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Atmospheric Environment 35 (2001) 5817–5821

ATMOSPHERIC ENVIRONMENT

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Scavenging of electrified radioactive aerosol

S.N. Tripathi, R.G. Harrison*

Department of Meteorology, The University of Reading, P.O. Box 243, Earley Gate, Reading RG6 6BB, UK

Received 1 November 2000; received in revised form 17 May 2001; accepted 29 May 2001

Abstract

Scavenging by water droplets is a mechanism for aerosol removal near clouds. Numerical methods are developed to quantify the removal of charged radioactive aerosols, including the electrical image force's contribution, attractive at small separations. Charging of radioactive aerosols is found to have significant effects on their collision efficiency and scavenging coefficient. The effect depends on the aerosol charge, and therefore, on the radioactive aerosol's decay rate and number concentration, but it does not depend significantly on the charge carried by the water drops. Scavenging coefficients are calculated for radioactive aerosols. For small particles at low aerosol concentrations ($Z \sim 10-100\,\mathrm{cm}^{-3}$), charging can increase the scavenging coefficients by up to an order of magnitude. Electrification will, therefore, encourage the removal of small radioactive aerosols from the atmosphere, more rapidly than equivalent non-radioactive aerosols. The increase in removal at low radioactive-aerosol concentration may account for under-predictions of surface concentrations and will contribute to spatial variations in aerosol removal.

Keywords: Collision efficiency; Aerosol charging; Wet removal; Radioactivity

1. Introduction

Radioactive aerosols differ from non-radioactive aerosols by their emission of charged α and β particles, but otherwise, their physical behaviour is usually assumed to be similar to non-radioactive aerosols. Each radioactive decay changes the charge of the emitting aerosol particle. It has previously been assumed (Greenfield, 1957) that radioactivity will lead to electrical neutralisation of radioactive aerosols and inhibition of the associated electric field growth (Spangler and Rosenkilde, 1979). Although at high activities this is likely to be the case, at lower activities, it has been shown (Clement and Harrison, 1992) that radioactive aerosols can acquire large charges, and recent experimental data supports the theory (Gensdarmes et al., 2000).

Scavenging of aerosols by water droplets is affected by the electrical forces between aerosol particles and droplets, principally for submicron aerosol (Pruppacher and Klett, 1997). In an intercomparison of wet precipitation scavenging schemes for the simulation of global transport and deposition of ^{210}Pb, Lee and Feichter (1995) observed that their model underpredicted the surface concentrations of ^{222}Rn. ApSimon et al. (1989) compared model results and observation data following the Chernobyl release and transport of radioactive debris, and found measured surface concentrations to be greater than those predicted from theory. Tinsley et al. (2000) have shown that the electrical image force is very significant in aerosol–droplet collisions as, unlike the Coulomb force, it is always attractive between the charged aerosol and water droplet at small separations. Numerical scavenging schemes may, therefore, be incomplete because they do not account for the effect of radioactive electrification on aerosol–droplet collisions.

2. Water droplet scavenging

For aerosol particles of radii $a > 0.4\,\mu\mathrm{m}$, the efficiency $E(A, a)$ with which a drop of radius A collides with an

*Corresponding author. Fax: +44-118-931-8905.
E-mail address: r.g.harrison@reading.ac.uk (R.G. Harrison).

1352-2310/01/$ - see front matter
PII: S1352-2310(01)00299-0

aerosol particle may be determined by integrating the equation of motion of an aerosol particle under the simultaneous influence of gravity, air drag, and electrical forces. The collision efficiency E is defined as

$$E(A,a) = \frac{\pi y_c^2}{\pi (A+a)^2}, \tag{1}$$

where A is the drop radius, a particle radius and y_c the largest horizontal offset which a particle can have from the drop axis and yet still collide with the drop (Pruppacher and Klett, 1997). The equation of motion of the aerosol is

$$m\frac{dV}{dt} = mg^* - \frac{6\pi\eta A(V-U)}{1+\alpha N_{Kn}} + F_e, \tag{2}$$

where m, V, a are the mass, velocity, and radius of the aerosol particle, respectively (Grover and Beard, 1975; McGann and Jennings, 1991) and $(1+\alpha N_{Kn})$ is the Stokes–Cunningham slip correction factor. $g^* = g(\rho_p - \rho_a)/\rho_p$, where g is the acceleration due to gravity, ρ_p the density of the aerosol particle, ρ_a the density of the air, U the velocity of the air around collector drop and F_e the electric force between water drop and aerosol. The Knudsen number N_{Kn} is given by λ_a/a, where λ_a is the mean free path of air molecules. The parameter α is given by Junge (1963) as

$$\alpha = 1.26 + 0.44\exp(1.07/N_{Kn}). \tag{3}$$

The particle's trajectory can be obtained by numerically integrating Eq. (2) in the reference frame of the drop (Fig. 1), and many trajectories are illustrated in Tinsley et al. (2000).

