Surface Charge Accumulation of Particles Containing Radionuclides in Open Air

Yong-ha Kim,¹ Sotira Yiacoumi,^{1,*} and Costas Tsouris^{1,2}

¹Georgia Institute of Technology, Atlanta, Georgia 30332-0373, United States

²Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6181, United States

*Corresponding author: E-mail sotira.yiacoumi@ce.gatech.edu

Submitted for publication in

Journal of Environmental Radioactivity

December 2014

Revised manuscript submitted in February 2015

Notice: This manuscript has been authored by UT-Battelle, LLC under Contract No. DE-AC05-00OR22725 with the US Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes. The Department of Energy will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (<http://energy.gov/downloads/doe-public-access-plan>).

Graphical Abstract

Surface Charge Accumulation of Particles Containing Radionuclides in Open Air

ABSTRACT. Radioactivity can induce charge accumulation on radioactive particles. However, electrostatic interactions caused by radioactivity are typically neglected in transport modeling of radioactive plumes because it is assumed that ionizing radiation leads to charge neutralization. The assumption that electrostatic interactions caused by radioactivity are negligible is evaluated here by examining charge accumulation and neutralization on particles containing radionuclides in open air. A charge-balance model is employed to predict charge accumulation on radioactive particles. It is shown that particles containing short-lived radionuclides can be charged with multiple elementary charges through radioactive decay. The presence of radioactive particles can significantly modify the particle charge distribution in open air and yield an asymmetric bimodal charge distribution, suggesting that strong electrostatic particle interactions may occur during short- and long-range transport of radioactive particles. Possible effects of transported radioactive particles on electrical properties of the local atmosphere are reported. The study offers insight into transport characteristics of airborne radionuclides. Results are useful in atmospheric transport modeling of radioactive plumes.

1. Introduction

Atmospheric dispersion of particles containing radionuclides may cause environmental and public health problems. The Chernobyl and Fukushima accidents led to radioactive contamination in many regions through transport and deposition of radionuclides (Chesser et al., 2004; Ooe et al., 1988; Pöllänen et al., 1997; Parache et al., 2011). Some areas may be contaminated for a long period of time due to the presence of long-lived radionuclides [e.g., ^{137}Cs (Yoshenko et al., 2006a, 2006b)]. Thus, accurate prediction of transport of radioactive particles is needed from the beginning of a nuclear event in order to reduce uncertainty in our evaluations of environmental and health risks.

Modeling efforts have been made to predict transport of airborne radionuclides (ApSimon et al., 1989; Lee and Feichter, 1995; Rehfeld and Heimann, 1995; Pöllänen et al., 1997). However, discrepancies between theory and observations have been reported. For the Chernobyl case, large radioactive particles, which in theory should settle quickly, were transported to areas far away from the site of the accident (Lujanienė et al., 2009; Pöllänen et al., 1997). Predictions that did not follow well the observations were also reported for other cases of radioactivity transport (ApSimon et al., 1989; Lee and Feichter, 1995; Rehfeld and Heimann, 1995). These discrepancies emphasize the need to improve predictive modeling of radioactivity transport.

The nature of particulates in radioactive plumes plays a major role in radioactivity transport (Pöllänen et al., 1997). So far, it has been assumed that, because radionuclides are typically carried by background aerosols (Kaneyasu et al., 2012), radioactive particles behave similarly to atmospheric particles, whose electrostatic surface interactions are neglected (Seinfeld and Pandis, 2006). It has been shown, however, that radioactive particles can be strongly charged (Clement

and Harrison, 1992; Gensdarmes et al., 2001; Kim et al., 2014; Kweon et al., 2013; Reed et al., 1977; Walker et al, 2010; Yeh et al., 1976); therefore, the assumption of neglecting electrostatic interactions may cause inaccuracies in predicting transport of radioactive particles. Nevertheless, the assumption has been used in many cases with the explanation that the presence of ionizing radiation induces charge neutralization of particles in open air (Greenfield, 1956, 1957). For example, Greenfield (1957) employed this assumption and found that the wet scavenging rate of radioactive particles can significantly decrease for a particle size range of 0.1 - 1 μm . This size range is typically called the Greenfield gap (Seinfeld and Pandis, 2006; Shaw, 1986). To reduce uncertainty in predictive modeling of radioactivity transport, there is a need to investigate whether radioactive particles can be charged in open air in order to evaluate the convenient assumption of negligible electrostatic interactions. In particular, although radioactive particles typically contain mixtures of radionuclides (Cuddihy et al., 1989; Devell et al., 1986; Kaneyasu et al., 2012; Kauppinen et al., 1986; Pöllänen et al., 1997), little information has been reported about ionizing radiation and particle charging under such conditions. Thus, this study examines charge accumulation and neutralization on the surface of particles containing various radionuclides in open air, under realistic conditions, to assess the importance of electrostatic interactions caused by radioactivity.

2. Methodology

2.1. Characteristics of Radioactive Particles

Various radionuclides (e.g., ^{99}Mo , ^{103}Ru , ^{106}Ru , ^{131}I , ^{132}Te , ^{134}Cs , and ^{137}Cs) were found in particles released from the Chernobyl and Fukushima accidents (Cuddihy et al., 1989; Devell et al., 1986; Kaneyasu et al., 2012; Kauppinen et al., 1986; Pöllänen et al., 1997). The radioactivity

65 η of individual particles containing radionuclides can be given by the number of radioactive
 66 atoms and decay constant λ :

$$67 \quad \eta_k = \sum_i \eta_{k,i} = \sum_i \frac{m_k N_A f_i}{\mu_{mean}} \lambda_i, \quad (1)$$

68 where m_k is the mass of size k particles, N_A is Avogadro's number, f_i is the mole fraction of a
 69 radionuclide i and μ_{mean} is the mean atomic weight.

