Studying the impact of radioactive charging on the microphysical evolution and transport of radioactive aerosols with the TOMAS-RC v1 framework

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1. Introduction

Human activities and events can release large amounts of radioactive particles into the atmosphere; nuclear reactor meltdowns such as the Fukushima and Chernobyl accidents, weapon tests, radioactive waste treatment, as well as coal fired power plants constitute important sources of airborne radionuclides (Chesser et al., 2004; Lujaniene et al., 2007; Yoshiko and Kanda, 2012; McBride et al., 1978; Mulpuru et al., 1992). The impacts of this radioactivity on the environment and human health depends largely on where it deposits, and underscores the need for its accurate prediction for policy and first response efforts to accidents and other release events.

Aerosols carrying radionuclides can spontaneously accumulate electrostatic charge, which affects their microphysical evolution in the atmosphere. Because of this, radioactive aerosol can exhibit a distinctly different behavior when compared to non-radioactive (and neutral) aerosol (Simons, 1981; Clement et al., 1995; Clement and Harrison, 1992; Harrison and Carslaw, 2003; Kim et al., 2014, 2015, 2016). Considering charging effects of radioactive aerosols immediately impacts their initial deposition patterns, and following the resistances in series model described in Seinfeld et al. (2006), it is expected that areas affected by radioactivity would present different surface resistances to charged aerosol populations. Therefore, the deposition rate for particles of a given size can be enhanced/reduced close to a radioactive surface. Secondary resuspension and redistribution mechanisms from wild fires, dust events and water runoff downwind of nuclear accidents (Lujaniene et al., 2007; Adeyini and Oladiran, 2006; Yoshenko et al., 2006a,b; Masson et al., 2011), all contribute to the long-term impacts of release events and are affected by where the primary deposition of radionuclides occur. For example, extensive forest fires between May and...
August of 1992 in the 30-km ring surrounding the Chernobyl-exclusion zone are known to have emitted radioactive aerosols to the atmosphere, detectable at considerable distances (Lujaniene et al., 2007; Yoshida and Kanda, 2012; Ooe et al., 1988). As a result, important secondary contamination was found in areas that were initially not contaminated by radioactivity. The remobilized radionuclides deposited after wild fires were more concentrated and water-soluble, and therefore easily redistributable by water runoff into subsurface water systems (Lujaniene et al., 2007; Yoshenko et al., 2006a,b).

Charging of radioactive aerosols occurs from the decay of radionuclides, which can be in either the form of α or β emission (Clement and Harrison, 1992). For alpha emission, an alpha particle of a charge of +2 is ejected from the aerosol, leaving a residual charge of −2. Nevertheless, in general, radioactive aerosols undergoing alpha decay become positively charged because alpha particles can cause significant ionization within the aerosols, which leads to the emission of secondary electrons. In β-decay, the emission of energetic electrons results in a residual charge of +1. These ions ejected and transferred their kinetic energy to the gas phase can combine with surrounding gas molecules or other aerosols through collision/adsorption, thereby producing many ion pairs and charged aerosols. Renard et al. (2013) found that many large aerosols in the upper troposphere and stratosphere can be easily charged by diffusion of ions, and the aerosols may gain more charges as their size increases. Negative ions produced from ionizing radiation tend to exhibit a higher mobility than positive ions produced (Gunn, 1954; Mohnen, 1976); this asymmetry in mobility leads to the charging of aerosols through the diffusion of ions onto their surface, with a surplus of negative gas-phase ions over positively charged ions. Ion mobilities are dependent upon the molecular weight, temperature and pressure of the surrounding gas (Harrison and Carslaw, 2003; Mohnen, 1976; Clement and Harrison, 1992). Therefore, variability of these factors can lead to a range of mobilities, and charging (Clement and Harrison, 1992). The direct ionization of particles from radioactive decay is called "self-charging", while collision/adsorption of gas-phase ions generated by the radioactive decay is called "diffusion charging" (Harrison and Carslaw, 2003; Clement et al., 1995; Clement and Harrison, 1992; Kim et al., 2014, 2015, 2016).

Because radioactive aerosols can be easily discharged by ionizing radiation (Greenfield, 1956), as well as neutralized by ions produced in the containment atmosphere (Clement et al., 1995; Clement and Harrison, 1992), the effects of electrostatic interactions on microphysical processes of aerosols have been frequently neglected (e.g., Greenfield, 1957). However, many radioactive aerosols can be appreciably charged in open air (Kim et al., 2015), suggesting that several microphysical processes of the aerosols can be affected by radioactive charging causing electrostatic interactions (e.g., coagulation (Clement et al., 1995; Kim et al., 2014, 2016)) and impacting the rate of wet scavenging (Tripathi and Harrison, 2001; Sow and Lemaître, 2016). Chemical transport models (CTMs) with explicit aerosol microphysics are well-poised to consider all the relevant processes that control the transport and deposition of radioactive aerosol (e.g., condensation/evaporation of semi-volatile species, coagulation of particles, cloud processing, wet/dry deposition and horizontal/vertical transport). State-of-the-art atmospheric models, however, do not account for radioactivity impacts on aerosol microphysics (e.g., Yoshenko et al., 2006a,b; Christoudias et al., 2013). This omission introduces an unknown, and potentially important, bias in the predicted deposition patterns following a radioactive release event.