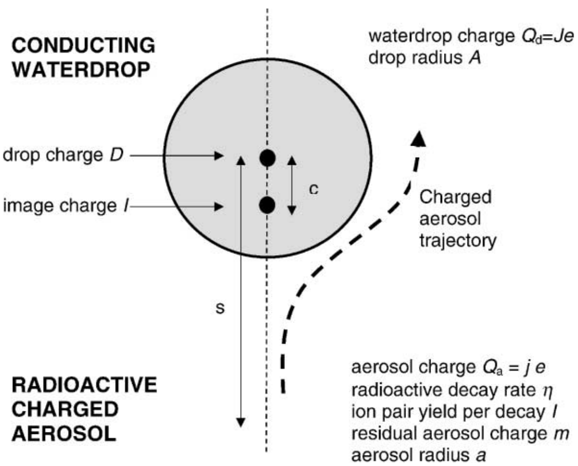

Fig. 1. Schematic diagram of the radioactively charged aerosol and the image charge I construction within a waterdrop of radius A. The aerosol and falling drop carry charges Q_a and Q_d, respectively, with $Q_d = I + D$, where D is the non-image charge considered at the centre of the drop.

2.1. Electrical forces

Fig. 1 also shows the partitioning of the water drop charge Q_d in response to the image charge induced by the charged aerosol particle (carrying a charge Q_a) brought close to the drop. Charge conservation requires that $Q_d = I + D$. In magnitude, $I = -(A/s)Q_a$, where s is the separation distance between the aerosol and drop centres. The image charge is located at a distance c from the centre of the drop (Jackson, 1975). Summing the Coulomb and image forces, the net electrical force acting between the particles' centres is

$$F_e = \frac{1}{4\pi\varepsilon_0}\left[\frac{Q_a I}{(s-c)^2} + \frac{Q_a D}{s^2}\right], \tag{4}$$

where a positive F_e is repulsive.

2.2. Radioactive aerosol electrification

Clement and Harrison (1992) showed that charging of radioactive aerosol could be characterised by three parameters. Radioactive decay particles were considered to leave an aerosol particle at a rate η. Each decay produces I ion-pairs in the local gas, with m self-generated electronic charge units remaining on the aerosol after each decay. The aerosol charge arises from the competition between the externally produced ions diffusing back to the aerosol and the self-generated charge associated with each decay.

The mean number of electronic charges j on a set of radioactive aerosol particles was found by Clement and Harrison (1992) using the iteration

$$j_{i+1} = my + j_i(x-1)\left[\exp(2\lambda j_i) - 1\right], \tag{5}$$

where $x = (\mu_+ n_+/\mu_- n_-)$ and $y = \eta\varepsilon_0/e\mu_- n_-$. Here $\mu_\pm$ is the positive (or negative) ion mobility, and $n_\pm$ the ion number concentration per unit volume. λ is the ratio of electrical energies to thermal energies of the ions, ε_0 the permittivity of free space and j_i an assumed initial number of charges, estimated using non-radioactive aerosol-charging theory. After convergence, the aerosol charge is found as $Q_a = je$. The radioactive charging parameters used by Clement and Harrison (1992) were chosen to be typical of a range of possible radioactive aerosols, and similar values are employed in this study.

2.3. Collision efficiency

The collision efficiency was computed from an analysis of the trajectories of the aerosol particles moving past the water drop. The assumption that the flow around the aerosol particle does not affect the drop motion is justified for ratios of particle mass to collector drop mass $<10^{-3}$. The flow fields outside a non-circulating water drop were computed by solving the complete Navier–Stokes equation of motion for steady, incompressible flow past a rigid sphere, using a method originally developed by LeClair et al. (1970). For a velocity field in spherical coordinates centred on the drop (Tripathi, 2000), the value of largest horizontal offset between droplet and aerosol, y_c, was found by choosing an initial offset value ($2.5A$), and integrating the resulting trajectory in time. For subsequent trajectories, the offset was determined by bisecting the previous offset value until a grazing collision occurs, when $(y_{i+1} - y_i)/y_i < \varepsilon_c$, where ε_c is the *collision tolerance*. For such a grazing collision, $y_c = y_{i+1}$. The collision tolerance was chosen as $\varepsilon_c = 10^{-4}$ (e.g. Miller and Wang, 1989), although the precise value does not materially affect the results. The initial aerosol–droplet separation was $20A$.