70 For beta decay, radionuclides emit electrons with kinetic energy T . Energy deposition by the
 71 emitted electrons causes atomic electron ejection from gas molecules in air, thereby generating
 72 ion pairs. The mean number of ion pairs produced per decay I (i.e., ionization rate coefficient)
 73 can be estimated using the linear energy transfer (LET) equation (Huizenga and Storchi, 1989;
 74 ICRU, 1984; Tomé and Palta, 1998) and the additivity rule (Bragg and Kleeman, 1905):

$$75 \quad I_i = \left[\sum_l f_l \left(\frac{1}{\rho} \frac{-dE}{dx} \right)_{\Delta,l} \right] \frac{\delta_i}{w_{air}}, \quad (2)$$

76 where f_l is the weight fraction of an element l , ρ is the air density, Δ is the cut-off energy, δ is the
 77 path length of electrons, and w_{air} is the ionization potential. LET, $(-dE/dx)_{\Delta,l}$, represents the mean
 78 energy loss of energetic electrons during their movement and involves energy transfer to air
 79 molecules. LET can be given as

$$80 \quad \left(\frac{1}{\rho} \frac{-dE}{dx} \right)_{\Delta,l} = \frac{2\pi r_e^2 m_e c^2}{\beta^2} \frac{Z}{uA} \left[\ln \left(\frac{T}{I_{excitation}} \right)^2 + \ln \left(1 + \frac{\tau}{2} \right) + G(\tau, \eta) \right] \quad (3)$$

where r_e is the classical electron radius, m_e is the rest mass of the electron, c is the velocity of light, β is the ratio of electron velocity to the velocity of light, Z is the atomic number, u is the atomic mass unit, A is the atomic weight, $I_{excitation}$ is the mean excitation energy of the element, G is the auxiliary function (Tomé and Palta, 1998), $\tau = T/m_e c^2$ and $\eta = \Delta/T$. In equation 3, the sum of the three terms in square brackets on the right-hand side (RHS) represents the dimensionless stopping number for elements (Seltzer and Berger, 1982). In particular, the last term restricts the amount of transferred energy up to Δ .

We assumed that air consists of N_2 , O_2 , CO_2 , and Ar and that the weight fraction of N, O, C, and Ar is comparable to the dry air around sea level. To secure validity of eq 3, the binding energies of atomic electrons should be smaller than Δ . Similarly to ICRU (1984) and Tomé and Palta (1998), it was assumed that $\Delta = 1$ keV for N, O, and C and $\Delta = 10$ keV for Ar, which can be greater than the binding energies of atomic electrons in K- and L-shells of the elements. Ionizing radiation caused by beta decay can polarize elements, which can influence LET. However, this effect is negligible for electrons with $T \leq 10$ MeV (ICRU, 1984). $I_{excitation}$ of the elements can be obtained from the empirical formula (Marmier and Sheldon, 1969). For beta decay, $w_{air} = 34$ eV (Whyte, 1963).

The path length of energetic electrons depends on their kinetic energy. Because it is difficult to predict the tortuous path of energetic electrons, the mean path length of the electrons was estimated using the semi-empirical range-energy relationship given by Katz and Penfold (1952):

$$R = 0.412T^{1.265-0.0954\ln T} \quad (0.01 < T \leq 2.5 \text{ MeV}) \quad (4)$$

where R is the range of electrons in g/cm^2 . In contrast to alpha decay, beta decay exhibits a continuous energy distribution because of the creation of antineutrinos. In this study, the ionization coefficients of radionuclides and the mean path length of energetic electrons were estimated using the mean value of the energy distribution, E_{mean} . Equations 1-2 provide the number of electrons emitted and ion pairs produced by beta decay of radionuclides.

2.2. Charge Balance Model

Electron emission and diffusion of positive and negative ions induce charge accumulation and neutralization on radioactive particles (Kim et al., 2014). Electron emission refers to the self-charging mechanism, while ion diffusion corresponds to the diffusion-charging mechanism. For beta decay, the net charge accumulation rate on radioactive particles is given by:

$$\frac{dJ}{dt} = \eta + \beta_J^+ n_+ - \beta_J^- n_- , \quad (5)$$

where J is the charge number, $\beta_J^\pm$ is the ion-particle attachment coefficient, $n_\pm$ is the ion concentration, and t is time. Clement and Harrison (1992) extended eq 5 to investigate the charging of monodispersed particles containing single radionuclides. Based on the work of Clement and Harrison, a sectional approach can give the charge distribution of polydispersed particles containing various radionuclides as:

$$\frac{dN_{kj}}{dt} = \eta_{k,j-1} N_{k,j-1} - \eta_{k,j} N_{k,j} + \beta_{k,j-1}^+ n_+ N_{k,j-1} - \beta_{k,j}^+ n_+ N_{k,j} + \beta_{k,j+1}^- n_- N_{k,j+1} - \beta_{k,j}^- n_- N_{k,j} , \quad (6)$$

where N_{kj} is the number concentration of radioactive particles of size k and charge j . In eq 6, the first two terms on the RHS represent the charge accumulation rate by the self-charging

mechanism and the remaining terms account for the charge accumulation rate by the diffusion-charging mechanism. The ion-particle attachment, ion-ion recombination, ionization, and electron emission can influence the ion concentration (Clement and Harrison, 1992; Renard et al., 2013), through the following equations:

$$\frac{dn_+}{dt} = -n_+ \sum_k \sum_j \beta_{k,j}^+ N_{k,j} - \alpha_{rc} n_+ n_- + q, \quad (7)$$

$$\frac{dn_-}{dt} = -n_- \sum_k \sum_j \beta_{k,j}^- N_{k,j} - \alpha_{rc} n_+ n_- + q + \sum_k \sum_j N_{k,j} \eta_{k,j}, \quad (8)$$

where α_{rc} is the recombination coefficient and q is the bipolar ion production rate. In eqs 7 and 8, the first two terms on the RHS represent ion loss through ion-particle attachment and ion-ion recombination. The third term represents ion production by beta radiation, as well as other ion sources, and can be obtained using

$$q = \sum_i q_{l,i} + q_b. \quad (9)$$

where $q_{l,i}$ and q_b are the bipolar ion production rates of beta-emitting radionuclides and other ion sources, respectively. $q_{l,i}$ can be calculated by multiplying the number of electrons emitted per unit time with their ionization rate coefficient. The last term of eq 8 accounts for the emitted electrons. Effects of each radionuclide on charge accumulation are included using η and $q_{l,i}$. Eqs 6-8, without radioactivity terms, can be valid in predicting electrification phenomena of aerosols in the upper troposphere and stratosphere (Renard et al., 2013). In this study, the ion-particle attachment coefficient in the charge balance equation was estimated according to Harrison and Carslaw (2003).