This study is an initial step to develop a comprehensive atmospheric modeling approach to account for the effects of radioactive charging on the microphysical evolution, transport and deposition of radioactive aerosol from the atmosphere. The established TwO-Moment Aerosol Sectional (TOMAS) aerosol microphysical model (Adams and Seinfeld, 2002) is used to simulate the microphysical evolution of atmospheric particulate matter, and is augmented to include electrostatic particle-particle interactions in the presence of radioactive charging. The expanded model, called TOMAS-RC (TOMAS with Radioactive Charging effects) is used to study, with idealized simulations, situations where charging exerts an important influence on the transport and deposition of radioactive particles.

2. Methods

2.1. TOMAS aerosol microphysical model

The version of the TOMAS model (Adams and Seinfeld, 2002) used here has a resolution of 30 bins, covering particle sizes from 10 nm to 10 μm. TOMAS accounts for all the relevant atmospheric processes of nucleation, condensation, coagulation, vertical mixing, cloud processing and deposition, in order to find the number concentration and size distribution of the modeled aerosol. For the needs of this study only dry deposition, vertical mixing and coagulation were active. TOMAS tracks two independent moments, number and mass, of the aerosol size distribution for each size bin:

$$N_k = \int_{x_k}^{x_{k+1}} n_k(x)dx$$

$$M_k = \int_{x_k}^{x_{k+1}} x n_k(x)dx$$

where $N_k$, $M_k$ are the total number and mass of aerosol in the $k$-sized bin, $n_k(x)$ is the number of particles with masses included between $x + dx$, and $x_k$ is the lowest boundary of the $k^{th}$ bin. The lowest and the largest boundaries of each cell are defined in terms of dry aerosol mass, in such a way that the lowest boundary has double the mass of the lowest boundary, something that allows for considerable gains in computational efficiency (Adams and Seinfeld, 2002). A detailed description of TOMAS is available elsewhere (Adams and Seinfeld, 2002; Lee and Adams, 2012).

The aerosol size distributions are influenced by microphysical processes (e.g., deposition, vertical layer mixing, and coagulation) occurring in each computational cell. The rate of change for $N_k$ and $M_k$ resulting from coagulation, which is a process modulated by radioactive charging, are given by (Fuchs, 1964; Tzivion et al., 1987, 1988; Adams and Seinfeld, 2002):

$$\frac{dN_k}{dt} = 0.5K_{k+1-k}N_{k+1} - K_{k-1}N_k - N_k\sum_{i=1}^{k-1} K_{i}I_{hi}N_i$$

$$+ \psi_{k-1}I_{hi} - \sum_{i=1}^{k-1} K_{i}I_{hi}M_i - \psi_{k-1}I_{hi} - \sum_{i=1}^{k-1} K_{i}I_{hi}M_i - K_{k-1}M_k$$

(3)

$$\frac{dM_k}{dt} = K_{k+1-k}M_{k+1} - K_{k-1}M_k - N_k\sum_{i=1}^{k-1} K_{i}I_{hi}M_i$$

$$- M_k\sum_{i=1}^{k-1} K_{i}I_{hi} - \psi_{k-1}I_{hi} - \sum_{i=1}^{k-1} K_{i}I_{hi}M_i - \sum_{i=1}^{k-1} K_{i}I_{hi}M_i + \frac{1}{2}\psi_{k-1}I_{hi}$$

$$- \frac{1}{2}\sum_{i=1}^{k-1} K_{i}I_{hi}M_i^2$$

(4)

where $I$ and $k$ represent the size bins between which coagulation takes place, $K_{k,i}$ is the coagulation coefficient between these two bins, $I$ is the total number of size bins which is equal to 30 for this case, $\psi$ and $f$ are weighting factors described in Tzivion et al. (1987), $\xi$ is the closure parameter determined size range of each bin (here, $\xi = 1.0625$), $x$ is the lowest dry mass boundary of the cell, $m_i$ is the average particle mass in bin $i$, and $t$ is the time.

In Eqs. (3) and (4), the coagulation coefficient, $K$, is the parameter controlling the coagulation rate of aerosols, and it can be affected by...
several collision mechanisms involving interparticle forces and flow regimes (Seinfeld et al., 2006). In TOMAS, it is assumed that Brownian motion is the dominant collision mechanism (Adams and Seinfeld, 2002; Lee and Adams, 2012), and the coagulation coefficient is obtained using the interpolation formula of Fuchs (1964) to consider aerosol coagulation in the continuum, transition, and free molecular regimes. In TOMAS-RC, the Brownian coagulation coefficient is augmented to include the effects of radioactive charging, following the suggestions of Clement et al. (1995); Kim et al. (2016), and as described below.

