Influence of Radioactive Decay on Atmospheric Transport of Radioactive Particles

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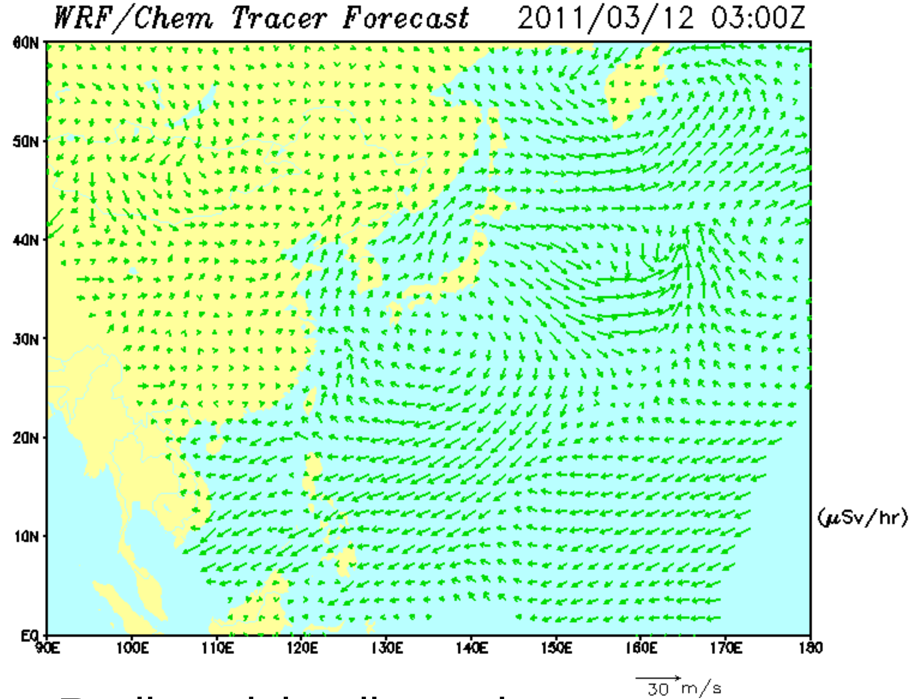
Contents

Release of Radionuclides by Nuclear Events

- Radionuclides can be intentionally or accidentally released from radioactive sources (e.g., radiological dispersal devices or nuclear power plant accidents)

  A recent case: Fukushima nuclear plant accident (2011)

< Initial radioactive plume >

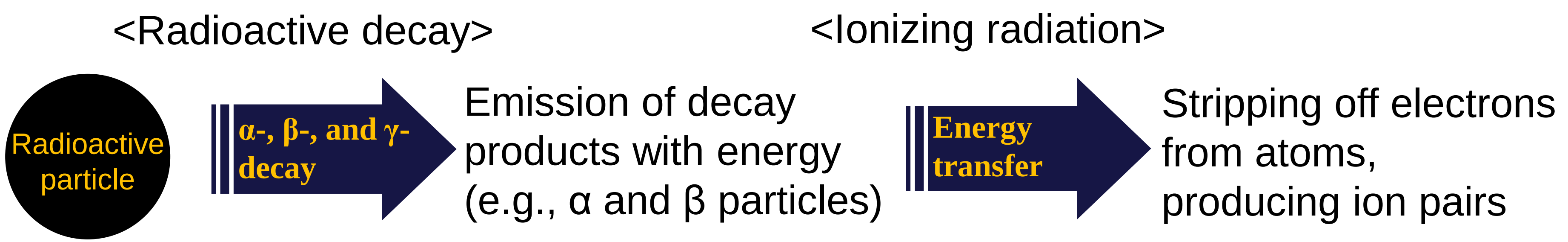


< Radioactivity dispersion>

A large amount of short-lived and long-lived radionuclides can be released into the environment by nuclear events

Harmful Effects of Radiation Exposure

❖ Radionuclides can cause severe damage to the human body through ionizing radiation



❖ Ionized atoms function differently in the human body

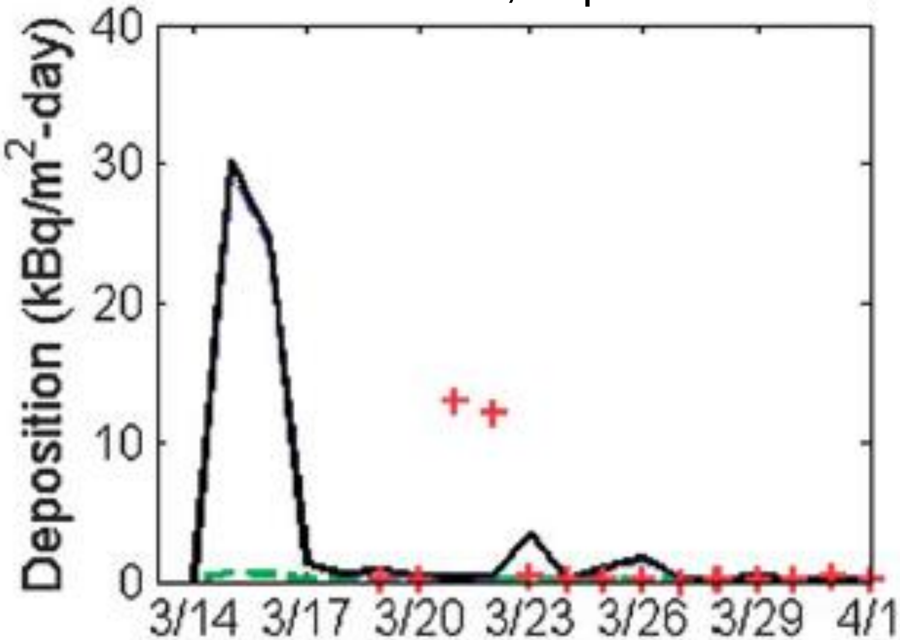
- ✓ Cell function disruption, breaking and mutating DNA chains
- ✓ Moderate to high-level radiation exposure of humans increases the cancer risk in most organs [2]
- ✓ When monitoring 94,000 atomic bomb survivors for 5 - 45 years, 5 - 8 % of cancer deaths resulted from radiation exposure [2]