2.3. Experimental Data

Experimental data from the literature were used to validate the charge balance model and ionization rate coefficient (Table 1). The charge distribution of ^{137}Cs particles was measured as a function of η and I . The charge accumulation of ^{198}Au particles was investigated as a function of η . The experiments were carried out in cylindrical tubes. Under such condition, the mean path length of electrons can be estimated as a function of the tube diameter, d_{tube} on the basis of the criteria of Clement and Harrison (1992) and Gensdarmes et al. (2001): if $\delta_{i,mean} < d_{tube}$, $\delta_i = (\pi/4)d_{tube}$, while $\delta_i = d_{tube}$ if $\delta_{i,mean} > d_{tube}$.

Table 1. Experimental observations on charging of beta-emitting radioactive particles

Radionuclide		¹³⁷ Cs				¹⁹⁸ Au		
Case number		1	2	3	4	5	6	7
Radioactivity <i>η</i> (mBq)		12.8	3.2	12.8	3.2	470	210	77
Ionization rate coefficient <i>I</i>		52	4,395	52	4,395	-	-	-
Particle diameter (μm)		0.82	0.82	1.05	1.05	0.53	0.53	0.53
Tube diameter (cm)		0.6	40	0.6	40	1.5	1.5	1.5
Ion mobility: (cm ² V ⁻¹ s ⁻¹)	Positive ion	1.19				1.2		
	Negative ion	1.54				1.2		
References		<i>Gensdarmes et al. (2001)</i>				<i>Clement and Harrison (1992)</i> <i>Yeh et al. (1976)</i>		

mBq: milliBecquerels

3. Results and Discussion

3.1. Ionization Rate Coefficient

The ionization rate coefficient I of radionuclides was estimated using LET (eq 2) and evaluated employing the charge balance model and experimental data (Cases 1-7). For comparison, I was also estimated from a typical approach using the Beer-Lambert (BL) law (Clement and Harrison, 1992; Cooper and Reist, 1973; Gensdarmes et al., 2001). Hereafter, we refer to estimation by the LET or BL approach, respectively. Note that the BL approach employed in this study includes the empirical formula for air (Martin, 2000), while previous studies have used that obtained for aluminium (Clement and Harrison, 1992; Cooper and Reist, 1973; Gensdarmes et al., 2001).

Figure 1(a) shows the charge accumulation rates for Case 1. The ionization rate coefficient was estimated as $I_{LET} = 29$ by the LET approach and $I_{BL} = 126$ by the BL approach. Both approaches predicted that ^{137}Cs particles initially accumulated positive charge, which neutralized the negative surface charge assumed initially based on the experiments by Gensdarmes et al. (2001). However, the results with I_{LET} were closer to the experimental data than those with I_{BL} . The discrepancy resulted from the difference in the ion concentration caused by ionization. For the case with I_{LET} , the ion concentration in the system increased more slowly and was lower than that estimated by I_{BL} , causing the particles to accumulate more positive charges through the self-charging mechanism until the diffusion-charging mechanism was strong enough to counterbalance the surface charge. As shown in Table 1, the mobility of negative ions is greater than that of positive ions, and this is the reason that the diffusion-charging mechanism neutralizes positive charge added by the self-charging mechanism.

Figure 1. Charge accumulation for Case 1 (a) and charge distribution for Cases 1-4 (b-e).

to obtain LET. To compute the amount of energy transferred to air molecules, we employed the mean kinetic energy obtained from the beta spectrum of ^{137}Cs [$E_{mean} = 0.192$ MeV (Mantel, 1972)], while Gensdarmes et al. (2001) assumed that the mean kinetic energy is one fourth of the maximum kinetic energy ($E_{mean} = 0.128$ MeV). For ^{137}Cs , a lower E provides a higher value for LET, thereby increasing its ionization rate coefficient. In this study, LET was estimated using the restricted LET formula with the cut-off energy, while Gensdarmes et al. (2001) obtained LET from a chart in Joffre (1963) without providing specific assumptions. Nevertheless, the LET approach of this study predicted the charge accumulation rates for Cases 1 and 3 more accurately than the LET approach described in Gensdarmes et al. (2001) [Figure 1(b) and 1(d)]. For Cases 2 and 4, the results of both approaches agreed well with the measurements [Figure 1(c) and (e)], but the LET approach forecasted more precisely the charge accumulation behavior of the ^{198}Au particles (Cases 5 to 7) (Figure 2). Compared to Reed et al. (1977) and Clement and Harrison (1992), the discrepancy in the predictions for Cases 5 - 7 results mainly from employing different ion mobilities and ionization rate coefficients. These results suggest that the LET approach employed in this study can reliably estimate ionization rate coefficients of radionuclides to predict charge accumulation on radioactive particles.

3.2. Charge Accumulation on Particles Containing Radionuclides

Charge accumulation on particles containing radionuclides was investigated using the charge balance model with eqs 1-3 under the experimental conditions of Cases 1-4. The mole fraction of ^{137}Cs contained in the particles was estimated using eq 1. It was assumed that (i) various short-lived and long-lived radionuclides are added to the ^{137}Cs particles, (ii) the mole fraction of added radionuclides equals that of ^{137}Cs , and (iii) their radioactivity is constant because of short charge accumulation time.

Figure 2. Charge accumulation rate for Cases 5-7: (a) Case 5, (b) Case 6, and (c) Case 7.

