SAND2017-7063C

Quantifying Electrostatic Resuspension of Radionuclides from Surface Contamination

Shaun Marshall[1], Charles Potter[2], David Medich[1]

[1]Worcester Polytechnic Institute, Worcester, MA 01609
[2]Sandia National Laboratories, Albuquerque, NM 87185

62nd Annual Meeting of the Health Physics Society
Raleigh Convention Center, Raleigh, NC – July 9, 2017

* Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525.

Outline

I. Introduction
- Inhalation Dosimetry Risk Assessment
- Summary of Resuspension Studies

II. Physical Modeling of Resuspension Factor (S_f)
- Catenary Kinetics Model
- S_f Weighted-Uncertainty Reassessment
- Model Discussion and Recommendations

III. Experimental Pilot Study Quantify Resuspension
- Indoor Resuspension Chamber
- Neutron Activation Analysis
- Spectroscopic Results and Resuspension Factor

IV. Conclusions and Future Work

I. Introduction

Inhalation Exposure Pathway

- Environmental exposure to airborne radionuclides following surface release
- Internal dose through inhalation dependent upon available pathways
- Resuspension as a mechanical rate constant for particulate transportation

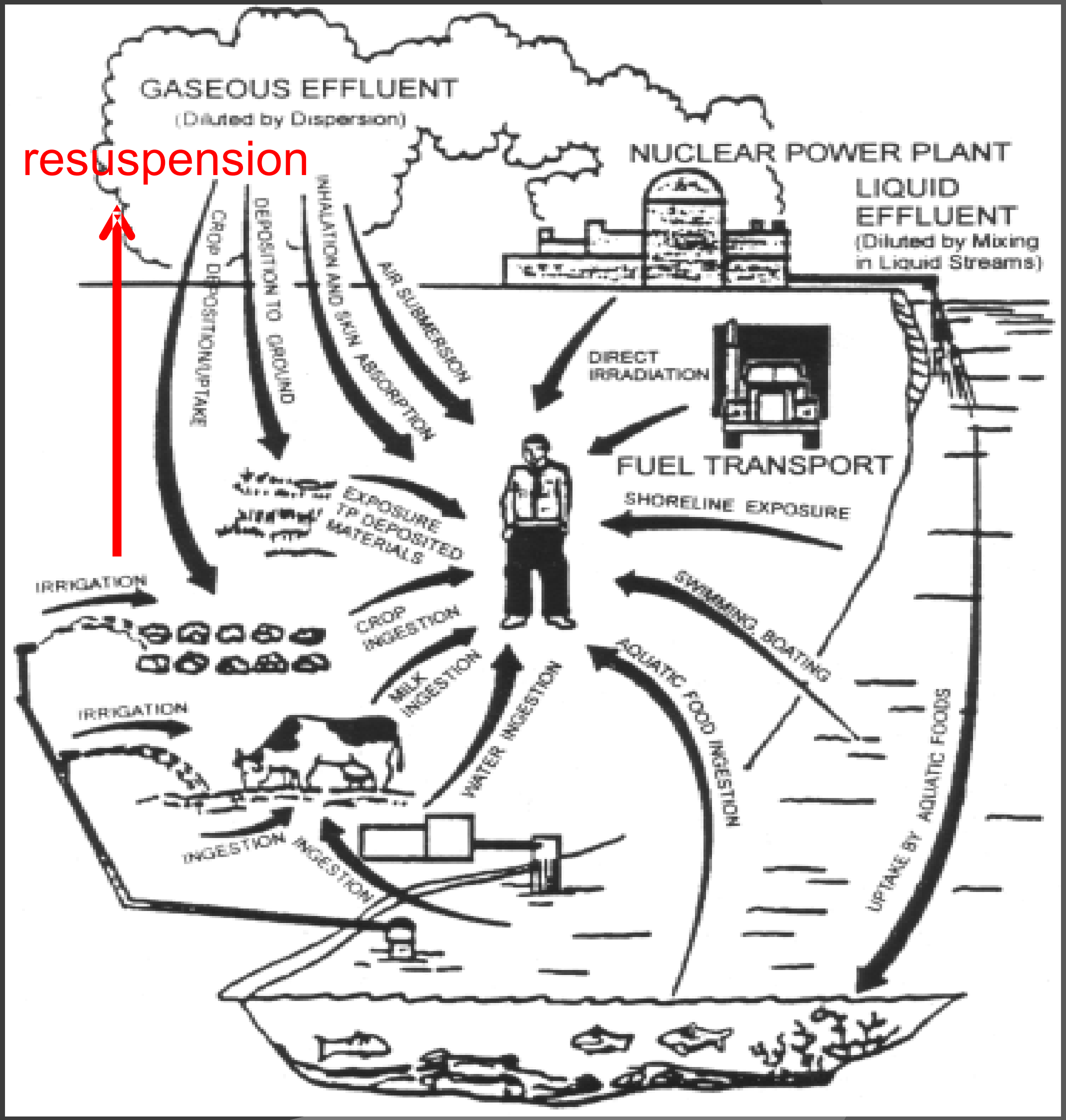

Figure 1: Potential exposure pathways in the event of a radionuclide release. (NRC, 2016)

Dose Prediction Model

- Dose due to inhalation of resuspended radionuclide*:

$$D_{inh} = C_{D,inh} \times \overline{f_B} \times KP,$$
$$KP = \int_{TP} Dp \times e^{-\lambda t} \times S_f(t)\, dt$$

Table 1: Formulaic breakdown of dose from inhalation of radionuclides.

Term	Description (units)
$C_{D,inh}$	inhalation committed dose coefficient (Sv Bq^{-1})
$\overline{f_B}$	Activity-averaged human breathing rate (~0.92 m^3 h^{-1})