Accurately predicting the transport of radionuclides from the beginning of nuclear events is important for risk evaluation

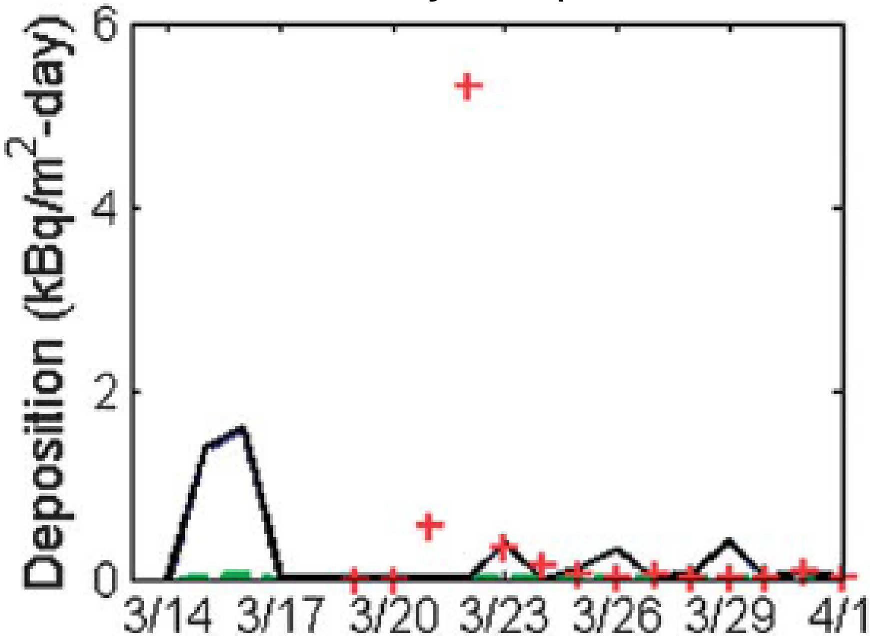
Theory vs. Observations

✓ Fukushima nuclear plant accident[3]

< Ibaraki, Japan >



< Tokyo, Japan >

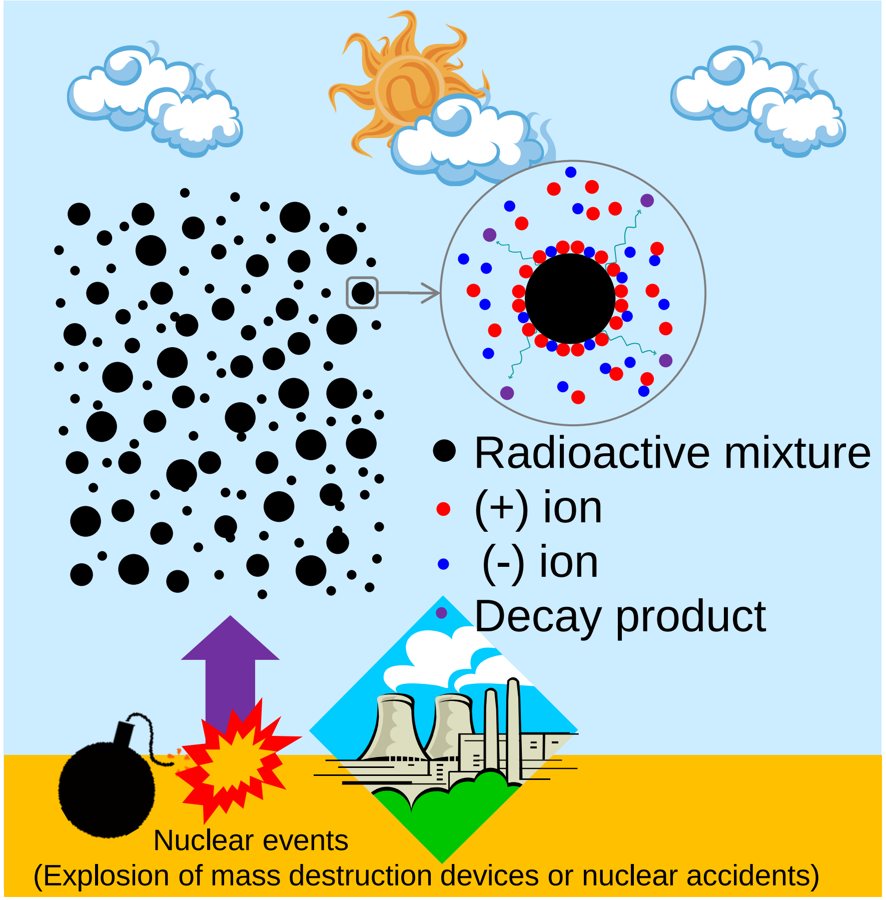


Black solid line: Total deposition (modeling); Green dash line: Dry deposition (modeling); Symbol: Total deposition (measurements)