Figure 3 shows charge distributions of ^{137}Cs particles containing other radionuclides under the experimental conditions of Cases 1 and 2. The relative difference of charge distributions represents the effects of each radionuclide on charge accumulation rates. The charge distributions of ^{137}Cs particles became quite different as short-lived radionuclides were added. The contained short-lived radionuclides increased the self-charging rates of the particles, thus accumulating more positive charges. Despite differences in ionization rate coefficients, the ^{137}Cs particles accumulated more positive charges as the half-life of the added radionuclides was much shorter ($^{132}\text{Te} > ^{131}\text{I} > ^{134}\text{Cs}$). As more ion pairs were created, the positively charged radioactive particles were effectively neutralized due to the diffusion of more negative ions (Cases 1 vs. 2).

Figure 3. Charge distributions of particles containing short- and long-lived radionuclides: (a) Case 1 and (b) Case 2.

Particles of lower mole fraction of radioactive elements accumulated fewer positive charges, but the effects of the contained short-lived radionuclides persisted (Cases 1 vs. 3 and 2 vs. 4; data not shown). These results suggest that the type, amount, and ionization rate coefficient of radionuclides contained in particles influence significantly charge accumulation on radioactive particles.

3.3. Charge Accumulation on Particles Containing Radionuclides in Open Air

3.3.1. Ionization Rate Coefficient of Beta-emitting Radionuclides in Dry Air

The ionization rate coefficient of various beta-emitting radionuclides in dry air was estimated using the LET approach. Energetic electrons can freely travel until their energy is completely dissipated. Compared to confined spaces (e.g., Case 1), the path length of energetic electrons can be maximized in open spaces, which can affect results of the LET approach (eq 2). Thus, the validity of the Katz and Penfold relationship (eq 4) was evaluated using the generalized semi-empirical range–energy relationship for monoenergetic electrons (Tabata et al., 1972) and the continuous-slowing-down approximation (CSDA) (ICRU, 1984) which can compute the mean path length of electrons in dry air. For various radionuclides released from the Chernobyl and Fukushima accidents, the range of energetic electrons given by eq 4 was similar to that by CSDA as well as that by the relationship for monoenergetic electrons (Figure 4), suggesting that the Katz and Penfold relationship can be used to approximate the mean path length of electrons in dry air.

Figure 4. Mean path length of electrons emitted during beta decay of various radionuclides in dry air.

Table 2 shows the ionization rate coefficient of various beta-emitting radionuclides estimated using the LET approach with the Katz and Penfold relationship.

Table 2. Ionization rate coefficient of various beta-emitting radionuclides in dry air
(Temperature = 295 K; Pressure = 100 kPa)

Radionuclide	Half-life	Energy of electrons (MeV)		Mean path length of electrons (cm)	Ionization rate coefficient, I
		E_{max}^a	E_{mean}^b		
^{95}Zr	64.032 d	0.3960	0.1191	15.35	1,261
^{95}Nb	34.991 d	0.1600	0.0463	2.91	472
^{99}Mo	2.7489 d	1.1800	0.3803	93.93	4,323
^{103}Ru	39.26 d	0.2294	0.0765 ^c	7.19	800
^{106}Ru	373.59 d	0.0394	0.0131 ^c	0.24	110
^{132}Te	3.204 d	0.2150	0.0717 ^c	6.41	748
^{131}I	8.01 d	0.6080	0.1812	30.43	1,945
^{134}Cs	2.0652 yr	0.6620	0.1581	24.45	1,688
^{137}Cs	30.16 yr	0.5140	0.1921	33.39	2,067
^{140}Ba	12.752 d	1.0200	0.2690	56.22	2,956
^{140}La	1.6781 d	1.3600	0.4360	114.30	5,041
^{141}Ce	2.508 d	0.4350	0.1453	21.32	1,547
^{144}Ce	284.91 d	0.3200	0.0778	7.41	814
^{239}Np	2.356 d	0.4380	0.1460 ^c	21.49	1,554

^a Mantel (1972) and Martin (2000).

^b E_{mean} of radionuclides was obtained via Mantel (1972) except for ^{103}Ru , ^{106}Ru , ^{132}Te , and ^{239}Np .

^c It was assumed that $E_{mean} = 1/3E_{max}$ (Cooper and Reist, 1973).

3.3.2. Charge Distribution of Radioactive Particles in Open Air

3.3.2.1. Testing the Validity of the Assumption of Greenfield (1956)

Charge accumulation on particles containing radionuclides was investigated under the ionization condition of Greenfield (1956), who showed that up to 75% of monodispersed radioactive particles ($\eta \approx 0.118$ Bq, $I = 15,000$, $N_t = 3.55 \times 10^{10}$ particles m^{-3}) are neutralized due to beta radiation. Figure 5 shows the steady-state number fraction of the charged and uncharged radioactive particles under the ionization condition of Greenfield (1956): $q_t = \eta I N_t = 6.3 \times 10^{13}$ ion pairs $\text{m}^{-3} \text{s}^{-1}$. The numbers of the positively and negatively charged particles were similar because of the symmetric particle charge distribution. Despite surface charge neutralization caused by beta radiation, more charged radioactive particles were generated at 250 K than estimated by Greenfield (1956). A similar result was obtained for 295 K. These discrepancies mainly resulted from the ion-particle attachment coefficient in eqs 6-8. We used the typical values of the ion-particle attachment coefficient in the lower atmosphere (e.g., $\beta_{-1}^+ = \beta_{+1}^- = 2.7 \times 10^{-12} \text{ m}^3 \text{s}^{-1}$ and $\beta_0^\pm = 1.5 \times 10^{-12} \text{ m}^3 \text{s}^{-1}$ at 250 K; $\beta_{-1}^+ = \beta_{+1}^- = 3.2 \times 10^{-12} \text{ m}^3 \text{s}^{-1}$ and $\beta_0^\pm = 1.9 \times 10^{-12} \text{ m}^3 \text{s}^{-1}$ at 295 K), while Greenfield assumed that $\beta_{-1}^+ = \beta_{+1}^- = 6 \times 10^{-12} \text{ m}^3 \text{s}^{-1}$ and $\beta_0^\pm = 10^{-12} \text{ m}^3 \text{s}^{-1}$ at 250 K. The values of the attachment coefficient estimated by Greenfield are beyond the typical range of values suggested more recently by many researchers (Harrison and Carslaw, 2003; Hoppel and Frick, 1986). The difference in the values of the ion-particle attachment coefficient resulted in dissimilar charge accumulation and neutralization rates of the radioactive particles, leading to the discrepancies mentioned above. The results of this study suggest that many radioactive particles can be charged despite the occurrence of charge neutralization induced by beta radiation in open air.