3. Scavenging coefficients

For an aerosol size distribution with N particles per unit volume of radius a, $N(a)$, the number of collisions between aerosol particles with radii in the range (a, $a +$ da) and a water drop of radius A is

$$\frac{\pi}{4}(A+a)^2\,[U(A) - V(a)]\,E(A,a)\,N(a)\,\mathrm{d}a, \tag{6}$$

where $U(A)$ and $V(a)$ are the terminal velocities of drop and particles, respectively (Seinfeld and Pandis, 1998). The below-cloud scavenging (washout) rate of aerosol particles of radius a can be written as

$$\frac{\mathrm{d}N_\mathrm{M}}{\mathrm{d}t} = -\Lambda(a)\,N_\mathrm{M}(a), \tag{7}$$

where the scavenging coefficient $\Lambda(a)$ is found from the collision efficiency using

$$\Lambda(a) = \int_0^\infty \frac{\pi}{4} A^2\,U(A)\,E(A,a)\,N(A)\,\mathrm{d}A. \tag{8}$$

Jylha (1991) determined scavenging coefficients from radar observations, finding an average value of $\Lambda = 1.02\,\mathrm{h}^{-1}$ for a rainfall rate of $5\,\mathrm{mm\,h}^{-1}$. Earlier work by Makhonko (1967) suggested values of the rainfall rate between 0.02 and $0.14\,\mathrm{h}^{-1}$ for the scavenging coefficients of fission products.

4. Calculated scavenging coefficients for charged radioactive aerosols

4.1. Monodisperse radioactive aerosol

Calculation of the aerosol scavenging rate, for a given monodisperse aerosol of radius a, therefore, requires a droplet size distribution $N(A)$ and the collision efficiency $E(A, a)$. The droplet size distribution, used here, was chosen to be fairly typical of stratocumulus, using flight A209 of the ASTEX atmospheric experiment (12 June 1992) (Hogan, 1998). Fig. 2 shows scavenging coefficients Λ for monodisperse radioactive aerosols found using Eq. (8), for drops of radii 40–100 μm in the size distribution, each carrying charges of $500e$ (e.g.

(a)

(b)

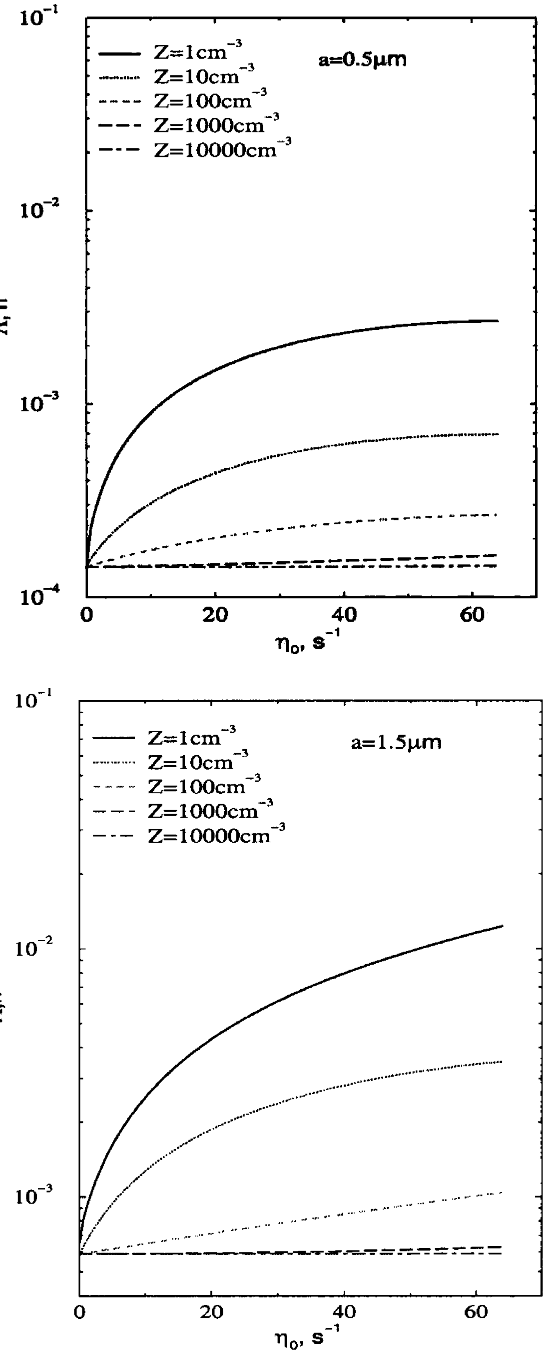

Fig. 2. Scavenging coefficient Λ plotted as a function of particle decay rate η_0 for different aerosol number concentrations Z. The monodisperse radioactive aerosol has radius: (a) 0.5 μm and (b) 1.5 μm.