It is still necessary to improve predictive transport models of radioactivity

Transport Modeling of Radioactivity

❖ Particles containing radionuclides do not behave like nonradioactive particles



- ✓ In case studies of nuclear reactor accidents and explosion of mass destruction devices, **nonradioactive particles are typically assumed**
- ✓ Nonradioactive particles normally show negligible electrostatic particle interactions
- ✓ However, **radioactive particles can be charged during radioactive decay**
- ✓ Particle charging can affect the microphysical behavior of airborne particles

Objectives: Investigate the charging of radioactive particles, involve the charging effects in transport modeling of radioactivity, and examine the charge effects on the transport of radioactivity

Modeling Radioactivity-induced Charging

❖ Charge Balance Model:[4]

Accumulation of radioactivity-induced charges J on a particle surface over time t
dJ/dt

=

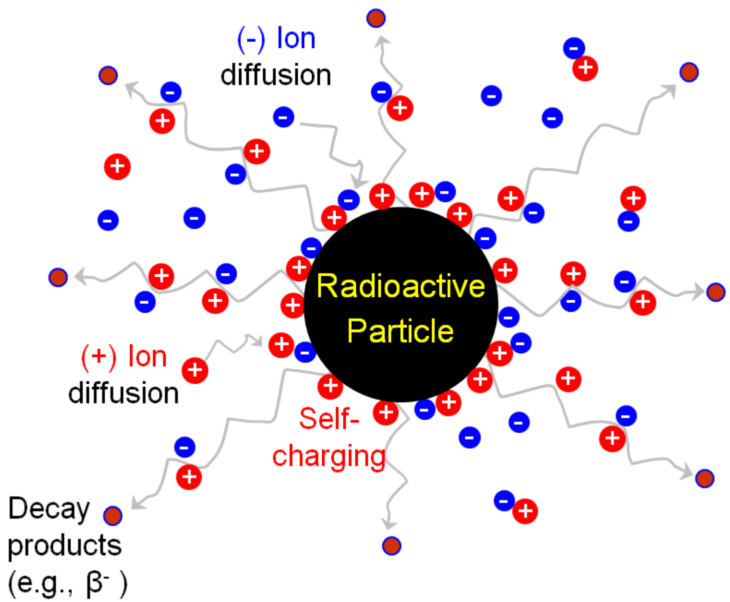
Production of positive charges through diffusion of positive ions (*Diffusion charging*):
$\beta_+ \times n_+ = f$ (**particle size, mobility and concentration of + ions**)

-

Loss of positive charges through diffusion of negative ions (*Diffusion charging*):
$\beta_- \times n_- = f$ (**particle size, mobility and concentration of - ions**)

+

Production of positive charges through α or β^- particle emission (*Self-charging*):
$m \times \eta = f$ (**self-charging coefficient m, radioactivity η**)



$\beta_\pm$ Ion-particle attachment coefficient
$n_\pm$ Ion concentration

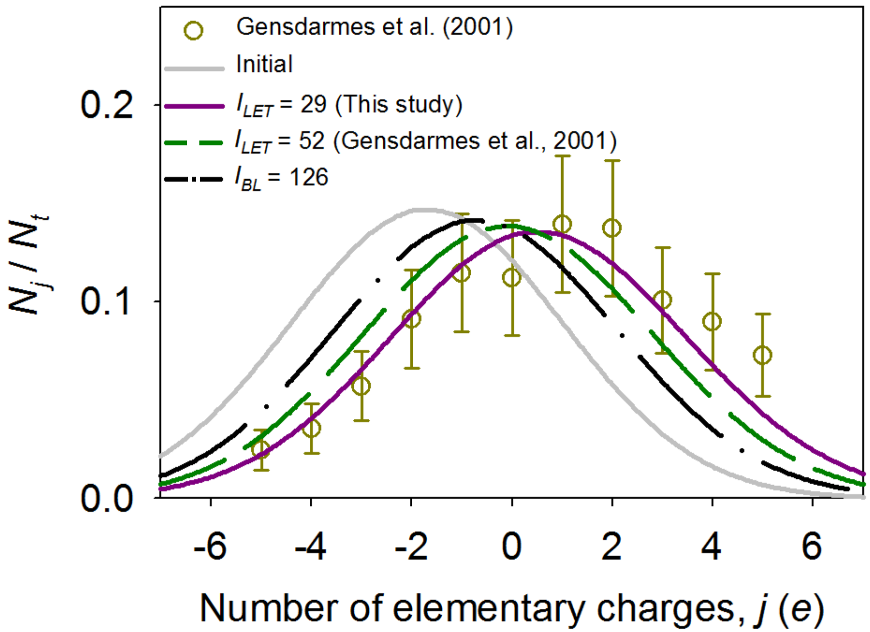
The charge balance equation is based on charging mechanisms, including diffusion charging and self-charging rates