Figure 5. Steady-state number fraction of charged and uncharged radioactive particles under the ionization condition of Greenfield (1956).

3.3.2.2. Effects of Radioactive Particles on Particle Charge Distribution in the Atmosphere

The Chernobyl accident released many particles containing beta-emitting radionuclides into the atmosphere. During short- and long-range transport, the radioactive particles became charged and ionized air molecules, thereby affecting the particle charge distribution in the atmosphere. Thus, effects of the Chernobyl radioactive particles on the particle charge distribution in the atmosphere were investigated here, using the charge balance model.

For single particle composition, we used observations of Devell et al. (1986) who measured properties of individual Chernobyl radioactive particles. Hereafter, we refer to the radioactive

274 particles by Ch1, Ch2, and Ch3: Ch1 contained ^{103}Ru , ^{99}Mo , ^{106}Ru , ^{132}Te , and ^{131}I ; Ch2 involved
 275 ^{103}Ru , ^{106}Ru , ^{132}Te , and ^{131}I ; Ch3 consisted of ^{106}Ru , ^{131}I , ^{140}Ba , ^{140}La , ^{95}Zr , ^{95}Nb , ^{141}Ce , ^{144}Ce ,
 276 and ^{239}Np . The radioactivity of each radionuclide was given by Devell et al. (1986). The total
 277 radioactivity of Ch1, Ch2, and Ch3 was 15,280 Bq, 8,780 Bq, and 2,740 Bq, respectively. The
 278 size distribution of background aerosols (BA) was obtained from Trier (1997). It was assumed
 279 that (i) the monodispersed Chernobyl radioactive particles were mixed with BA, (ii) their total
 280 particle number concentrations were similar, and (iii) all particles were initially neutralized. The
 281 charge balance equation was defined for each particle group and all equations were solved
 282 simultaneously. The ionization rate coefficients shown on Table 2 were used to calculate $q_{l,i}$ in
 283 eqs 7 and 8. Some typical values used in simulation were obtained from Harrison and Carslaw
 284 (2003) by assuming the fair-weather atmosphere (e.g., $q_b = 10^7$ ion pairs $\text{m}^{-3} \text{s}^{-1}$ and $\alpha_{rc} =$
 285 $1.6 \times 10^{-12} \text{ m}^3 \text{s}^{-1}$.)

286 Figure 6 shows the steady-state particle charge distributions in fair-weather atmosphere
 287 involving various Chernobyl radioactive particles. Beta radiation caused by the radioactive
 288 particles generated numerous ion pairs in the atmosphere. The ions were rapidly captured by the
 289 background aerosols and the particles quickly obtained a unimodal symmetrical charge
 290 distribution (i.e., the Boltzmann distribution). However, despite ionization induced by beta
 291 radiation, most Chernobyl radioactive particles accumulated more positive than negative charges
 292 because of the self-charging mechanism. The presence of the positively charged particles
 293 produced asymmetric bimodal particle charge distributions in the atmosphere. When the self-
 294 charging mechanism was neglected, all particle charge distributions became Boltzmann
 295 distributions.

Figure 6. Particle charge distributions in fair-weather atmosphere containing various Chernobyl radioactive particles (Ch1, Ch2, and Ch3; $d_p = 1 \mu\text{m}$). The geometric mean diameter, d_g and geometric standard deviation σ_g of the background aerosols (BA) are $0.116 \mu\text{m}$ and 1.46 , respectively (Trier, 1997). $N_{BA} = N_{Ch1} = N_{Ch2} = N_{Ch3} = 6.718 \times 10^9 \text{ particles m}^{-3}$.

The particle charge distributions in fair-weather atmosphere involving radioactive particles were investigated using polydispersed conditions. We assumed ammonium sulfate particles carrying ^{103}Ru , ^{99}Mo , ^{106}Ru , ^{132}Te , and ^{131}I which correspond to the radioactive composition of Ch1. Radioactivity of single particles was calculated using eq 1. The initial size distribution of polydispersed radioactive particles was obtained from Tripathi and Harrison (2001).

Figure 7 shows the steady-state particle charge distributions in the atmosphere containing the polydispersed radioactive particles. For all cases, the particle charge distribution was different from the Boltzmann distribution. Some particles were highly radioactive and accumulated many

positive charges until reaching steady state. As the mole fraction of the radionuclides increased, the number of highly radioactive particles increased and thus, more positively charged particles were generated. If the mole fraction of the radionuclides is similar, large particles can contain more radionuclides than small particles (eq 1). Thus, the self-charging rate of single radioactive particles was dissimilar for different-size particles, and this led to the creation of the asymmetric multimodal particle charge distributions in the atmosphere.

Figure 7. Particle charge distributions in fair-weather atmosphere containing background aerosols and particles containing ^{103}Ru , ^{99}Mo , ^{106}Ru , ^{132}Te , and ^{131}I . $d_{g,BA} = 0.116 \mu\text{m}$, $\sigma_{g,BA} = 1.46$, $d_{g,ChI} = 0.52 \mu\text{m}$, $\sigma_{g,ChI} = 2.4$, and $N_{BA} = N_{ChI} = 6.718 \times 10^9 \text{ particles m}^{-3}$.