2.2. Coagulation of radioactive aerosols

Charging effects on the coagulation rate between particles $i$ and $k$ are introduced through a "correction factor" multiplier, $W_{k,i}$, applied to the Brownian coagulation kernel at each aerosol microphysical step (Fig. 1). $W_{k,i}$ in TOMAS-RC is based on the stability function correction factor formulation neglecting the effects of image forces (Fuchs, 1964; Spielman, 1970; Seinfeld et al., 2006):

$$W_{k,i} = \frac{\gamma}{\varepsilon^2 - 1}$$  \hspace{1cm} (5)

where $\gamma = \frac{k_b T}{2e\varepsilon_0}$, $k_b$ is the Boltzmann constant, $T$ is the temperature. $W_{k,i}$ is applied as an enhancement factor to the Brownian kernel; as the charge of either the colliding particles approaches zero (i.e., $j_{k,i} \rightarrow 0$), $\gamma \rightarrow 0$ and $W_{k,i} \rightarrow 1$ (because $e' \sim 1 + \gamma$ for small $\gamma$), and thus, coagulation rates of the particles become similar to those expected from Brownian diffusion of neutral particles. This limit is relevant for the coagulation between small particles that typically carry an average charge of less than 1 ($\gamma < 0.1$), and subsequently their coagulation is not impacted by charging effects. In the case of particles with opposite sign charges, $\gamma > 0$ and $W_{k,i} > 1$ meaning that coagulation is enhanced; for particles with like charges, $\gamma > 0$ and $W_{k,i} < 1$ leading to inhibition of coagulation.

Equation (5) is derived assuming coagulation of charged particles in the continuum regime. The correction factor formulations for the transition and molecular regimes are available elsewhere (e.g., Marlow (1980) and Huang et al. (1990)). Compared to these formulations, equation (5) is less accurate (e.g., up to 10% errors for the transition regime (Huang et al., 1990)). In contrast to these formulations requiring high computational costs, however, equation (5) is much simpler and computationally more efficient, indicating that the equation may be more suitable for use in three-dimensional transport models. Also, equation (5) has been used in various modeling and experimental investigations into coagulation of charged particles in the molecular and transition regimes [e.g., Maisels et al. (2002a, 2002b)] because the equation may still provide reliable computational results despite the possibility of errors. For instance, Maisels et al. (2002b) estimated the coagulation coefficient of charged particles in the transition regime using the interpolation formula of Fuchs (1964) and found that the calculation was in good agreement with the measurements. Thus, in this study, equation (5) was used to include the effects of particle charging on particle coagulation in all flow regimes.

Equation (5) depends strongly on the number of charges existing on the coagulating particles. An appropriate theory is therefore required to calculate at each coagulation timestep (Fig. 1) the number of charges that develop on the aerosol population. Kim et al. (2016) has evaluated an approach assuming a Gaussian distribution to approximate the charge distribution and found that the errors associated with such an assumption only become significant for particles with diameters smaller than 40 nm. An explicit representation of the charge distribution would be extremely computationally demanding when compared to a Gaussian, as demonstrated in Clement et al. (1995) and Kim et al. (2016). Furthermore, the average charge and deviation values used in the Gaussian distribution are approximated from the exact distributions (Clement and Harrison, 1992), which further reduce the error while achieving desirable computational efficiency. In the presence of self-charging and diffusion charging, the Gaussian distribution used to describe the charge distribution that develops for particles in each size bin $k$ (Clement et al., 1995; Gensdarmes et al., 2001; Kim et al., 2016) is:

$$N_{j,k} = \frac{1}{\sqrt{2\pi \sigma_j}} \exp \left( -\frac{(j - \mu_j)^2}{2\sigma_j^2} \right)$$  \hspace{1cm} (6)

where $N_{j,k}$ is the number concentration (m$^{-3}$) of aerosols in size bin $k$ carrying charge $j$. $N_k$ is the total number of particles in bin $k$ ($N_k = \sum N_{j,k}$) and $\mu_j, \sigma_j$ are the mean aerosol charge and standard deviation of the aerosol charge distribution for size bin $k$, given by:

Fig. 1. Flow diagram of the computational scheme in TOMAS-RC. The number concentration and diameter of each bin are passed from the coagulation subroutine to the aerosol charging subroutine, where the average charge and charge distributions are calculated, and then used to estimate the enhancement factor, which is returned to the TOMAS coagulation module. Processes in bold are the ones active in TOMAS-RC.
\[
\sigma^2 = y_k + 1 + \frac{1}{2a_k}, J_k = \begin{cases} 
\frac{y_k - \left(\frac{y_k(3X-1)}{exp(2a_k)X-1}\right)}{y_k + \frac{X-1}{2a_k}} & \text{if } y_k > 0.22 \\
\frac{y_k - \left(\frac{y_k(3X-1)}{exp(2a_k)X-1}\right)}{y_k + \frac{X-1}{2a_k}} & \text{if } y_k \leq 0.22 \end{cases} 
\]

where \(\omega_k = \frac{e^2}{8\pi\varepsilon_0a_k}\) is a parameter describing the effects of diffusion charging, \(X = \frac{\mu_+}{\mu_-}\) is the asymmetry/mobility ratio, \(\mu_+\) and \(\mu_-\) are the mobilities of positive and negative ions, respectively (m² V⁻¹s⁻¹), \(y_k\) is the positive charge accumulated via self-charging and \(n_k\) is the total number of ions in the air. The Gaussian charge distribution we utilize has been used in previous literature reports (Clement and Harrison, 1992; Clement et al., 1995; Kim et al., 2016) as an approximation of the exact charge distribution calculated numerically in Clement and Harrison (1992). The normal distribution presents a simple, yet accurate representation of the charge distribution of radioactive aerosols (e.g., Gensdarmes et al., 2001). For the internally mixed aerosol populations presented here, the Gaussian distribution constitutes an accurate simplification of the steady-state charge distribution as shown in Kim et al. (2016). The size-dependent radioactive decay per particle \(n_0\), in each size bin, required to obtain \(y_k\) is determined from the specific radioactive decay rate \(\eta_0\) for each species (Clement and Harrison, 1992):

\[
\eta_k = \eta_0 r_k^3 \tag{7}
\]