 5. Kim et al., *J. Environ. Radioactivity*, **2015**; 6. Gensdarmes et al., *J. Aerosol. Sci.*, **2001**

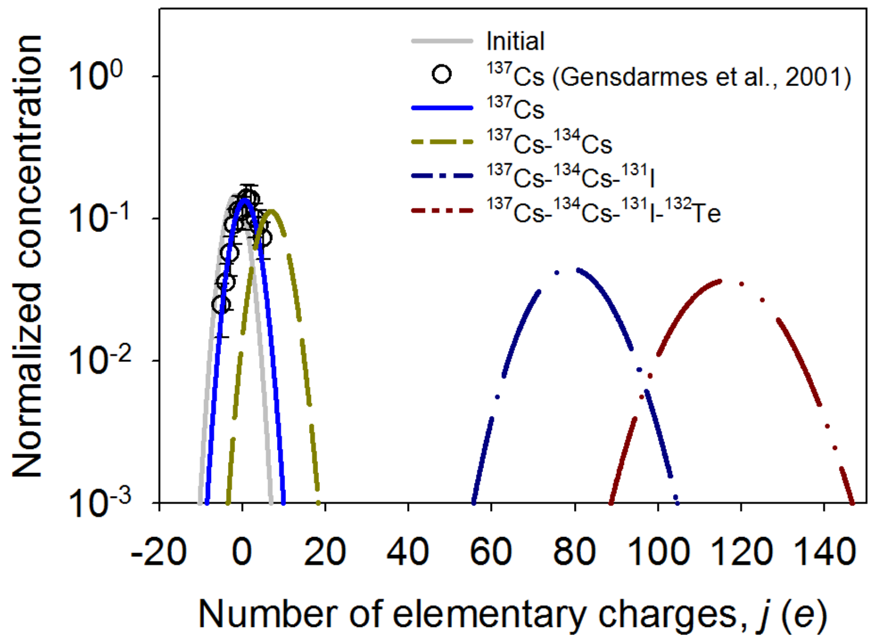
Charging of Radioactive Particles in Tubes of 6 mm Diameter

❖ Charging of ^{137}Cs aerosols (Size = 0.82 μm; Radioactivity = 1.28x10^{-2} Bq; Charging time = 1,060s)[5]

- Single-radionuclide aerosols (Aerosols containing single radionuclides)



- Multicomponent radionuclide aerosols (Aerosols containing various radionuclides)



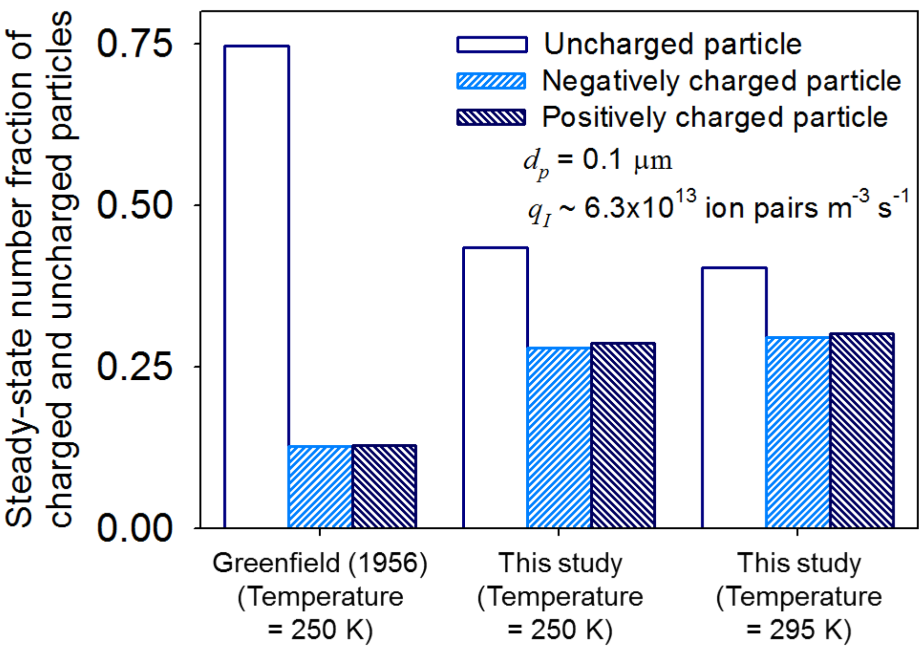
- Results obtained using the calculated ionization rate coefficient are in good agreement with measurements of Gensdarmes et al.[6]
- Short-lived radionuclides can significantly affect the charging of multicomponent radionuclide aerosols

Radioactive particles can be highly charged

Can Radioactive Particles Be Charged in Open Air?

