

DEVELOPING SURROGATE FAR-FIELD NUCLEAR FALLOUT AND ITS RAPID DECONTAMINATION FROM AIRCRAFT SURFACES

Radioactive Particles and Soluble Radionuclides

William C. Jolin, Ph.D.; Michael Kaminski, Ph.D.; Argonne National Laboratory
Frederick Lancaster; Naval Air Command

ABSTRACT

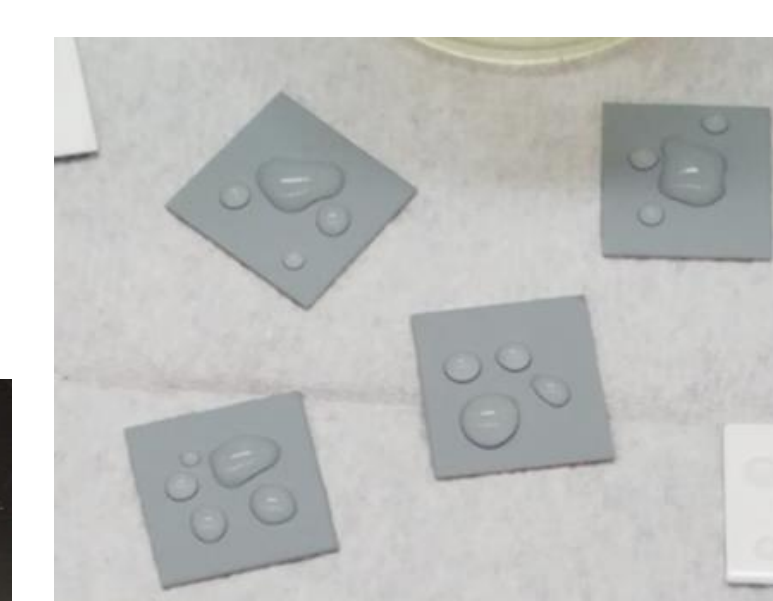
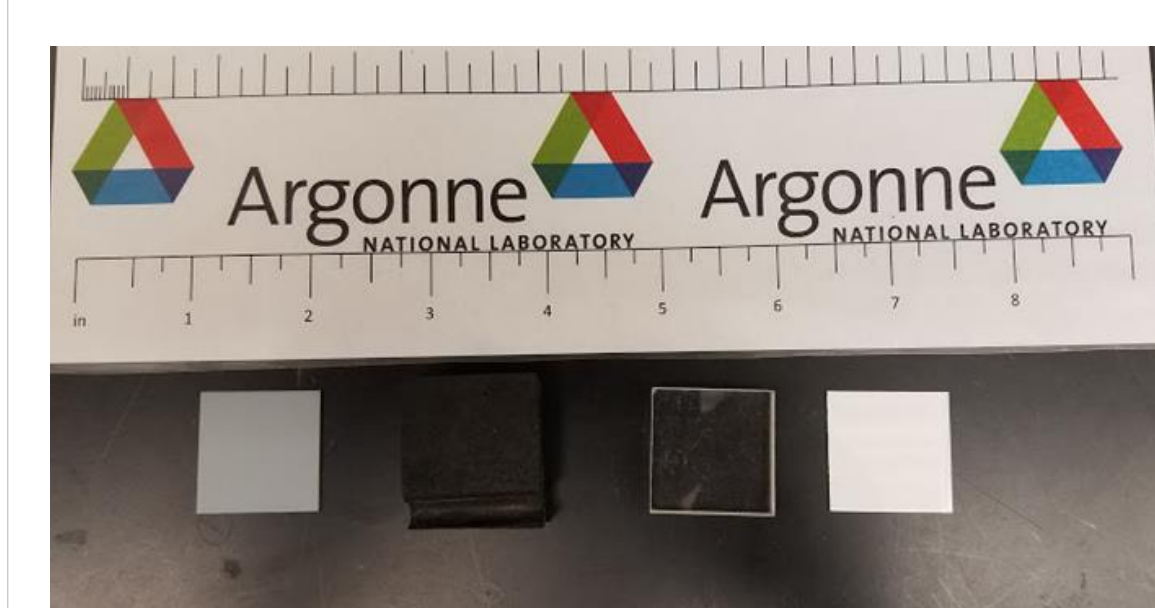
Naval aircraft and vehicles deployed in response to the Tsunami of 2011 and subsequent nuclear meltdown at the Fukushima Daachi nuclear power station were exposed to and contaminated with radioactive materials released to the atmosphere. In response, the Navy and Argonne National Laboratory are currently testing various decontamination methods to clean aircraft surfaces exposed to far-field nuclear fallout. Far-field fallout, which can travel hundreds of miles, includes dissolved radionuclides and fine particulate matter. To simulate contamination with such materials, dissolved Cs-137 and radiolabeled particles were deposited on coupons (1" x 1" sections) of tire, acrylic, and painted metal surfaces. These coupons were subjected to six different decontamination methods including four strippable products, a soap solution, and tap water. Removals of dissolved Cs-137 were generally high, over 90% of the deposited radionuclide was removed after three decontaminations.