Calculation of \(y_k\) also requires the total number \(n_0\) of ions (positive and negative) produced from natural radioactivity, cosmic rays, and decay of radionuclides attached to aerosols and is given by (Clement et al., 1995; Harrison and Carslaw, 2003):

\[
n_0 = \frac{q_i + q_f}{\sigma_{cr}}, \tag{8}
\]

where \(q_i\) is the rate of ionization by radon and cosmic radiation, \(q_f\) is the rate of ionization caused by radioactive aerosols, and \(\sigma_{cr}\) is the rate coefficient of ion-ion recombination.

Equation (6) applies to background aerosols as well; in this case \(y_k \rightarrow 0\) so \(\sigma_k \rightarrow 0\). The resulting distribution has a Gaussian mean charge, \(\bar{X} = \frac{y_k}{n_0}\), that represents the effects of diffusion charging acquired by particles from background radiation, and has been validated in a number of studies (e.g., Gensdarmes et al., 2001; Kim et al., 2014; Kim et al., 2016).

The size and charge are important variables in Eqs. (5)-(7) and continuously vary over the whole size distribution. A singular charge distribution \(\bar{X} = y_k\) (Eq. (6)), reduces to the symmetrical Boltzmann distribution which occurs as the ion asymmetry ratio \(X\) approaches 1. The mean charge of each aerosol size bin can then be used to approximately calculate the correction factor. For lower values of \(X\) however, the resulting charge distributions are asymmetric and not well approximated by the Boltzmann distribution due to the higher mobility of the negative ions (Clement and Harrison, 1992), making the average charge an insufficient proxy for the correction factor. To overcome this limitation, the average correction factor between particles of size \(i\), \(\bar{W}_{i,j}\) proposed by Clement et al. (1995) and validated by Kim et al. (2016), which can consider the interaction of all charged aerosols, was employed.

\[
\bar{W}_{i,j} = 1 + \frac{\sum_j N_{i,j} N_{j,i} (W_{i,j} - 1)}{\sum_j N_{i,j} \sum_j N_{j,i}} \tag{9}
\]

If repulsive electrostatic forces are predominant among aerosols, the sign of the fractional term in the right-hand side of Eq. (9) is changed to minus and the average correction factor is less than unity (i.e. radioactive charging inhibits coagulation). Radioactive charging impacts in TOMAS-RC are introduced by multiplying the Brownian coagulation coefficient by \(\bar{W}_{i,j}\) (Fig. 1).

2.3. Optimization of coagulation corrections for broad charge distributions

During simulations, the width of the charge distribution for each bin, approximated by \(\bar{X} \pm 5\sigma\), is proposed Kim et al. (2016) to determine the summations in Eq. (9). The larger the average charge and the deviation of the distribution for that bin, the larger the summation index \(l = \bar{X} \pm 5\sigma\). Of Eq. (9) becomes, which, in the case of large particles carrying significant charges (Clement et al., 1995; Clement and Harrison, 1992; Kim et al., 2014) can lead to values of \(l\) of the order 10³.

The computational burden in such situations quickly becomes overwhelming, as the required calculations for the correction factor scale with \(k^2\) at each microphysical TOMAS-RC timestep (because the summations of Eq. (9) need to be recalculated every time to consider all interactions between all size bins \(k\), as well as every possible charge value for bin \(l\)). In the case of radioactive particles with a diameter greater than 2μm, the average charge attained can be, depending on the radionuclide of question, on the order of thousands, which increases the computation time by at least 10⁴ when compared to smaller particles with \(\bar{X}\) values less than 10. The computational burden is further increased by broadening of the charge distribution – which expands the summation in Eq. (9) to include substantially more terms.

To accelerate calculations while minimizing loss in accuracy, instead of iterating over all possible values of \(l\) for each size bin, iterations are done over a limited charge interval about the mean \([\bar{X} - 9\sigma, \bar{X} + 9\sigma]\), which for the normal charge distribution encompasses 95% of the possible charge values. Other values for the intervals were tested, spanning from \(\bar{X} \pm 5\sigma\) to \(\bar{X} \pm 6\sigma\) and the results showed the best agreement with the lowest associated computational cost for \([\bar{X} - 2\sigma, \bar{X} + 2\sigma]\). We additionally employ an adaptive, linearly increasing, step for the iterator \(j\) of Eq. (8), which was used when the average charge exceeded 100. This step was derived empirically based on simulation results for the cases where \(\bar{X}\) and \(\sigma\) were highest, by determining the limiting case from all the simulation scenarios, which occurred for \(^{131}\text{I}\) when the particle size and concentrations were maximum. We find that using a step of \(\Delta n = \frac{\text{bin}}{100}\) (where \(n\) is the step size for a given particle size \(\bar{X}\), \(\Delta n\) is the deviation of the charge distribution for size bin \(k\)) considerably accelerates the calculations at a minimal loss of computational accuracy.

