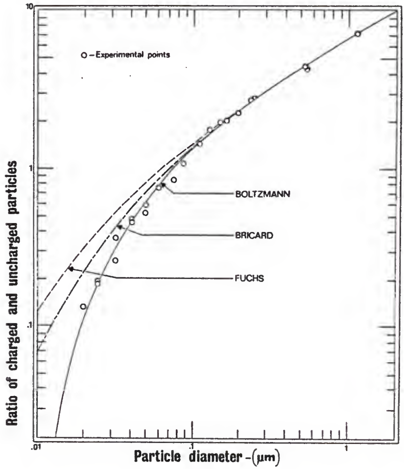

.15
.1
.05
.01
Relative concentration
-1
-5
-10
-15
-20
Particle charge-(e)

fig.8

FIGURE 9.

Comparisons between the experimental results, the Boltzmann distribution[5], Bricard's theory[1], and Fuchs' theory. This figure is a modified version of one occuring in the paper by Liu and Pui[12].



10
1
.1
.01
.1
1
O – Experimental points
BOLTZMANN
BRICARD
FUCHS
Ratio of charged and uncharged particles
Particle diameter -(μm)

fig.9

APPENDIX A

Electron Interactions and Attachment Rates

The rate equation for the concentration of free electrons can be written as

$$\dot{n}_e = q_e - (\alpha_o n_o + \alpha_o' n + \beta N + \gamma C) n_e \quad , \qquad \text{(A.1)}$$

where q_e is the electron production rate density, $\alpha_o n_o$ is the attachment rate to neutral molecules, $\alpha_o' n$ is the attachment rate to all other ionic species, βN is an effective attachment rate to particles, and γC is an effective attachment rate to cloud droplets. The electron attachment to gas molecules occurs by three-body processes, almost all of which generate[7] O_2^- . The combination of the various processes yields an effective attachment rate of $\alpha_o n_o \cong 5 \times 10^7 [s^{-1}]$. The electron-ion attachment studied by Biondi[15], yields an effective attachment coefficient of $\alpha_o' \cong 5 \times 10^{-3} [m^3 s^{-1}]$. An estimate for the electron-particle, or electron-droplet, attachment coefficient is $\beta \cong 4\pi a D_e$, where a is the radius of the collector and $D_e \cong 1.4 \times 10^{-2} [m^2 s^{-1}]$ is the diffusion coefficient for electrons. Hence, for a 1[μm] particle and a 10[μm] droplet we get $\beta \cong 1.8 \times 10^{-7} [m^3 s^{-1}]$ and $\gamma \cong 1.8 \times 10^{-6} [m^3 s^{-1}]$. Now using typical values of $n \cong 8 \times 10^{13} [m^{-3}]$ (see Figure 1), $N \cong 1 \times 10^{10} [m^{-3}]$, and $C \cong 3 \times 10^8 [m^{-3}]$, we have $\alpha_o' n \cong 40 [s^{-1}]$, $\beta_e N \cong 1.8 \times 10^3 [s^{-1}]$, and $\gamma_e C \cong 5.4 \times 10^2 [s^{-1}]$, all of which are small compared with $\alpha_o n_o$. Hence, we can approximate

$$\dot{n}_e \cong q_e - \alpha_o n_o n_e \qquad \text{(A.2)}$$

Hence, the steady-state concentration of electrons is given by $n_e = q_e(\alpha_o n_o)^{-1}$. If we now take $q_e \cong 1 \times 10^{16}[m^{-3}s^{-1}]$, we find that $n_e \cong 2 \times 10^8[m^{-3}]$. Comparing this with the ion concentration (Figure 1), we see that the free electrons may be ignored.

APPENDIX B

Solution of Rate Equations for Steady-State Ion Concentrations

The rate equations governing the concentrations of small atmospheric ions are expressed as

$$\dot{n}_1 = q - (\alpha n_2 + \beta N + \gamma C)n_1 \quad , \tag{B.1}$$

$$\dot{n}_2 = \alpha_e n_o n_e - (\alpha n_1 + \beta' N + \gamma' C)n_2 \tag{B.2}$$

where $q \cong \alpha_e \alpha_o n_e$ are the ion production rates, $\alpha n_1 n_2$ is the ion-ion recombination rate per unit volume, $\beta N \cong \beta' N$ are the attachment rates to the particles, and $\gamma C \cong \gamma' C$ are the attachment rates to the droplets. To solve this in the steady-state we let $\dot{n}_1 = \dot{n}_2 = 0$, $\gamma \cong \gamma'$, $\beta \cong \beta'$, and $q = \alpha_e n_o n_e$. It is clear