KP	resuspension parameter, which considers airborne concentration during time phase TP following deposition (Bq s m^{-3})
Dp	initial areal deposition (Bq m^{-2})
λ	radionuclide decay constant (s^{-1})
$S_f(t)$	empirical resuspension factor (m^{-1})

* (SNL, 2015); simplified

An Improved Model for Prediction of Resuspension

Figure 2: Resuspension factors calculated from historical dataset of realistic and synthetic particle dispersion experiments. (Maxwell and Anspaugh, 2011)

Resuspension Factor Development

- Current FRMAC model*:

$$S_f(t) = 10^{-9} + 7 \times 10^{-9} e^{-0.002t} + 5 \times 10^{-6} e^{-0.07t}$$

- Motivation for reassessment of resuspension factor
  - Sustained availability for inhalation uptake
  - Recently argued* to be 10-100x greater than original estimates (ppm)
  - No directly applicable resuspension data for Am-241
  - Need validation and consensus of applicable models
    - Differentiated by nuclide or environment
    - Differentiated by synthetic and realistic updraft

* Maxwell and Anspaugh, 2011

II. Physical Modeling of Resuspension Factor (S_f)

S_f Reassessment Methodology

- Resolve early timeframe of historic datasets
  - Average and standard deviation S_f bins of width τ_b
  - ~3-4 days to avoid undersampling first process (10-100 d)

- Fit averaged data to current semi-empirical model
  - S_f-data weighted by uncertainty in data for each time period: $w_i = \sigma_{S_f}^{-1}$
  - Applied fixed ($X_0 = 10^{-9}$) and unfixed ($X_0 \neq 0$) long-term steady-state constant

- Extract analytical constants from regression coefficients
  - Reconstruction of resuspension factor following “ideal” surface release