Fig. 2 shows the comparison between the approach described in Clement et al. (1995) and the aforementioned scheme, showing the average enhancement factors between particles of a given size \(r_k\), as a function of a coagulating particle with size, \(r_s\). For particles with \(r_s\) values of less than 0.345μm, coagulation tends to be unaffected for smaller sizes and enhanced for larger ones, since particles of that size carry a very small negative charge of less than ~1. As small charge means that their coagulation with other small particles is not inhibited, while enhancement is seen for larger sized particles carrying a large positive charge (\(\bar{X} > 100\)). For particles with \(r_s\) values of 0.801μm (\(\bar{X} \approx 2\)), there is an initial enhancement between the negatively charged, small-sized part of the distribution, an inhibition for mid-sized, positively charged particles, and a subsequent enhancement for the larger, strongly positive particles. Since these particles carry the same sign charge, this enhancement can only be explained by a considerably negatively charged tail of the charge distribution (\(q_0 = 3.71\)) of the particles with \(r_s = 0.801\mu m\), which shows that the average charge is not a good predictor of radioactive coagulation, and the use of a distribution is necessary. For the larger particles with \(r_s = 1.6\mu m\) (\(\bar{X} \approx 21.5\)), there is an initial expected enhancement of coagulation between them and the negatively charged smaller particles, and a strong inhibition for the positively charged larger particles.

Using the optimizations described above, calculation of \(\bar{W}_{i,j}\) in TOMAS-RC is accelerated by up to 3 orders of magnitude. When compared to using the exact summation calculations over a charging distribution that considers ± 5\(\sigma\) about the mean \(\bar{X}\), there is no apparent loss in accuracy, as the enhancement factors computed for coagulation
of $^{137}$Cs aerosol (Fig. 2) are similar to the ones reported by Clement et al. (1995).

2.4. Atmospheric simulation scenarios

To demonstrate the capabilities of TOMAS-RC, it is used to simulate the deposition of $^{137}$Cs and $^{131}$I during an idealized radionuclide release incident. Simulation results for neutral background aerosol are obtained under the same initial conditions and given for reference. The specific radionuclides are considered, as they have been released during nuclear plant accidents (Masson et al., 2011; Yoshida and Kanda, 2012; Kauppinen et al., 1986). The characteristics of the idealized simulations are provided in Table 1. Values for radioactivity pertinent parameters were obtained from previous studies (Clement and Harrison, 1992; Clement et al., 1995; Kim et al., 2015). The height of the top layer was set to 1 km so that it could capture a potential plume from a nuclear accident (Chesser et al., 2004).

For all simulations considered, three aerosol microphysical processes were accounted for i.e., coagulation, vertical mixing (turbulent diffusion) and dry deposition (Fig. 1) – as a means of carrying out a semi-Lagrangian simulation, where an airmass is tracked as it advected away from its release point, but still allowed to vertically mix. Such a simulation resolves the processes that impact the microphysical evolution of the aerosols contained within the column and also can be used to compute the depositional loss of radioactive. The simulation duration is set to 5 days, which covers most of the lifetime of tropospheric aerosol (Seinfeld et al., 2006). We assume that $\eta_0$ of the radioactive aerosol is constant throughout the simulation. The effects of radioactive charging were assessed by examining the aerosol fields with and without radioactivity effects and for four different values of the ion mobility ratio ($\chi$): 0.7, 0.8, 0.9 and 1, for each radionuclide considered. In all simulations, it was assumed that each of the five vertical levels had an initial particle population that followed a log-normal distribution with a single mode. Dry deposition was the only active mechanism of aerosol loss from the atmospheric column.

The microphysical characteristics of the radioactive aerosol emissions are highly uncertain, so we assess the importance of radioactive charging for a wide range of initial distributions, by varying the geometric mean diameter, $D_m$, initial concentration of particles, $N_0$, and the geometric standard deviation $\sigma_g$ of the radioactive aerosol. The range of values used for this study is shown in Table 2. The results of the sensitivity analysis were evaluated using the column integrated mass ratio (CIMR) of aerosol remaining in the column:

$$\text{CIMR} = \frac{\text{MR}_t}{\text{MNR}_t}$$  \hspace{1cm} (10)

where $\text{MR}_t$ and $\text{MNR}_t$ are the total amounts of mass residing in the column after time $t$ for the radioactive and nonradioactive cases, respectively.

3. Results

Deposition characteristics of radioactive aerosols in the column consisting of five vertical layers were investigated using the TOMAS model with and without the average correction factor (hereafter radioactive and nonradioactive cases, respectively). The column spanned altitudes from 0 to 1000 m with a resolution of 200 m, with each layer having the same amount of initial radionuclide mass. Coagulation characteristics of radioactive aerosols in one layer were investigated first before analyzing the vertical transport and deposition of the aerosols. Then the aerosol size distribution and total mass residing in each layer were analyzed to elucidate the effects of the radioactive charging on the deposition of radioactive aerosols. During these investigations using the TOMAS-RC model under various initial conditions, computational issues (e.g., computational instability which can suddenly increase computational costs) were not observed.