In the simulations, we assumed similar ion mobilities for positive and negative ions, which correspond to those in fair-weather atmosphere (Harrison and Carslaw, 2003), but positive ions

are typically less mobile than negative ions (Clement and Harrison, 1992; Hoppel and Frick, 1986; Gensdarmes et al., 2001). However, the asymmetric bi- and multi-modal charge distributions created by the Chernobyl radioactive particles were maintained for both equal and unequal ion mobilities of positive and negative ions (data not shown).

In fair-weather atmosphere, where cosmic ray and natural radioactivity are major ionization sources, the time needed to reach a steady-state charge distribution can be delayed due to low ion concentrations (Yair and Levin, 1995), but for all cases, the steady-state particle charge distribution was attained within a few seconds due to the rapid ionization by beta radiation. Thus, other processes, such as aggregation, may not affect the steady-state charge distributions caused by the Chernobyl radioactive particles because of the short simulation time, as well as the low particle concentrations. Effects of aggregation on the particle charge distributions should be evaluated if timescale to reach the steady-state particle charge distributions is long or particle concentrations are high.

These results suggest that radioactive particles can significantly influence the particle charge distributions in the atmosphere during short- and long-range transport.

3.3.2.3. Possible Effects of Radioactive Particles on Transport of Radioactivity and Atmospheric Electricity

Charge accumulation on radioactive particles can significantly influence short- and long-range transport of radioactivity because it induces strong electrostatic interactions. The generation of an asymmetric bimodal particle charge distribution would produce strong electrostatic particle-

particle interactions, which would greatly affect particle growth (Matsoukas, 1997; Oron and Seinfeld, 1989a, 1989b). For example, positively charged radioactive particles may attract negatively charged background aerosols. Additionally, radioactive particles can favorably interact with raindrops due to the electrostatic image force generated between them. Recent experiments of Walker et al. (2010) suggested that radioactivity increased the adhesion force between a charged particle and a conductive surface through the generation of the attractive image force. Radioactive surfaces can be charged at humid conditions (Kweon et al., 2013) and the surface charge of the particles in the proximity of raindrops can generate an attractive electrostatic image force (Tripathi and Harrison, 2001). Thus, the scavenging rate of charged radioactive particles may be enhanced by radioactivity-induced electrostatic interactions. Hence, the dry and wet deposition rates of radioactive particles may be significantly affected by electrostatic interactions caused by radioactivity.

During atmospheric transport, radioactive particles can significantly influence the electrical properties of the local atmosphere because they can ionize air molecules. Table 3 shows changes in electrical properties of the local atmosphere by radioactive particles measured after nuclear events, including the Chernobyl accident. During short- and long-range transport, radioactive particles were accumulated on ground surfaces via various mechanisms, such as thundershower, and dry and wet deposition. For example, radioactive particles flying at 5-km altitude were carried to ground surfaces by thundershower (Harris, 1955). Ionization induced by the radioactive particles increased ion concentrations near ground surfaces and this led to changes in electrical properties of the local atmosphere. High ion concentrations increased the electrical conductivity of the local atmosphere to much higher than normal levels. This could result in short-circuiting of the electric field (Israelsson and Knudsen, 1986; 1987), thereby decreasing

potential gradient and space charge in the local atmosphere. The duration time of these effects may depend on radioactivity in air and on ground surfaces because radioactive particles can be transported to other places by various processes, such as runoff (Israelsson and Knudsen, 1986) and wind-driven resuspension (Yamauchi et al., 2012).

Table 3. Changes in electrical properties of the local atmosphere by transported radionuclides.

Nuclear events	Emission sources	Observations	Distance from emission sources	Major transport mechanisms (from the local atmosphere to ground level)	Electrical conductivity ^a	Potential gradient ^a	Space charge ^a	Reference
Weapon tests	Nevada, USA	Tucson, USA	850 km	Thundershower	10 times increase	6 times decrease	-	Harris (1955)
Nuclear plant accidents	Chernobyl, Ukraine	Uppsala, Sweden	1,300 km	Wet deposition	11 times increase	10 times decrease	10 times decrease	Israelsson and Knudsen (1986)
		Athens, Greece	1,500 km	Dry deposition	7 times increase	2 times decrease	-	Retalis and Pitta (1989)
		Vantaa, Finland	1,050 km	Dry and wet deposition	10 times increase	10 times decrease	-	Toumi (1989)
	Fukushima, Japan	Kakioka, Japan	150 km	Dry and wet deposition	-	10 times decrease	-	Takeda et al. (2011)
		Kakioka, Japan	150 km	Dry and wet deposition Resuspension	-	10 times decrease	-	Yamauchi et al. (2012)

^a Changes in electrical properties were obtained by comparing maximum values for observations with normal levels of observation locations.

After the Chernobyl accident, abnormal lightning activity was observed at contaminated radioactive sites in Sweden (Israelsson and Knudsen, 1987). Lightning flashes of the contaminated sites significantly increased for a few months after the accident. The average lightning frequency of the contaminated sites was up to five times higher than that of low-radioactivity sites. Israelsson and Knudsen (1987) suggested that the observations of lightning activity enhancement may be attributed to ionization caused by radioactivity.

4. Conclusions

In case of a nuclear plant accident, accurate prediction of radioactivity transport is crucial in evaluating environmental and health risks caused by radioactive contamination. Electrostatic interactions can be important in simulating various atmospheric processes, but they are typically neglected in modeling studies of radioactivity transport. This study has shown that radioactivity can induce surface charge accumulation of radioactive particles and background aerosols, significantly influence the particle charge distribution in open air, and change the electrical properties of the local atmosphere. Based on the charging behavior of radioactive particles and the expected influence of radioactivity-induced charge on particle interactions with other particles, raindrops, and other environmental surfaces, electrostatic interactions should be considered in predictive studies of short- and long-range radioactivity transport.

ACKNOWLEDGMENTS. This work was supported by the Defense Threat Reduction Agency under grant number DTRA1-08-10-BRCWMD-BAA. The manuscript has been co-authored by UT-Battelle, LLC, under Contract No. DEAC05-00OR22725 with the U.S. Department of Energy.