$$0 = (n_1 \dot{n}_2) = (n_1 + n_2)(q - \alpha n_1 n_2) - 2n_1 n_2 \chi \tag{B.3}$$

$$0 = (n_1 \dot{+} n_2) = 2q - (2\alpha n_1 n_2 + (n_1 + n_2)\chi) \tag{B.4}$$

where $\chi \equiv \beta N + \gamma C$. Solving (B.4) for $n_1 + n_2$, substituting into (B.3), and solving (B.3) for $n_1 n_2$ we obtain

$$C_o \equiv (n_1 n_2) = q\alpha^{-1} + (\chi^2(2\alpha^2)^{-1})[1 \pm (1 + 4\alpha q\chi^{-2})^{1/2}] \tag{B.5}$$

and

$$B \equiv -(n_1 + n_2) = \chi\alpha^{-1}(1 \pm [1 + 4\alpha q\chi^{-2}]^{1/2}) \quad . \tag{B.6}$$

Hence, we must have

$$n_1 = \frac{1}{2}[-B \pm (B^2 - 4C_o)^{1/2}] \quad . \tag{B.7}$$

Inspection shows that $B^2 - 4C_o \equiv 0$. Hence $n_1 = -\frac{1}{2}B$, which implies $n_1 = n_2$. This enables (B.1) to be written as a quadratic equation in terms of $n_1 = n_2 = n$. Solving this for n, we find

$$n = \frac{1}{2}\chi\alpha^{-1}[1 + (1 + 4\alpha q\chi^{-2})^{1/2}]. \qquad (B.8)$$

The limiting cases, $n \cong (q\alpha^{-1})^{1/2}$, and $n \cong q\chi^{-1}$, follow when $4\alpha q\chi^{-2} >> 1$, and $4\alpha q\chi^{-2} << 1$, respectively.

APPENDIX C

Approximations in the Low Activity Cases

The low activity cases occur when we have $A_p(\beta_{1,j}n_1)^{-1} << 1$. By using (4.3) and (4.1) we know that the general formula for all low activity cases is (4.6)

$$N_j N_0^{-1} = \rho^j S_j T_j \quad , \tag{4.6}$$

where we have defined $\rho = n_1 n_2^{-1}$, $S_j \equiv \prod_{\ell=1}^{j} (\beta_{1,\ell-1}\beta_{2,\ell}^{-1})$, and $T_j \equiv \prod_{\ell=1}^{j} (1 + A_p(\beta_{1,\ell-1}n_1)^{-1})$. The problem is to evaluate S_j and T_j for the cases involving the weak ($\alpha << 1$) and strong ($\alpha >> 1$) external electric fields.

The case involving the weak external electric field is developed by using the $\beta_{i,j}$ defined by (3.7). The approximation of S_j is done as follows. We note that

$$\beta_{1,\ell-1}\beta_{2,\ell}^{-1} = \eta(\ell-1)I_1''(\ell-1)[-\ell I_2''(\ell)]^{-1} \quad , \tag{C.1}$$

where we have defined $\eta \equiv D_1 D_2^{-1}$, and we recall from (3.7) that

$$I_i''(\ell) \equiv \left(1 + \frac{s_i \alpha 2b\ell}{2(1-\exp\{-s_i 2b\ell\})}\right)\left(\frac{\exp(-s_i 2b\ell)}{1-\exp(-s_i 2b\ell)}\right) \quad . \tag{3.8}$$

It is convenient to define the function

$$F(\ell) \equiv (\ell)^{-1}(1 - \exp(-2b\ell)) \quad , \tag{C.2}$$

where $F(0) \equiv \lim_{\ell \to o} F(\ell) = 2b$. It follows that

$$(\ell-1)I_1''(\ell-1)[-\ell I_2''(\ell)]^{-1}$$

$$= F(\ell)[F(\ell-1)]^{-1}[1 + \frac{1}{2}\alpha(2b)(\ell-1)\{1-\exp(-2b(\ell-1))\}^{-1}]$$

$$\cdot[1 + \frac{1}{2}\alpha(2b\ell)\{\exp(2b\ell)-1\}^{-1}]^{-1}\exp\{2b(\ell-1)\} \quad . \tag{C.3}$$