Given that multiple charging mechanisms exist and different particle sizes are involved in each one, a wide range of particle size distributions was used. Particles formed through ion-induced nucleation are a few nanometers in size (Harrison and Carslaw, 2003), while particles in volcanic ash or dust clouds that are charged through friction, reside mostly on the coarse mode with diameters greater than 10 $\mu$m (Langmann, 2013). For charged radionuclides resuspended during fires, the bulk of radioactive material is contained in giant particles of diameters greater than 25 $\mu$m as shown for the case of forest
fires conducted in controlled conditions in the Chernobyl exclusion zone (Yoshenko et al., 2006a,b). Note that this refers to radionuclides that were already deposited on the ground and not new particles formed through charging mechanisms.

3.1. Aerosol size distributions

Fig. 3 presents the microphysical evolution, expressed as changes in the size distribution, of $^{137}$Cs aerosols at 400-m altitude over time. The initial size distributions were selected so that a significant number of large particles were present, leading to an appreciable impact of radioactive charging. Shown are the changes that would occur in the presence (solid lines) and absence (dashed lines) of radioactive charging. In the presence of charging effects, $^{137}$Cs aerosols coagulate more slowly than nonradioactive aerosols. This behavior can be seen in a less dramatic shift of the modal peak towards larger aerosol sizes over time when charges are present (Fig. 3). The slow coagulation occurs because numerous negative ions in the atmosphere diffuse more quickly toward the aerosol surfaces (i.e., $X < 1$), so most aerosol acquires a net negative charge (Fig. 4a). Since many particles smaller than 0.8 $\mu m$ are not charged (Fig. 4a), their coagulation is almost unaffected between neglecting and considering charging effects, while particles between 0.8 and 5 $\mu m$ exhibit the most dramatic inhibition (Fig. 3), since these are the particles that accumulate the largest amount of negative charges (Fig. 4a). Particles larger than 5 $\mu m$ carry even larger amounts of negative charge, but are not present in significant concentrations ($d_{g} = 1.5 \mu m$) to affect the shape of the number distribution (Fig. 3).

The evolution of the size distributions of $^{131}$I aerosols at 400-m altitude is presented in Fig. 5. Similarly to $^{137}$Cs, particles below 4 $\mu m$ are negatively charged, inhibiting their coagulation. Even for a low value of $X$ (0.7), however, all particles above 4 $\mu m$ are positively charged (Fig. 4b), leading to notable differences in the predicted distributions when neglecting and considering charging effects. Particles up to 1.3 $\mu m$ in diameter are slightly more negatively charged than $^{137}$Cs, and the coagulation between those particles and the positively charged larger bins with a diameter of 5 $\mu m$ and above is significantly enhanced leading to reduced predicted number concentrations of both the large and small particles when compared to assuming Brownian coagulation alone (Fig. 5). For particles close to the initial modal diameter of 1.5 $\mu m$ (where most aerosol numbers lie), and up to sizes of 3 $\mu m$, coagulation of the particles is strongly inhibited due to their negative charges (Fig. 4b). The rationale behind looking at such a large range of particle sizes is to evaluate the effects of radioactive charging on the microphysical evolution of aerosol populations over a wide range covering from the Aitken to the coarse mode. The presence of such large particles is also expected in dust and volcanic ash clouds (Langmann, 2013), as well as radiological debris created during nuclear events.

3.2. Dry deposition fluxes of radionuclides

Fig. 6 shows contour plots of CIMR from Eq. (10), for the $^{137}$Cs aerosol simulation after 5 days of microphysical evolution, as a function of initial geometric mean diameter and initial concentration. As the geometric mean diameter and total concentration of the initial aerosol size distribution increase, so does the CIMR, indicating that fewer $^{137}$Cs aerosols were deposited. This behavior means that the lifetime of the radioactive aerosol can increase and particles may deposit over a longer distance away from the source. For the more extreme cases, where the concentrations were upwards of $10^{13} \text{ m}^{-3}$ and the geometric mean diameter was around 1 $\mu m$, the coagulation between these particles was strongly inhibited, leading to an appreciable increase in the CIMR and, consequently, a longer lifetime of the aerosol.