REFERENCES

- ApSimon, H., Wilson, J. J. N., Simms, K. L., 1989. Analysis of the dispersal and deposition of radionuclides from Chernobyl across Europe. *Proc. R. Soc. A* 425, 365-405.
- Bragg, W. H., Kleeman, R., 1905. XXXIX. On the α particles of radium, and their loss of range in passing through various atoms and molecules. *Philos. Mag.* 10, 318-340.
- Chesser, R.K., Bondarkov, M., Baker, R.J., Wickliffe, J.K., Rodgers, B.E., 2004. Reconstruction of radioactive plume characteristics along Chernobyl's Western trace. *J. Environ. Radioactivity* 71, 147-157.
- Clement, C. F., Harrison, R. G., 1992. The charging of radioactive aerosols. *J. Aerosol Sci.* 23, 481-504.
- Cooper, D. W., Reist, P. C., 1973. Neutralizing charged aerosols with radioactive sources. *J. Colloid Interface Sci.* 45, 17-26.
- Cuddihy, R. G., Finch, G. L., Newton, G. J., Hahn, F. F., Mewhinney, J. A., Rothenberg, S. J., Powers, D. A., 1989. Characteristics of radioactive particles released from the Chernobyl nuclear reactor. *Environ. Sci. Technol.* 23, 89-95.
- Devell, L., Tovedal, H., Bergström, U., Appelgren, A. I., Chyssler, J., Andersson, L., 1986. Initial observations of fallout from the reactor accident at Chernobyl. *Nature* 321, 192-193.
- Gensdarmes, F., Boulaud, D., Renoux, A., 2001. Electrical charging of radioactive aerosols—comparison of the Clement-Harrison models with new experiments. *J. Aerosol Sci.* 32, 1437-1458.
- Greenfield, S. M., 1956. Ionization of radioactive particles in the free air. *J. Geophys. Res.* 61, 27-33.

416 Greenfield, S. M., 1957. Rain scavenging of radioactive particulate matter from the atmosphere.
 417 J. Atmos. Sci. 14, 115-125.

418 Harrison, R. G., Carslaw, K. S., 2003. Ion-aerosol-cloud processes in the lower atmosphere. Rev.
 419 Geophys. 41, 1012.

420 Hoppel, W. A., Frick, G. M., 1986. Ion-aerosol attachment coefficients and the steady-state
 421 charge distribution on aerosols in a bipolar ion environment. Aerosol Sci. Technol. 5, 1-
 422 21.

423 Huizenga, H., Storchi, P. R. M., 1989. Numerical calculation of energy deposition by broad
 424 high-energy electron beams. Phys. Med. Biol. 34, 1371-1396.

425 International Commission on Radiation Units and Measurements (ICRU), 1984. Stopping
 426 powers for electrons and positrons, ICRU Report 37, ICRU, Bethesda, M. D.

427 Israelsson, S., Knudsen, E., 1986. Effects of radioactive fallout from a nuclear power plant
 428 accident on electrical parameters. J. Geophys. Res. 91, 11909-11910.

429 Israelsson, S., Schütte, T., Pislér, E., Lundquist, S., 1987. Increased occurrence of lightning
 430 flashes in Sweden during 1986. J. Geophys. Res. 92, 10996-10998.

431 Joffre, H., 1963. Les Problèmes physiques de la radioprotection; Institut national des sciences et
 432 techniques nucléaires, Paris, France.

433 Kaneyasu, N., Ohashi, H., Suzuki, F., Okuda, T., Ikemori, F., 2012. Sulfate aerosol as a potential
 434 transport medium of radiocesium from the Fukushima nuclear accident. Environ. Sci.
 435 Technol. 46, 5720-5726.

436 Kauppinen, E. I., Hillamo, R. E., Aaltonen, S. H., Sinkko, K. T. S., 1986. Radioactivity size
 437 distributions of ambient aerosols in Helsinki, Finland, during May 1986 after the
 438 Chernobyl accident: preliminary report. Environ. Sci. Technol. 20, 1257-1259.

439 Katz, L, Penfold, A. S., 1952. Range-energy relations for electrons and the determination of
 440 beta-ray end-point energies by absorption. *Rev. Mod. Phys.* 24, 28-44.

441 Kim, Y.-H., Yiacoumi, S., Lee, I., McFarlane, J., Tsouris, C., 2014. Influence of radioactivity on
 442 surface charging and aggregation kinetics of particles in the atmosphere. *Environ. Sci.*
 443 *Technol.* 48, 182-189.

444 Kweon, H., Yiacoumi, S., Lee, I., McFarlane, J., Tsouris, C., 2013. Influence of surface potential
 445 on the adhesive force of radioactive gold surfaces. *Langmuir* 29, 11876-11883.

446 Lee, H., Feichter, J., 1995. An intercomparison of wet precipitation scavenging schemes and the
 447 emission rates of ^{222}Rn for the simulation of global transport and deposition of ^{210}Pb . *J.*
 448 *Geophys. Res.-Atmos.* 100, 23253-23270.

449 Harris, D. L., 1955. Effects of radioactive debris from nuclear explosions on the electrical
 450 conductivity of the lower atmosphere. *J. Geophys. Res.* 60, 45-52.

451 Lujanienė, G., Aninkevičius, V., Lujanas, V., 2009. Artificial radionuclides in the atmosphere
 452 over Lithuania. *J. Environ. Radioactiv.*, 100, 108-119.

453 Mantel, J., 1972. The beta ray spectrum and the average beta energy of several isotopes of
 454 interest in medicine and biology. *Int. J. Appl. Radiat. Is.* 23, 407-413.

455 Marmier, P., Sheldon, E., 1969. *Physics of Nuclei and Particles (Volume I)*, Academic Press,
 456 New York.

457 Martin, J. E., 2000. *Physics for Radiation Protection*, John Wiley & Sons, New York.

458 Matsoukas, T., 1997. The coagulation rate of charged aerosols in ionized gases. *J. Colloid*
 459 *Interface Sci.* 187, 474-483.