Pruppacher and Klett, 1997; Chalmers, 1967), although sensitivity to the drop charge is small. There is an order of magnitude increase in Λ for 0.5 μm radius aerosol for decay rates increasing from 0 to $64\,s^{-1}$, when the aerosol concentration is very low. The rate of increase in scavenging coefficient is relatively large for small changes in the decay rate when the decay rate is less than $20\,s^{-1}$. Asymptotic behaviour of Λ is observed for large values of decay rate, as any further increase in decay rate does not change the aerosol charge. It is also seen that for aerosols with $Z = 10{,}000\,cm^{-3}$, no appreciable change in Λ is found, even for large decay rates, as the aerosol particles do not obtain large charges. Insignificant changes in Λ were also found even for large decay rates with aerosol radii bigger than 3 μm, because inertial forces dominate over the electrical forces.

4.2. Polydisperse radioactive aerosol

Clement et al. (1995) extended the charging theory to include polydisperse radioactive aerosols. One effect of a range of particle sizes on electrification is the competition for ions between the larger and smaller aerosols.

In Fig. 3, scavenging coefficients are plotted for a lognormal polydisperse aerosol (mean radius $a_m = 0.26\,\mu m$, geometric deviation $\sigma_g = 2.4$) collected by drops of radii 70 and 102 μm, respectively. From Fig. 3(a), for $\eta_0 = 0.47\,s^{-1}$, the increase in scavenging coefficient is insignificant compared to neutral aerosols as small as 1 μm. In the higher decay rate case, Fig. 3(b), there is almost a five-fold increase in scavenging coefficient for aerosols of radii 3 μm, compared with neutral aerosols. This result is different from that obtained for monodisperse aerosols, where, the increase in scavenging coefficient was more pronounced only for small aerosols with $\eta_0 = 64\,s^{-1}$. This may be because the larger aerosols tend to attain large charges at the expense of smaller aerosols. As number concentration Z increases, the change in scavenging coefficient decreases, a result similar to the monodisperse aerosol case.

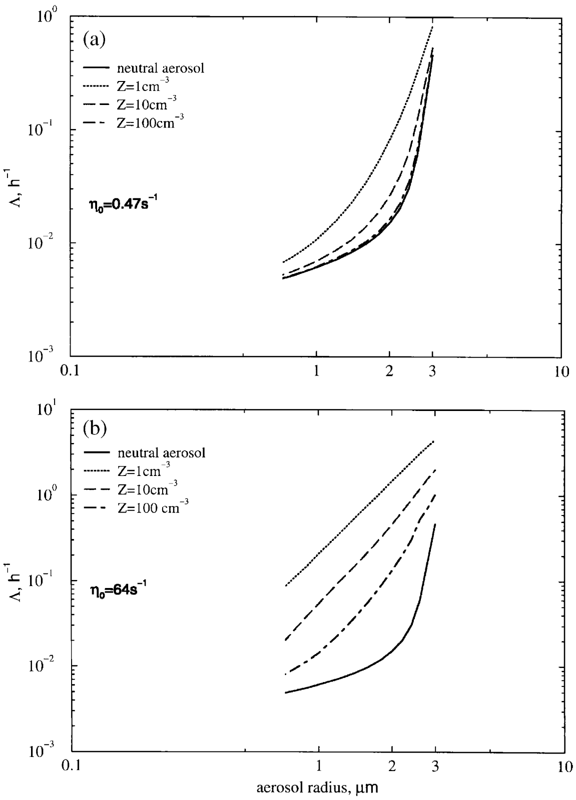

Fig. 3. Scavenging coefficient Λ plotted as a function of particle radius in a polydisperse aerosol population, when collected by a drop of radius 70 μm in: (a) low activity case with decay rate $\eta_0 = 0.47\,s^{-1}$ and (b) high activity case with decay rate $\eta_0 = 64\,s^{-1}$.

5. Conclusions

Inconsistencies between modelled and observed data on radioactive-aerosol removal appear to be partly explained by the electrical forces between the drop and aerosol, as the electrical image force is always attractive at small separations, no matter what charge the drop carries. The collision efficiency is relatively insensitive to the sign and magnitude of drop charge. Scavenging coefficients calculated for micron-sized monodisperse radioactive aerosols show that, for low aerosol concentrations, charging can increase the scavenging coefficients by an order of magnitude. Scavenging coefficients calculated for polydisperse radioactive aerosols, show an increase for the larger particles due to the large charges acquired by them at the expense of smaller particles. These variations are within the range of scavenging coefficients observed for radioactive particles. Consequently, it is necessary to include electrical effects in detailed calculations of atmospheric radioactive-aerosol removal, as spatial variability in natural cloud, aerosol and ion conditions will cause regions where electrically enhanced scavenging is especially significant for small radioactive-aerosol removal.

Acknowledgements

One of the authors (SNT) acknowledges financial support from The Felix Foundation. RGH acknowledges many helpful conversations with Prof. Brian Tinsley at Dallas.

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