Fig. 4. Mean charge values for particle sizes between 0.1 and 10 $\mu m$, for $^{137}$Cs (a) and $^{131}$I (b), for $X$ values of 0.7, 0.8, 0.9 and 1. For the case of $^{137}$Cs, the average charge is negative for all size bins when $0.7 \leq X = 0.8$, leading to inhibition of coagulation for all size bins. For $X = 0.9$, the mean charge is negative for all particles below 3 $\mu m$, but become positive for larger sizes, leading to inhibition of coagulation between particles of small sizes, and enhancement between particles below and above that threshold. $^{131}$I exhibits similar behavior, but because of its much higher decay rate, particles tend to accumulate much smaller negative charges and significantly larger positive ones. For $0.7 \leq X = 0.8$, particles up to 4 $\mu m$ accumulate a small amount of negative charge, while, above that size cutoff all particles are strongly positively charged, implying accelerated coagulation rates between particles belonging to the different sides of the cutoff and inhibition for ones belonging on the same. Larger $X$ leads to the entire size spectrum being positively charged and inhibition across all particle sizes.
diameter was larger than 3 μm, the amount of 137Cs aerosols remaining suspended in the atmosphere, and therefore available for transport, was increased by up to 260% (Fig. 6a), compared to the case of neutral coagulation. Increasing particle concentrations for a given initial particle diameter, meant that the number of particles that could carry a larger amount of same charges was increased, even though the average charge is independent of the number concentration, due to the deviation of the charge distribution (Eq. (6)), leading to inhibition. At the same time, increasing the initial particle size for a constant number concentration augments the number of larger particles that are able to sustain a larger number of charges (Fig. 4a and b), which in turn translates to stronger impacts of charging on coagulation.

It is important to understand the change in deposition behavior of 137Cs particles as the relative mobility of positive vs. negative particles (i.e., X) changes, especially if the radioactive decay has a range of negative/positive ions produced (certainly the case for diffusion charging in air, where the carrier molecules of ions may be N2 or O2 (Harrison and Carslaw, 2003). For situations where X is less than 0.9 (Fig. 6a and b), the entire particle distribution is negatively charged (Fig. 4a), which leads to a distribution-wide inhibition of coagulation. The inhibition is stronger for the lowest value of X = 0.7, since the higher mobility of negative ions leads to particles becoming strongly negatively charged, thereby limiting their interactions. When X ~ 1 (Fig. 6d), particles up to sizes of 3 μm have a mean charge of zero, while larger ones are all positively charged with an average charge increasing exponentially with size (Fig. 4a), meaning that as the concentration of aerosol larger than 3 μm increases, the coagulation between them is suppressed due to the accumulation of large charges of the same sign. In the case of a mobility ratio of less than 0.9 (Fig. 6a and b), the charge of all particles is negative, leading to strong coagulation inhibition.

An interesting behavior is observed on CIMR when X increases from 0.9 to unity (Fig. 6c and d); while the amount of mass deposited is observed between X = 0.9 and 1 (Fig. 6c and d), with the mass deposited decreasing with an increase in X. The reason for this behavior is that the mean charge of each bin changes sign and from negative becomes positive, even for the smaller sized bins (Fig. 4a). This change increases the coagulation rates between the smaller, negatively charged particles and the larger positively charged particles, but at the same time, inhibits the coagulation between the same large particles. An explanation can be found by looking at the definition of the average charge of each bin. For all 137Cs cases simulated, \( N_{\omega} \neq 0.2 \) and therefore \( Y = Y_k + \frac{1}{1+2\omega} \) (Eq. (6)). When X = 0.8, the fractional term is negative and larger than \( Y_k \). However, when X = 0.9 and large enough particle sizes, \( Y_k \) becomes greater than the negative fractional term, leading to these particles becoming positively charged.

Fig. 6. CIMR for 137Cs aerosols after 5 days for X values of (a) 0.7, (b) 0.8, (c) 0.9 and (d) 1; \( \sigma = 2.0 \).

Fig. 7 shows CIMR for 131I aerosols after 5 days under various initial aerosol size distributions. As with 137Cs, less 131I aerosols were deposited, as the mean diameter or total concentration increased. Because the radioactive decay rate of 131I aerosols is nearly 700 times higher than that of the 137Cs aerosols (Table 1), the impact of charging is much more pronounced and can lead to up to a factor of 30 more mass remaining after 5 days of atmospheric processing (Fig. 7d). Similar behavior to 137Cs was observed, with the average charge of some bins changing sign depending on the mobility ratio (Fig. 4b). However, in stark contrast with the 137Cs simulations for the same conditions, all particle sizes above 5 μm are strongly positively charged regardless of the value of the mobility ratio, with the charge of the largest bins being in excess of a 100. The largest impact of radioactive charging for 131I can be seen in the case of X = 0.9 and 1. Looking at the average charge of each bin for these cases, reveals that the entire distribution is positively charged (Fig. 4b), which in turn leads to very strong inhibition between all bins, and a resulting 30-fold increase in mass remaining after 5 days, if the emitted particles contain large amounts of radionuclides (Fig. 7c and d). The effect is slightly more pronounced for the case of a mobility ratio of 0.9, because the maximum charge attained by
the largest particles can be higher than for the case of a mobility ratio of unity (Fig. 4b). For $X$ values of 0.7 and 0.8, particles up to 5 μm carry a negative charge that enhances their coagulation with larger positively charged particles, but the significant positive charges attained by the larger particles (Fig. 4b) still leads to overall inhibition, and subsequently to high CIMR values (Fig. 7a and b), albeit smaller than the ones predicted when $X \geq 0.8$, due to the aforementioned enhancement.