460 Ooe, H., Seki, R., Ikeda, N., 1988. Particle-size distribution of fission products in airborne dust
 461 collected at Tsukuba from April to June 1986. *J. Environ. Radioactivity* 6, 219-223.

462 Oron, A., Seinfeld, J. H., 1989a. The dynamic behavior of charged aerosols: II. Numerical
 463 solution by the sectional method. *J. Colloid Interface Sci.* 133, 66-79.
 464 Oron, A., Seinfeld, J. H., 1989b. The dynamic behavior of charged aerosols: III. Simultaneous
 465 charging and coagulation. *J. Colloid Interface Sci.* 133, 80-90.
 466 Parache, V., Pourcelot, L., Roussel-Debet, S., Orjollet, D., Leblanc, F., Soria, C., Gurriaran, R.,
 467 Renaud, P., Masson, O., 2011. Transfer of ^{131}I from Fukushima to the vegetation and
 468 milk in France. *Environ. Sci. Technol.* 45, 9998-10003.
 469 Pöllänen, R., Valkama, I., Toivonen, H., 1997. Transport of radioactive particles from the
 470 Chernobyl accident. *Atmos. Environ.* 31, 3575-3590.
 471 Reed, L. D., Jordan, H., Gieseke, J. A., 1977. Charging of radioactive aerosols. *J. Aerosol Sci.* 8,
 472 457-463.
 473 Rehfeld, S., Heimann, M., 1995. Three dimensional atmospheric transport simulation of the
 474 radioactive tracers ^{210}Pb , ^7Be , ^{10}Be , and ^{90}Sr . *J. Geophys. Res.-Atmos.* 100, 26141-
 475 26161.
 476 Renard, J. B., Tripathi, S. N., Michael, M., Rawal, A., Berthet, G., Fullekrug, M., Harrison, R. G.,
 477 Robert, C., Tagger, M., Gaubicher, B., 2013. In situ detection of electrified aerosols in
 478 the upper troposphere and in the stratosphere. *Atmos. Chem. Phys.* 13, 11187–11194.
 479 Retalis, D., Pitta, A., 1989. Effects on electrical parameters at Athens, Greece by radioactive
 480 fallout from a nuclear power plant accident. *J. Geophys. Res.* 94, 13093-13097.
 481 Seinfeld, J. H., Pandis, S.N., 2006. *Atmospheric Chemistry and Physics: From Air Pollution to*
 482 *Climate Change*, John Wiley and Sons, New Jersey.
 483 Seltzer, S. M., Berger, M. J., 1982. Evaluation of the collision stopping power of elements and
 484 compounds for electrons and positrons. *Int. J. Appl. Radiat. Is.* 31, 1189-1218.

485 Shaw, G. E., 1986. Size distribution of atmospheric aerosols at Ross Island. *Antarct. J. U.S.* 21,
486 241-242.

487 Tabata, T., Ito, R., Okabe, S., 1972. Generalized semiempirical equations for the extrapolated
488 range of electrons. *Nucl. Instrum. Methods* 103, 85-91.

489 Takeda, M., Yamauchi, M., Makino, M., Owada, T., 2011. Initial effect of the Fukushima
490 accident on atmospheric electricity. *Geophys. Res. Lett.*, 38, L15811.

491 Tomé, W. A., Palta, J. R., 1998. On the calculation of mean restricted collision stopping powers.
492 *Med. Phys.* 25, 758-772.

493 Trier, A., 1997. Submicron particles in an urban atmosphere: a study of optical size
494 distributions—I. *Atmos. Environ.* 31, 909-914.

495 Tripathi, S., Harrison R. G., 2001. Scavenging of electrified radioactive aerosol. *Atmos. Environ.*
496 35, 5817-5821.

497 Walker, M. E., McFarlane, J., Glasgow, D. C., Chung, E., Taboada-Serrano, P., Yiacoumi, S.,
498 Tsouris, C., 2010. Influence of radioactivity on surface interaction forces. *J. Colloid*
499 *Interface Sci.* 350, 595-598.

500 Whyte, G., 1963. Energy per ion pair for charged particles in gases. *Radiat. Res.*, 18, 265-271.

501 Yair, Y., Levin, Z., 1995. Charging of polydispersed aerosol particles by attachment of
502 atmospheric ions. *J. Geophys. Res.-Atmos.* 94, 13085-13091.

503 Yamauchi, M., Takeda, M., Makino, M., Owada, T., Miyagi, I., 2012. Settlement process of
504 radioactive dust to the ground inferred from the atmospheric electric field measurement.
505 *Ann. Geophys.* 30, 49–56.

506 Yeh, H. C., Newton, G. J., Raabe, O. G., Boor, D. R., 1976. Self-charging of ^{198}Au -labeled
 507 monodisperse gold aerosols studied with a miniature electrical mobility spectrometer. J.
 508 Aerosol Sci. 7, 245-253.

509 Yoshenko, V.I., Kashparov, V.A., Protsak, V.P., Lundin, S.M., Levchuk, S.E., Kadygrib, A.M.,
 510 Zvarich, S.I., Khomutinin, X.V., Maloshtan, I.M., Lanshin, V.P., Kovtun, M.V.,
 511 Tschierisch, J., 2006a. Resuspension and redistribution of radionucleotides during
 512 grassland and forest fires in the Chernobyl exclusion zone. Part I: fire experiments. J.
 513 Environ. Radioactivity 86, 143-163.

514 Yoshenko, V.I. Kashparov, V.A., Levchuk, S.E., Glukhovskiy, A.S., Khomutni, Y.V., Protsak,
 515 V.P., Lundin, S.M., Tschiersch, J., 2006b. Resuspension and redistribution of
 516 radionucleotides during grassland and forest fires in the Chernobyl exclusion zone. Part
 517 II: modeling the transport process. J. Environ. Radioactivity 87, 260-278.