❖ A scenario of Greenfield[7]: radioactive particles are generated at 6 km altitude

Previous suggestion: most radioactive particles are neutralized due to beta radiation which generates 6.3×10^{13} ion pairs m^{-3} s^{-1} [7]



- ✓ Yes, many radioactive particles can be charged even in open air in contrast to the previous suggestion[5]
- ✓ The discrepancies resulted from the difference in simulating diffusion charging of radioactive particles
- ✓ Electrostatic interaction can occur among radioactive particles, which can influence the microphysical behavior of the particles (e.g., size growth by coagulation)

Investigations into electrostatic interactions of radioactive particles are necessary to better understand the microphysical behavior of particles

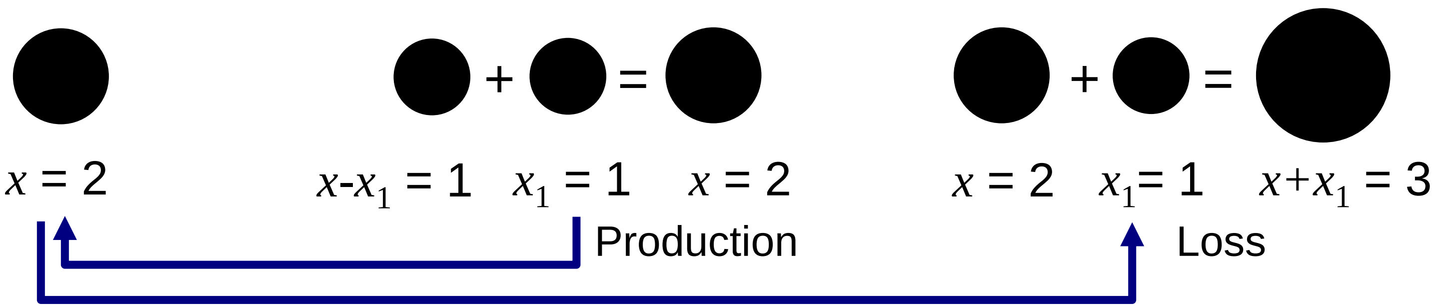
4. Kim et al., *Atmos. Chem. Phy.*, **2016**

Modeling Particle Aggregation and Charging

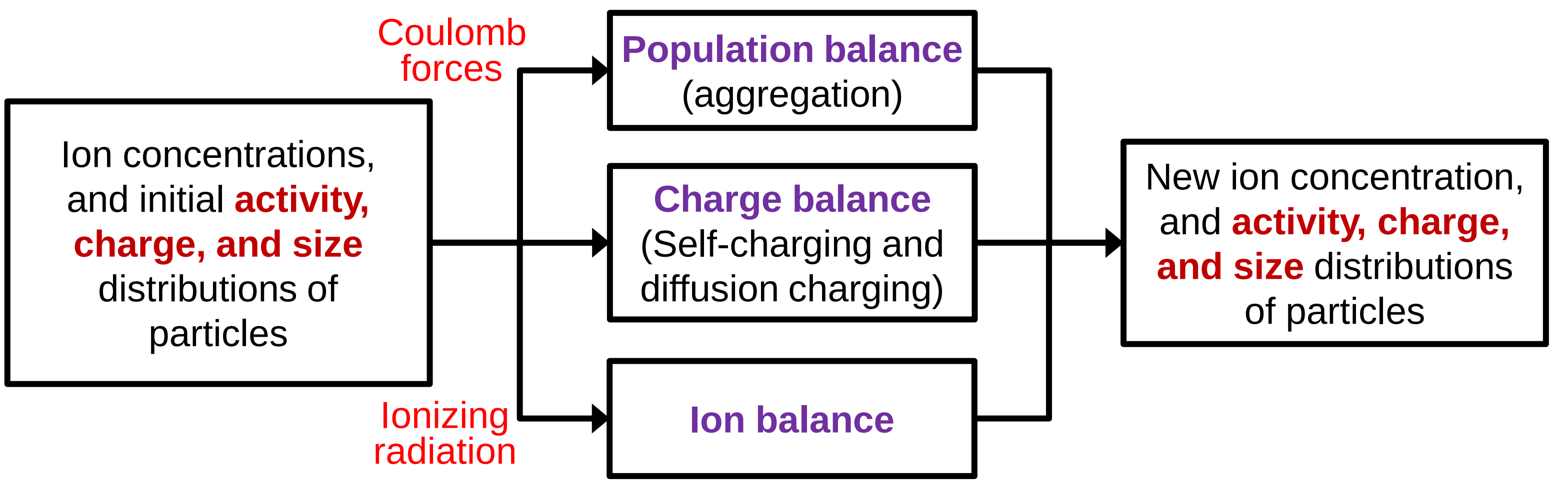
❖ Population Balance Model (PBM)

Changes in number density n of particles of size x over time	=	Rate of production of particles of size x by coagulation of smaller particles	+	Rate of loss of particles of size x by coagulation of particle x with any size particles

e.g.,

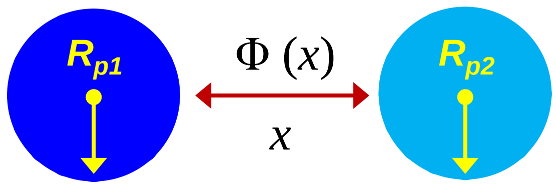


❖ How to add charging effects on aerosol microphysics? [4]