We define the two quantities

$$U_j \equiv \prod_{\ell=1}^{j} [1 + \frac{1}{2}\alpha(2b(\ell-1))\{1-\exp(-2b(\ell-1))\}^{-1}] \quad , \tag{C.4}$$

and

$$V_j \equiv \prod_{\ell=1}^{j} [1 + \frac{1}{2}\alpha(2b\ell)\{\exp(2b\ell) - 1\}^{-1}] \quad . \tag{C.5}$$

It follows

$$S_j = (\eta)^j F(j)[F(o)]^{-1}\exp(-bj(j-1))U_j V_j^{-1} \quad . \tag{C.6}$$

Expand $U_j V_j^{-1}$, keeping only terms of first order in α, to obtain

$$U_j V_j^{-1} \cong 1 + \frac{1}{2}\alpha[1 + bj^2-bj-2bj(\exp(2bj)-1)^{-1}] \quad . \tag{C.7}$$

By anticipating $bj << 1$, we approximate $U_j V_j^{-1}$ by

$$U_j V_j^{-1} \cong 1 + (1/2)\alpha bj^2 \cong \exp(\frac{1}{2}\alpha bj^2) \quad . \tag{C.8}$$

A useful identity, easily proved by direct substitution, is

$$F(j)[F(0)]^{-1}\exp(-bj(j-1)) = (bj)^{-1}\exp(-bj^2)\sinh(bj) \quad . \tag{C.9}$$

Hence we find that an approximation for the product S_j is

$$S_j \cong \eta^j (bj)^{-1}\exp\{-bj^2(1-(1/2)\alpha) \sinh(bj) \quad . \tag{4.7}$$

We now need to approximate T_j. Expand T_j by using the low activity assumption $(A_p(\beta_{1,\ell}n_1)^{-1} << 1)$. Furthermore, since $\alpha << 1$, we neglect terms of order $\alpha A_p(\beta_{1,\ell}n_1)^{-1}$. Hence, we have

$$T_j \cong 1 + A_p(4\pi a n_1 D_1)^{-1}\sum_{\ell=1}^{j} (\exp(2b(\ell-1))-1)[2b(\ell-1)]$$

$$= 1 + 2bA_p\varepsilon_o\sigma_1^{-1}\sum_{\ell=1}^{j} [(\exp\{2b(\ell-1)\}-1)(2b(\ell-1))^{-1}] \quad , \tag{C.10}$$

where $n_1D_1 = kT\sigma_1e^{-2}$ follows from the Einstein relation, $D(kT)^{-1} = \mu e^{-1}$, and the definition of electrical conductivity, $\sigma = e\mu n$. Now, by using the approximation $2bj << j$, we can expand (C.10) and keep only terms of first order in b to obtain

$$T_j \cong 1 + 2bA_p\varepsilon_o j\sigma_1^{-1}$$

$$\cong \exp\{2bA_p\varepsilon_o j\sigma_1^{-1}\} \quad . \tag{4.8}$$

The case involving the strong external electric field is developed by using the $\beta_{1,j}$ defined by (3.10). We must approximate S_j, and T_j, in (4.6), by using the high field approximations $\gamma << 1$, and $K << 1$, where we have defined

$$\gamma \equiv (e^2(8\pi\varepsilon_o a)^{-1})[eaE_o]^{-1} \quad , \tag{C.11}$$

and

$$K \equiv eA_p(3\pi a^2\sigma_2E_o)^{-1} \quad . \tag{C.12}$$

The approximation for S_j is done first. We first note

$$\beta_{1,j}\beta_{2,j+1}^{-1} = (n_2\sigma_1)(n_1\sigma_2)^{-1}(1-\tfrac{2}{3}\gamma j)^2(1+\tfrac{2}{3}\gamma(j+1))^{-2} \quad . \tag{C.13}$$

Now by using the high field approximation, we can approximate S_j to first order in γ as

$$S_j = [(n_2\sigma_1)(n_1\sigma_2)^{-1}]^j[\prod_{\ell=1}^{j} \{1-(2/3)\gamma(2\ell-1)\}]^2$$

$$\equiv [(n_2\sigma_1)(n_1\sigma_2)^{-1}]^j P^2 \quad . \tag{C.14}$$

Now, by taking the natural logarithm of P, we can replace the produce with a summation of logarithms which can also be approximated since $\gamma << 1$. After evaluating the summation, exponentiating it and completing the square, we find

$$S_j \cong [(n_2\sigma_1)(n_1\sigma_2)^{-1}]^j \exp(-\tfrac{4}{3}\gamma j^2) = (n_2 n_1{}^{-1})^j \exp\{-\tfrac{4}{3}\gamma j^2 + j\ln(\sigma_1\sigma_2{}^{-1})\} \tag{4.15}$$