S_f Reassessment Results

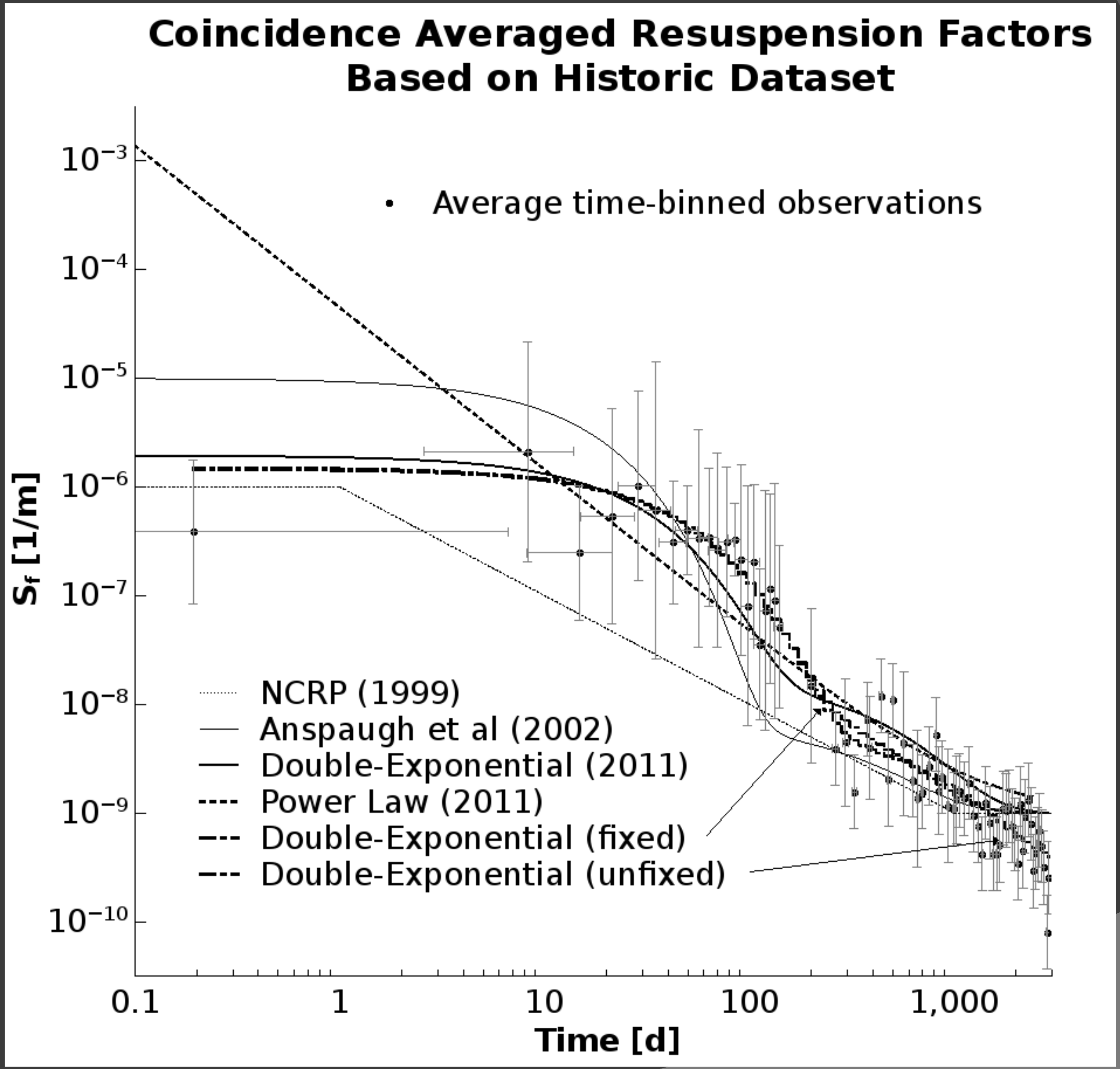

Figure 3: Averaged resuspension factor observations, overlaid with recent resuspension factor models including this work (indicated with arrows).

Resuspension Catenary Kinetics

- Fractional population of air (A), surface (S), ground (G) compartments of catenary system behave similarly as:

$$\boxed{A} \underset{k_{A\leftarrow S}}{\overset{k_{A\rightarrow S}}{\rightleftarrows}} \boxed{S} \underset{k_{S\leftarrow G}}{\overset{k_{S\rightarrow G}}{\rightleftarrows}} \boxed{G}\,; \qquad X_A(t) = X_0 + X_1 e^{-\omega_1 t} + X_2 e^{-\omega_2 t}$$

Table 2: Kinetic rate constant (s^{-1}) definitions for three-compartment catenary model.

Term	Description
$k_{A\rightarrow S}$	Deposition or settling rate: gravitational (proportional to $v_{terminal}$)
$k_{A\leftarrow S}$	Resuspension rate: electrostatic upward drift into atmosphere
$k_{S\rightarrow G}$	Migration rate: based on ground porosity and colloidal properties
$k_{S\leftarrow G}$	(Bio)turbation rate: mixing by decontamination, biota activity or long-term geological movement

- Since $S_f(t) \in [0,1]$, $S_f(t) \cong X_A$
  - $\therefore$ S_f contains initially AND re-suspended particles!

S_f Reassessment Results

Table 3: Best-fit linear regression parameters in log-space of averaged observations with varied constant offset.