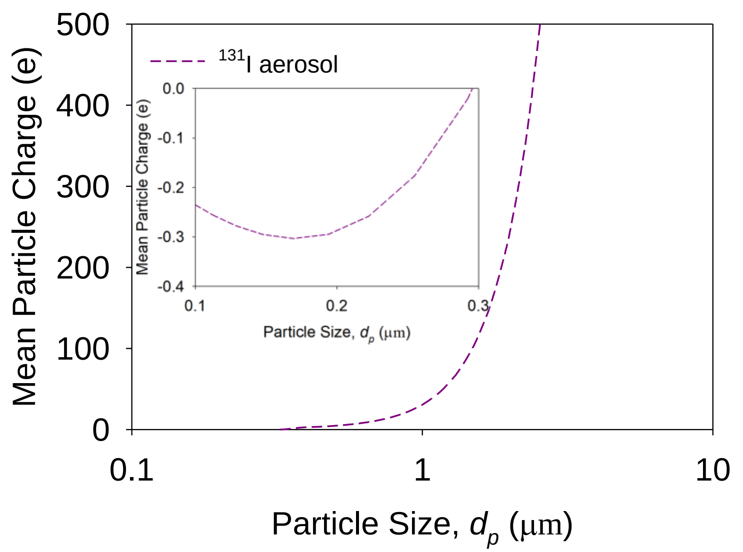
Effects of Radioactivity-Induced Charging on Particle Interactions

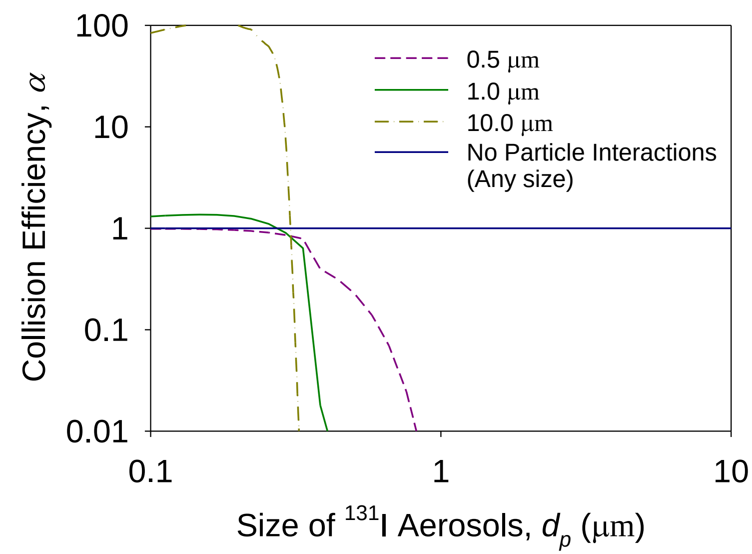
❖ Highly Radioactive Particles vs Highly Radioactive Particles in Highly Ionized Air



$$\alpha = \left(R_{p1} + R_{p2} \int_{R_{p1}+R_{p2}}^{\infty} \frac{1}{x^2} \exp\left(\frac{\Phi(x)}{kT} \right) dx \right)^{-1} \quad 8$$

$\alpha > 1$: Particle-Particle Attraction $\alpha = 1$: No Net Effects of Particle Interactions $\alpha < 1$: Particle-Particle Repulsion





- Strong electrostatic attractive forces can be produced between the aerosols

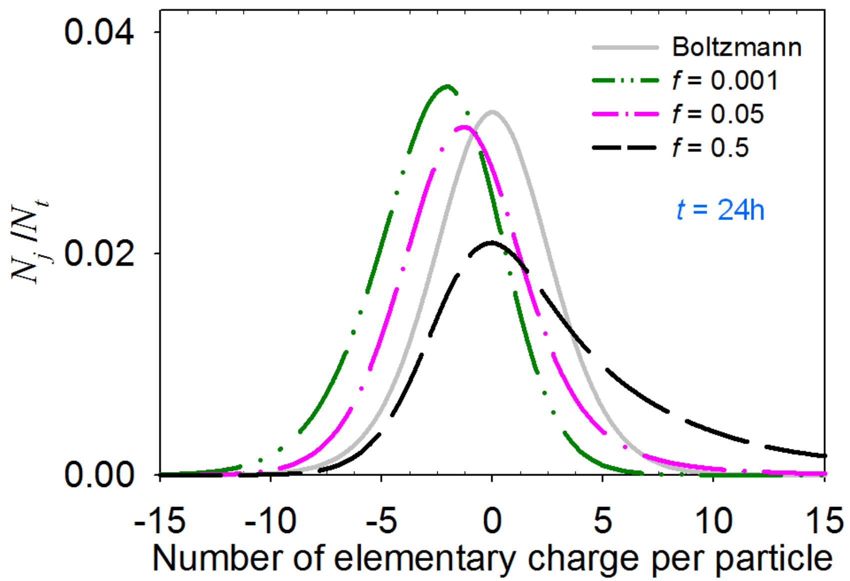
Radioactivity-induced charging impacts interactions of highly radioactive particles

Coagulation of Multicomponent Radionuclide Particles [9]

