ABSTRACT
Radioactive aerosols differ physically from non-radioactive aerosols by the appreciable charge that they may carry. This arises from a competition between the charge generated by the radioactive decays occurring within each aerosol and the subsequent diffusion of the ions formed back onto the particle. At high levels of activity, the particles may self-discharge, at low and intermediate activity levels, however, appreciable charging may occur. In the atmosphere, charged particles are removed preferentially over neutral particles by water droplets. At the surface, deposition rates of charged particles are greater than those of neutral particles, depending on their radii.

INTRODUCTION
Natural radioactive aerosols are ubiquitous in the atmospheric surface layer and artificial radioactive aerosols may also be released by human activities. A key aspect in which radioactive aerosols differ from non-radioactive aerosols is that, in many situations, they may acquire appreciable electric charge. This modifies their physical behaviour. Theoretical estimates of radioactive aerosol charging have recently been extensively experimentally verified [Gensdarmes et al., 2001]. Using the theoretical charge estimates, model results show that radioactive aerosols are preferentially removed by water droplet scavenging in clouds [Tripathi and Harrison, 2001]. When a radioactive aerosol release occurs, the resulting environmental effects include a combination of atmospheric droplet scavenging, electrical changes in aerosol coagulation and enhanced surface deposition. The effects on surface deposition are considered further here. The theoretical problem for small aerosol, however, is complicated further by the presence of the background electric field, which may be modified by the radioactive aerosol [Clement and Harrison, 2000; Mayya et al., 2002]. The general radioactive deposition problem requires coupled consideration of particle mechanics, atmospheric electrostatics and surface layer turbulence. Using a simplified model, the effects of radioactive charging on particle deposition rates are calculated, for different aerosol sizes and radioactive decay rates. An important finding is that in only some cases is the aerosol discharged by its radioactivity: in many other cases the deposition is enhanced. This also has implications for radioactive aerosol deposition in the human lung. In this paper we consider the effect of different aerosol charge distributions on the resulting deposition behaviour, extending previous results to include radioactively-charged aerosol.

AEROSOL DEPOSITION THEORY
Aerosols which are radioactive acquire charge, in general, more readily than a comparable non-radioactive aerosol because additional external ions are created by the radioactive decay particles. McMurry and Rader (1985) gave an aerosol deposition theory which included the
effect of an aerosol charge distribution, such as from radioactive aerosols. This theory gives
better agreement with the experiments than the equivalent neutral deposition theory, even using a
highly simplified charge distribution. McMurry and Rader (1985) followed Crum and
Seinfeld (1981) but extended the fractional deposition coefficient to \( \chi(r,j) \), including both the
aerosol radius \( r \) and charge \( j \) as

\[
\chi(r,j) = \frac{3\sqrt{k_0 D}}{\pi R X} \left\{ \frac{(X+Y)^2}{2} + (X+Y) \text{DI}(X+Y) + (X-Y) \text{DI}(X-Y) \right\}
\]

where \( \text{DI}(z) \) is the Debye function. \( X \) and \( Y \) are dimensionless parameters given by
\( X = \pi v_s / 2(k_0 D)^{1/2} \) and \( Y = \pi v_e / 2(k_0 D)^{1/2} \), where \( v_s \) is sedimentation velocity, \( v_e \) the electrical
migration velocity, \( D \) the Brownian diffusion coefficient and \( k_0 \) the coefficient of eddy diffusion.

Two important assumptions were made in deriving this formula. Firstly, electrostatic forces were
assumed to be the dominant forces acting on particles. Secondly, symmetry in deposition was
assumed for oppositely charged particles. This assumption will break down under strong ion
asymmetry where electric and dynamic effects will be coupled. This second assumption is
generally valid for non-radioactive aerosols as charges carried by them are not large. The
associated change in electric field can be shown to be small by simple calculations using
Gaussian law, from which the change in electric field is given by

\[
dE = \frac{d\sigma}{dt} = \frac{N_j v_e je}{\varepsilon_0}
\]

where \( \sigma \) is the surface charge density, \( N_j \) the number of aerosols carrying \( j \) charges and \( e \) the
elementary charge. In the case of highly-charged radioactive aerosols this assumption will break
down. From equation (2), it can be shown that for aerosols of 0.5 \( \mu \)m radius, the change in
electric field will be \( \sim 100\% \) for particles carrying elementary charges greater than 400. This
situation could arise for low particle concentrations with modest decay rates in indoor
environments, but a full treatment is not considered further here.