Term	Fixed	Unfixed
X_0	1.00×10^{-9}	3.307×10^{-10}
X_1	1.371×10^{-8}	6.118×10^{-9}
X_2	1.501×10^{-6}	1.450×10^{-6}
ω_1	-0.00346	-0.00157
ω_2	-0.0264	-0.0253

Table 4: Initial fractional quantities and kinetic rate constants for unfixed three-compartment catenary model as determined by historic dataset

Fractional Quantity	
$X_A(0)$	1.456×10^{-6}
$X_S(0)$	0.9999984
Rate constants (d^{-1})	
$k_{A \rightarrow S}$	0.0253
$k_{A \leftarrow S}$	1.535×10^{-10}
$k_{S \rightarrow G}$	1.484×10^{-3}
$k_{S \leftarrow G}$	8.555×10^{-5}

S_f Reassessment Results

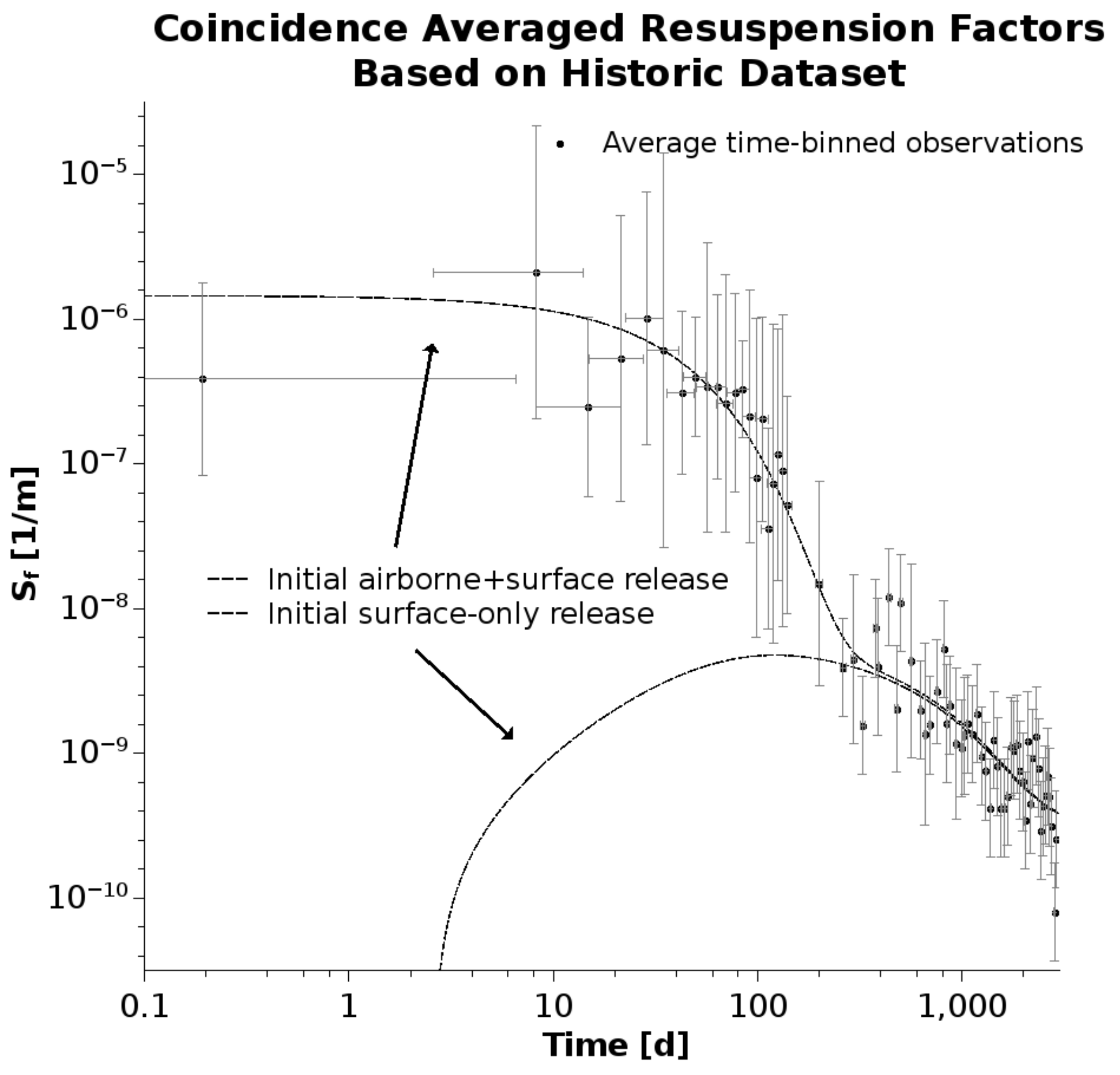

Figure 4: Averaged historic resuspension factor observations, overlaid with this work's suspension factor model and its equivalent for an ideal surface release.