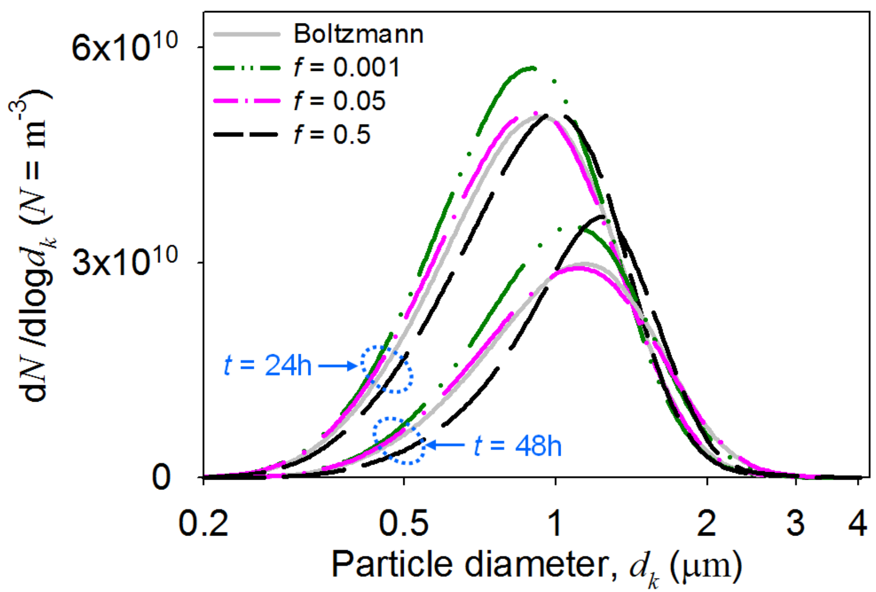
- ✓ Initial size distribution: Lognormal distribution (d_g = 0.5 µm; σ_g = 1.5; $N_{total} = 10^{11}$ m^{-3})
- ✓ Initial particle composition: $(NH_4)_2SO_4$-^{132}Te
- ✓ Final particle composition: $(NH_4)_2SO_4$-^{132}Te-^{132}I-^{132}Xe

f: Mole fraction of radioactive components

< Charge distribution >



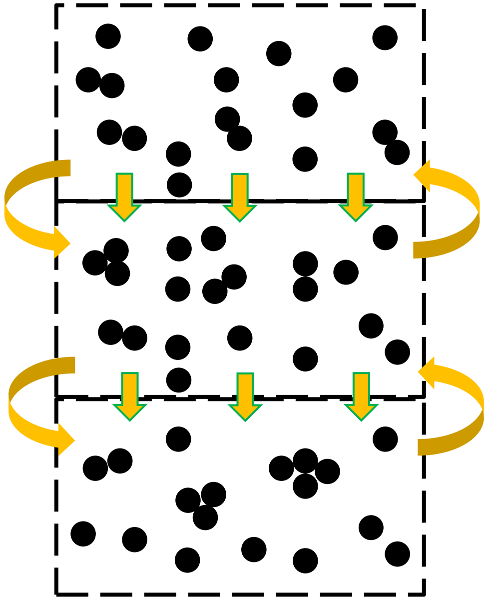
< Size distribution >

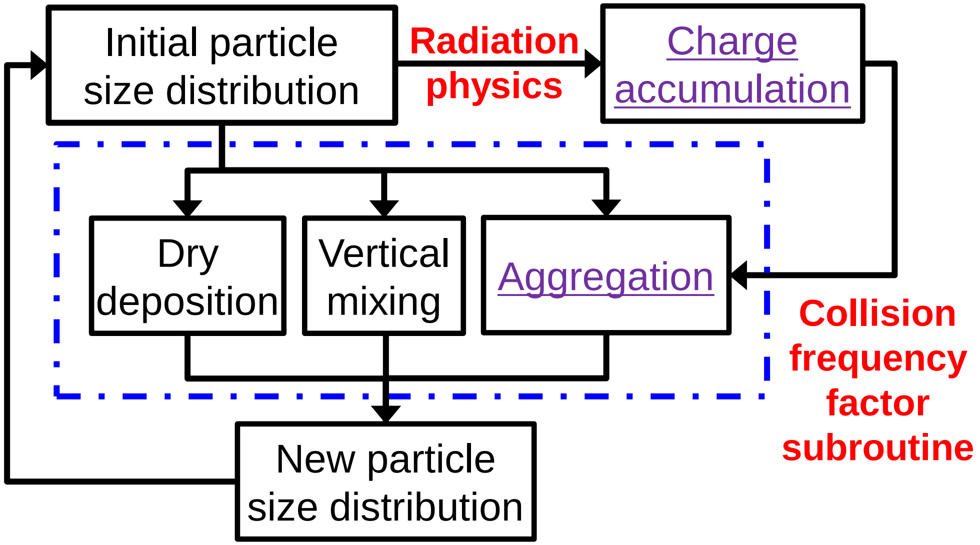


The charging of radioactive particles can influence the coagulation of particles

Radioactive Particle Transport Modeling [10]

✓ Example: a simple vertical transport model of radioactive particles including aggregation, charge accumulation, dry deposition, and vertical mixing