To further determine the impact of possible diffusion charging effects on non-decaying particles, simulations were carried out where the radioactive decay rate of the aerosol population was set to a value of 0. In this case, $\eta_0 = 0 \Rightarrow \omega_0 Y_0 = 0$ and therefore $Y_0 = \frac{Y_1}{\omega_0}$ (Eq. (6)). Diffusion charging can have a significant impact on very coarse aerosol (Gunn, 1954) such as dust and volcanic ash, regardless of its composition. Following the same analysis as with the radioactive particles, the same scenarios were studied and results analyzed using the CIMR. While the impact of diffusion charging on small particles (Fig. 8) is negligible, for very large aerosol and high enough concentrations the impact can be significant, especially in environments where many ion pairs are produced, and where the mobility of negative ions is greater than that of the positive ones ($X < 1$). For such cases, all particles gather large negative charges and enhancement factors of less than unity, which inhibits their coagulation and lead to increased aerosol lifetimes.

For all cases described above, the geometric standard deviation of the initial aerosol size distribution, $\sigma_g$, was also found to affect CIMR. Larger values of $\sigma_g$ simultaneously increase the number of small and large particles, which in effect augments both the enhancement and inhibition for all cases. Inhibition tends to be favored, since wider distributions carry a higher number of large particles, which subsequently become a larger portion of the total aerosol mass, given that the mass scales cubically with size. These large particles tend to carry the largest amount of charges (Fig. 4a and b) and are affected the most by radioactive charging. Therefore, increasing their mass fraction for any value of the mobility ratio will lead to overall inhibition, and also extend the impact of radioactivity to lower initial particle concentrations and diameters, since even in this case an abundance of large particles will be present.

4. Conclusions

We present the development of the TOMAS-RC model, an expanded version of the TOMAS microphysics model to explicitly treat the effects of radioactive charging on the microphysical evolution during the transport and deposition of radioactive particles away from a radiation source. Electrostatic charges induced by radioactive decay, through the mechanisms of self- and diffusion charging, can magnify or reduce the coagulation rates of radioactive particles by producing electrostatic attractive or repulsive forces. Effects are introduced as a multiplication factor applied to the Brownian coagulation kernel of TOMAS; explicit treatment of the discretized nature of the size distribution, the charge distribution within each size bin and accelerations of the calculation procedure are discussed in detail.

The TOMAS-RC model was applied to study the dispersion and microphysical evolution of radioactive particles within a vertically-dispersing atmosphere, and simulations were carried out for aerosols carrying numerous $^{131}$I and $^{137}$Cs atoms for a wide range of initial aerosol conditions and ion mobilities. Radioactive charging for both radionuclides was found to often have an important impact on the evolution of the particle size distributions and subsequent dry deposition of radioactive aerosol. The effects increased as the initial size or concentrations of radioactive particles increased, due to different ion concentrations determined by equations (7) and (8). The ratio of mobilities of the positive ions to the negative ions, $X$, was found to also have a very strong effect on the evolution of the particle size.
distribution and deposited particles, especially for the case of $^{137}\text{Cs}$ particles, where depending on the value of $X$, some bins of the aerosol distribution can switch sign and behavior altogether (charging enhancing vs. inhibiting coagulation). It should be noted that in this study, the charge signs of $^{137}\text{Cs}$ and $^{131}\text{I}$ aerosols can be opposite because the signs depend on their radioactivity levels and ion concentrations which determine their dominating charging mechanisms, as shown for the case of particles larger than 5 μm for all $X$ values.

Through a series of highly idealized simulations, we have shown that the charging of radioactive aerosols can have an appreciable effect on how particles are deposited, with important implications for predicting dispersal patterns after nuclear accidents. However, it has been assumed that only one type of radionuclide is present. More realistic aerosol may contain external or internal mixtures of radionuclides, with very different microphysical evolution from the ones describe here. For example, if both $^{137}\text{Cs}$ and $^{131}\text{I}$ are present in the same column as externally-mixed aerosol, the coagulation would be highly facilitated and thus the deposition rates of radioactive aerosols should be enhanced. Multigenerational products from the decay of radionuclides during atmospheric transport can create conditions of variable $X$ (dependent on particle size and composition) that can lead to very different depositional patterns as well. Coagulation of externally mixed bins, or condensation/evaporation of semi-volatile radionuclides (e.g. $^{131}\text{I}$, $^{132}\text{I}$) can create particles of varying compositions, which in turn translates to different rates of radioactive decay and charging mechanisms. Further decay products of radionuclides (e.g. $^{132}\text{Te}$ to $^{132}\text{I}$) can shift the balance between diffusion and self-charging (Kim et al., 2017), and that behavior can induce significant uncertainties in modeling.

Since this study only considers dry deposition, the potential role of radioactive charging on wet scavenging is not considered. However, the potential acceleration of the coagulation between fine particles from charging could make these particles larger and therefore more effective cloud condensation nuclei (CCN), increasing the efficiency by which they are removed from the atmosphere through wet deposition and warm rain process modulation. Coagulation of radionuclides with larger, charged dust particles which are very efficient ice nuclei (IN) may also affect deposition patterns by promoting cold rain processes. Future and ongoing modeling activities should focus on a full account of all such effects to fully elucidate the impact that radioactive charging effects can have on the transport of radioactivity across the globe.

Code and data availability

The TOMAS-RC code, as well as the data, are available upon request.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.