THEORY FOR CHARGE DISTRIBUTIONS
Aerosols charge as a result of collisions with ions, or, in the case of radioactive aerosols, from
the competition between self-generated charges from radioactive decays and the diffusion of ions
generated back onto the aerosol particle. Theories for charge distributions on radioactive and
non-radioactive aerosols are discussed in Clement and Harrison (1992), in terms of ion
asymmetry. Radioactive aerosols directly emit alpha or beta radiation and each radioactive decay
influences the charge of the active aerosol particle. In the theory of Clement and Harrison
(1992), charging of radioactive aerosol was characterised by three parameters. Radioactive decay
particles are considered to leave an aerosol particle at a rate \( \eta \), and then each produces \( I \) ion-pairs
in the local gas with \( m \) electronic charge units left on the aerosol after each decay. The aerosol
mean charge \( j \) can be found using the iteration

\[
J = m x + j \times (x - 1)[\exp(-2\lambda j) - 1]
\]

where \( x = (\mu n_+ / \mu n_-) \), \( y = \eta \varepsilon_0 / e \mu n_- \) and \( j_1 \) is mean charge given initially by from non-
radioactive charging theory, when \( j \) is small.
RESULTS AND DISCUSSION
Deposition calculations were performed to investigate the effect of different charge distributions on deposition, for the enclosed container described by equation (1). Surface deposition coefficients are plotted for aerosols charged by radioactive decays using mean charges calculated by the iterative approach given in equation (3). The values of radioactive decay rate $\eta$, ion production constant $I$ and self-charging coefficient $m$ are taken from Clement and Harrison (1992) as typical values for a radioactive aerosol.

Figure 1(a) Surface deposition rates $\chi$ of radioactive aerosols as a function of radius for a low concentration (number concentration $Z = 100\,\text{cm}^{-3}$), slightly radioactive aerosol (decay rate $0.077\,\text{s}^{-1}$), under different assumptions of asymmetry in ion mobility ($\mu_+ / \mu_-$). The lower solid line represents the neutral deposition rate calculation from McMurry and Rader (1985).

Figure 1(b) Surface deposition rates $\chi$ of radioactive aerosols as a function of radius for a highly radioactive aerosol, (decay rate $64\,\text{s}^{-1}$), for different aerosol number concentrations $Z$. The lower solid line represents the neutral deposition rate calculation from McMurry and Rader (1985).

In the case of radioactive aerosols, differences up to an order of magnitude are found between this work and the deposition coefficients expected from McMurray and Rader (1985) particularly at the lower end of size spectrum. Deposition rates could be up to a few ten percent greater in
the case of beta-emitting aerosols than alpha-emitting aerosols because of more effective charging. Table 1 summarises the removal times which are correspondingly greatly reduced for both alpha and beta-emitting aerosols. For accurate deposition calculations the shape of the charge distribution must also be included, rather than just the mean charges. The ion asymmetry factor is the most important parameter in the case of non-radioactive aerosol, but decay rate and number concentrations are more important for radioactive aerosols. Deposition increases in general when at least one of the following occurs: (1) ion concentration asymmetry exists, (2) low aerosol concentrations (1-10 cm$^{-3}$) exist, (3) particle decay rates are greater than 1.0 s$^{-1}$. Consequently for most radioactive contamination situations, aerosol deposition calculations requires the charging of radioactive aerosols to be taken into account.

**Table 1** Changes in the half removal timescales of aerosols of radius $a$ with respect to neutral aerosols for different values of ion asymmetry factor $x$ and number concentration $Z$.

<table>
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<th>Type of aerosol</th>
<th>change in removal time scale $\tau$, hour</th>
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<tr>
<td></td>
<td>$0.05 \leq r \leq 0.1 \mu m$</td>
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<tr>
<td>Alpha-active aerosol</td>
<td>Z cm$^{-3}$</td>
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<tr>
<td></td>
<td>1</td>
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<td></td>
<td>10</td>
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<tr>
<td>Beta-active aerosol</td>
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<tr>
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REFERENCES