We can now approximate T_j by first noting

$$\{1 + A_p(\beta_{1,\ell}n_1)^{-1}\} = 1 + K[1-\tfrac{2}{3}\gamma j]^2 \quad . \tag{C.15}$$

We now notice that $K << 1$, since we have low activity and a strong electric field. This implies that the terms of order γK can be neglected. Hence, we have

$$T_j \cong \prod_{\ell=1}^{j} (1 + K) \quad . \tag{C.16}$$

Now we can take the natural logarithm of T_j and replace the product by a sum of logarithms, which can be expanded for small K. After summing and exponentiating we find

$$T_j \cong \exp(jK) \quad . \tag{4.16}$$

As shown in Chapter IV, it follows that

$$\bar{j} = \overline{(\Delta j)^2}\ln(\sigma_1\sigma_2{}^{-1}) + A_p\varepsilon_o\sigma_1{}^{-1} \quad , \tag{4.18}$$

where

$$\overline{(\Delta j)^2} = 3\, a^2 \varepsilon_o E_o e^{-1} \tag{4.19}$$

It has been shown[3] that for strong external electric fields, with non-radioactive aerosols, the mean charge is given by

$$\frac{\bar{j} e}{12\pi\varepsilon_o a^2 E_o} = \frac{(\sigma_1\sigma_2^{-1})^{1/2} - 1}{(\sigma_1\sigma_2^{-1})^{1/2} + 1} \quad . \tag{C.17}$$

By using (4.18) and (4.19), when $A_p = 0$, we obtain

$$\bar{j} = \frac{3\pi\varepsilon_o a^2 E_o}{e} \ln(\sigma_1\sigma_2^{-1}) = \frac{6\pi\varepsilon_o a^2 E_o}{e} \ln((\sigma_1\sigma_2^{-1})^{1/2}) \quad . \tag{C.18}$$

Now recall the formula (valid for all $x > o$)

$$\ln(x) = 2\left(\frac{x-1}{x+1} + \frac{1}{3}\left(\frac{x-1}{x+1}\right)^3 + \frac{1}{5}\left(\frac{x-1}{x+1}\right)^5 + \cdot\cdot\cdot\right) \quad . \tag{C.19}$$

Hence, if $(\sigma_1\sigma_2^{-1}) \doteq 1$, we have

$$\ln((\sigma_1\sigma_2^{-1})^{1/2}) \doteq 2\left(\frac{(\sigma_1\sigma_2^{-1})^{1/2} - 1}{(\sigma_1\sigma_2^{-1})^{1/2} + 1}\right) \quad , \tag{C.20}$$

so we have

$$\bar{j} \doteq \frac{12\pi\varepsilon_o a^2 E_o}{e}\left(\frac{(\sigma_1\sigma_2^{-1})^{1/2} - 1}{(\sigma_1\sigma_2^{-1})^{1/2} + 1}\right) \quad , \tag{C.21}$$

which agrees with (C.17).

APPENDIX D

Approximations in the High Activity Case

The high activity case occurs when we have $A_p(\beta_{1,j}n_1)^{-1} >> 1$. By using (4.5), and the $\beta_{i,j}$ defined by (3.7), the telescoping product for $N_jN_0^{-1}$ is given by (4.20),

$$N_jN_o^{-1} = (A_p\varepsilon_o\sigma_2^{-1})^j(j!)^{-1}S_jT_j^{-1} \quad , \tag{4.20}$$

where

$$S_j \equiv \prod_{\ell=1}^{j} \{1-\exp(-2b\ell)\} \quad , \tag{4.21}$$

and

$$T_j \equiv \prod_{\ell=1}^{j} \{1 - \alpha b\ell[1-\exp(2b\ell)]^{-1}\} \quad . \tag{4.22}$$