III. Experimental Pilot Study to Quantify Resuspension

Air Sampling Chamber

- Release known mass m_0 of Eu_2O_3 particles (D_p=1-10 µm)
- Deposited at h=0 or h=2m, sampled at h=1m with low flowrate
- Filters changed by hour, day, week

Figure 4: Close-up of particulate deposits on concrete surface following air release.

Figure 5: Resuspension chamber with vacuum pump head.

Thermal Neutron Activation

- Used filters stored in envelopes, positioned against beamline
  - Blocked with solid water to increase neutron scatter and flux
  - Used gold foil flux monitor (~1-2x10^8 n cm^{-2} s^{-1})

Figure 6: 1MW D-D research reactor beamline at WPI.

Gamma Spectroscopy

- Activated samples placed directly on Ge detector

- Obtained histograms with GENIE*
  - Pulse Height Analysis
  - Gamma spectrum 1-2000 keV

- Reduced background with Cadmium shielding
  - Decreased minimum detectable activity (MDA)

Figure 7: Broad energy Li-drifted Ge detector system with Cd shielding at WPI.

* GENIE 2000, Canberra Industries, Inc., Meriden, CT

Spectroscopic Analysis and Assessment

Table 5: Mass of Eu_2O_3 content on air sampler filters following surface release via neutron activation analysis for each time-phase sample, and corresponding minimum detectable mass (MDM) and suspension factor (S_f) for each analysis. Irradiation time (τ) and proceeding delay (t_d) and detection times (Δt) are given for each trial.

Sample	τ [h]	t_d [h]	Δt [h]	MDM [g]	MDS_f [m^{-1}]	S_f [m^{-1}]
Day 1 #1	8	0.083	2	2.435×10^{-12}	7.738×10^{-13}	$< MDS_f$
Day 1 #2	8	0.083	24	9.258×10^{-14}	2.942×10^{-14}	$< MDS_f$
Day 1 #3	8	0.083	24	9.258×10^{-14}	2.942×10^{-14}	$< MDS_f$
Day 1 #4	8	0.083	24	9.258×10^{-14}	2.942×10^{-14}	$< MDS_f$
Day 1 #5	8	0.083	24	9.258×10^{-14}	2.942×10^{-14}	$< MDS_f$
Week 1 #1	2	0.5	2	8.146×10^{-12}	3.698×10^{-13}	$< MDS_f$
Week 1 #2	24	0.5	24	5.148×10^{-14}	2.337×10^{-15}	$< MDS_f$
Week 1 #3	24	0.5	24	5.148×10^{-14}	2.337×10^{-15}	$< MDS_f$
Week 1 #4	24	0.5	24	5.148×10^{-14}	2.337×10^{-15}	$< MDS_f$
Week 1 #5	24	0.5	24	5.148×10^{-14}	2.337×10^{-15}	$< MDS_f$
Biweek 1 avg	4	0.5	2			

Spectroscopic Analysis and Assessment

Table 6: Mass of Eu_2O_3 content on air sampler filters following air release via neutron activation analysis for each time-phase sample, and corresponding minimum detectable mass (MDM) and suspension factor (S_f) for each analysis. Irradiation time (τ) and proceeding delay (t_d) and detection times (Δt) are given for each trial.

Sample	τ [h]	t_d [h]	Δt [h]	MDM [g]	MDS_f [m^{-1}]	S_f [m^{-1}]
Hour 1 avg	2	0.1	2	7.907×10^{-12}	6.031×10^{-11}	6.599×10^{-9}
Hour 2 avg	2	0.1	2	7.907×10^{-12}	6.031×10^{-11}	1.344×10^{-9}
Hour 3 avg	2	0.1	2	7.907×10^{-12}	6.031×10^{-11}	9.778×10^{-10}

IV. Conclusions and Recommendations

Conclusions and Recommendations

- “Suspension factor” should be used in place of “resuspension factor” to remove ambiguity of particulate mechanics
- Used NAA to observe suspension factor 100x lower than prediction for indoor electrostatic resuspension
- Initial particulate dispersion dramatically affects observed air concentration
- Long-term suspension factor observations will enable tuning of kinetic rate constants

Future Work

- Impact and validation of similar catenary models
  - Esp. open catenary systems (no constant term)

- Assess resuspension perturbation from contributing sources
  - Wind speed and gust frequency
  - Ground chemical identities and roughness
  - Humidity or other atmospheric content
  - Anthropological activity

- Resuspension of other elements/isotopes
  - Inclusive of radioactive fuel “flea” dynamics
  - (Initial) particle size distribution analysis