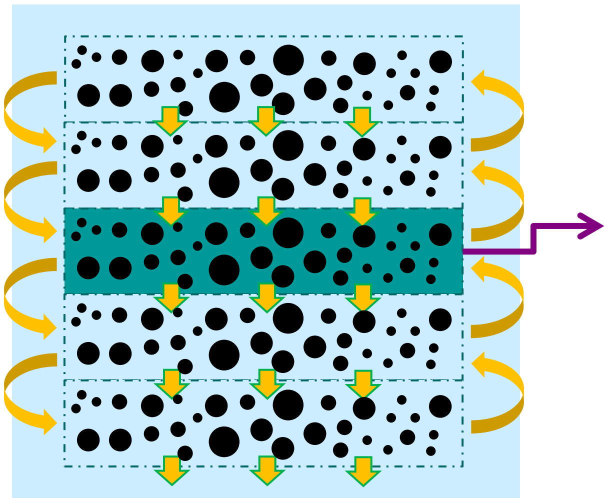


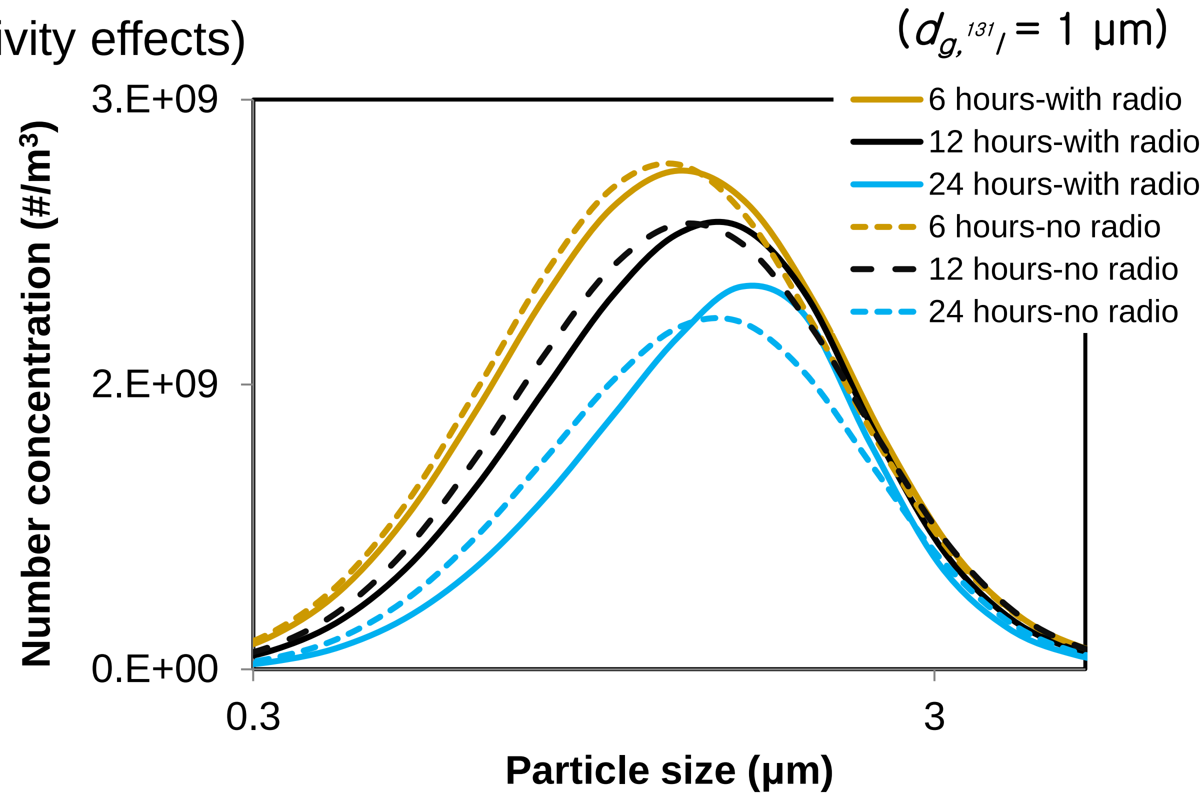
✓ Scenarios for Radiological Debris Release

- ^{131}I particles; 5 vertical layers with 200 m height; Initially log-normal size distribution; Decay rates are constant during simulation; Charging of radioactive particles quickly occurs and reaches steady state rapidly; No humidity effects

Vertical Transport of 131I Particles (600 m altitude)

✓ ^{131}I aerosol time dependent distributions (with and without radioactivity effects)





- With a large initial particle size, the radioactive decay of ^{131}I has an appreciable effect on the particle charging and evolution of the particle size distribution
- Beyond a certain particle size, aggregation is suppressed due to strong repulsive forces generated by the self-charging mechanism of ^{131}I

Conclusions

✓ The **charging and aggregation kinetics** of particles containing various radionuclides have been investigated

- Particles containing short-lived and long-lived radionuclides can be charged due to self-charging and diffusion charging
- Radioactivity-induced charge can generate repulsive and attractive electrostatic forces, which can suppress or facilitate aggregation of radioactive particles
- The dry deposition rates of radioactive particles can be different from those of nonradioactive particles

Radioactive particles behave differently from nonradioactive particles

Radioactivity-induced charging must be taken into consideration in the 3D transport modeling of radioactivity for more accurate predictions

Acknowledgements

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Thank you very much for your attention!