We can now approximate S_j by first taking its natural logarithm, which reduces the product to that of a summation of logarithms. This summation can be replaced by an integration. However, calculations show this to be too crude. A good method to approximate this summation is obtained by looking only at the last (j-1) terms of the summation. Since any integral can be approximated by both upper and lower Riemann sums, we notice that

$$\int_2^{j+1} G(\ell)d\ell < \sum_{\ell=2}^{j} G(\ell) < \int_1^{j} G(\ell)d\ell \quad , \tag{D.1}$$

where $G(\ell) \equiv \ell n(1-\exp(-2b\ell))$. These integrals may be evaluated directly, added to the first term of the summation, and then exponentiated to obtain two different approximations for S_j. The final approximation for S_j is then obtained by taking the geometric mean of these two approximations. The result is

$$\tilde{S}_j \equiv (1-\exp(-2b))(\frac{e}{2b})^{3/2}(\frac{1}{2})\exp\{(2b)^{-1}(\exp(-2bj)-\pi^2/b)\} \quad . \tag{D.2}$$

This approximation is compared to the actual products in Table I. It is clear that the approximation is good for $j \gtrsim 50$. We now must approximate T_j. Expanding (4.21), and keeping only terms of first order in α, we obtain

$$T_j \cong 1 + \alpha b \sum_{\ell=1}^{j} \{\ell(\exp(2b\ell)-1)^{-1}\} \quad . \tag{D.3}$$

If the summation is replaced by an integration we find that the resulting integral can only be evaluated numerically. However, we can approximate the summation by first noting

$$\int_0^\infty x(e^x-1)^{-1}dx = \pi^2/6 \quad , \tag{D.4}$$

$$\int_1^{j+1} \frac{\ell d\ell}{e^{2b\ell}-1} < \sum_{\ell=1}^{j} \ell\{\exp(2b\ell)-1\}^{-1} < \int_0^j \frac{\ell d\ell}{e^{2b\ell}-1} \quad , \tag{D.5}$$

$$\int_0^j \frac{\ell d\ell}{e^{2b\ell}-1} = (2b)^{-2}\left(\frac{\pi^2}{6} - \int_{2bj}^\infty x(e^x-1)^{-1}dx\right) \quad , \tag{D.6}$$

and

$$\int_1^{j+1} \frac{\ell d\ell}{e^{2b\ell}-1} \cong (2b)^{-2}\left(\frac{\pi^2}{6} - \int_{2b(j+1)}^\infty \frac{xdx}{e^x-1} - \int_o^{2b} \frac{xdx}{e^x-1}\right) \quad . \tag{D.7}$$

If we now note $2bj >> 1$, and $b << 1$, we can make the approximations

$$\int_{2bj}^\infty \frac{xdx}{e^x-1} \cong \int_{2bj}^\infty xe^{-x}dx \quad , \tag{D.8}$$

and

$$\int_0^{2b} \frac{x\,dx}{e^x-1} \cong \int_0^{2b} dx = 2b \quad . \tag{D.9}$$

These approximations, when used with the above formulae, yield both upper and lower bounds for $\sum_{\ell=1}^{j} \{\ell[\exp(2b\ell)-1]^{-1}\}$. The two approximations for this summation are averaged geometrically. By using this geometric mean for the approximation of the summation we find

$$T_j \cong 1 + \frac{1}{4}\frac{\alpha}{b}\left\{[\frac{\pi^2}{6} - (1 + 2bj)e^{-2bj} - 2bj][\frac{\pi^2}{6} - (1 + 2bj)e^{-2bj}]\right\}^{1/2}. \tag{D.10}$$

Since $\alpha << 1$ we find

$$T_j^{-1} \cong 1 - \frac{\alpha}{4b}\left\{[\frac{\pi^2}{6} - (1 + 2bj)e^{-2bj} - 2bj][\frac{\pi^2}{6} - (1 + 2bj)e^{-2bj}]\right\}^{1/2} . \tag{D.11}$$

For the high activity case we expect $j >> 1$. Hence

$$S_jT_j^{-1} \cong (1-e^{-2bj})(\frac{e}{2bj})(\frac{1}{2})\exp(\frac{-\pi^2}{12bj})(1 - \frac{\alpha\pi^2}{24b}) \quad . \tag{D.12}$$

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CHARGE DISTRIBUTIONS FOR RADIOACTIVE AEROSOLS IN A BIPOLAR ATMOSPHERE PERMEATED BY AN ELECTRIC FIELD

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Steady-state charge distributions are obtained for radioactive aerosol particles in bipolar ionized atmospheres with concentrations of 10^2 to 10^8 ions/cm^3. Ion-particle attachment is based on a diffusion-conduction model with unequal ion mobilities. Asymptotic formulae for charge distributions are derived from charge transfer rate equations. Cases include low and high specific activities with weak and strong electric fields. Mean charge and variance are obtained for Poisson and shifted Gaussian approximations in the appropriate cases. Comparisons are made with numerically calculated distributions.