To simulate exposure to radioactive particles, silicate particles were developed as surrogate fine particulate matter. A method was developed to radiolabel silica particles by first depositing Eu-152 or Am-241 on the particle surface as sesquioxides, then coating the particle with sodium silicate. Scanning electron microscopy confirmed particle size was not altered while leaching studies into numerous solutions and decontamination agents confirmed that minimal amounts of the radiolabel dissolved in solution. Three different methods to apply the particles were then evaluated. Pressing the coupons into a dish containing the particles resulted in particle clumping on the surface, while applying the particles within water also resulted in a heterogeneous surface coverage. Suspending the particles in ethanol and then applying them to the surface supplied a homogenous surface coverage that provided acute and precise removal percentages. Finally, particle-surface interactions seemingly play an important role in radiological decontamination as removals of particles were lower than those of soluble Cs-137 for aqueous decontamination methods. These decreased removals indicate that more complex removal methods such as the strippable decontamination agents are required to remove particulate far-field fallout from aircraft and similar vehicle surfaces.

MOTIVATION

- Develop surrogate far-field nuclear fallout and application method for decontamination testing
- Test the efficacy of four strippable agents, a soap solution, and tap water to remove soluble Cs-137 and radioactive particulates from four aircraft surfaces (high and low gloss paint, acrylic, tire)

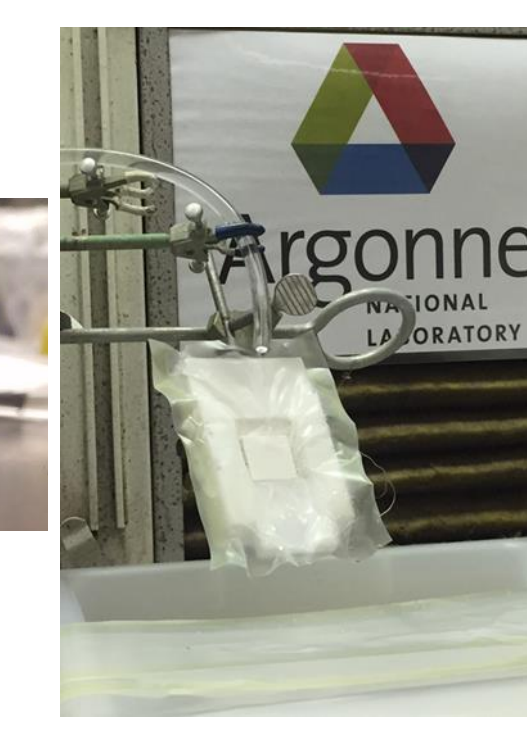
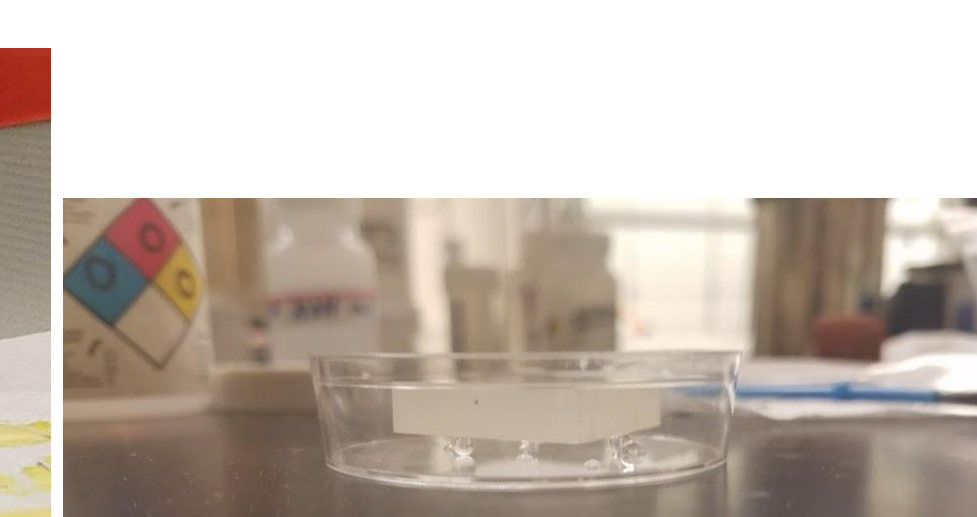
(Below) Test Surfaces



(Above) Dissolved Cs-137 Application

METHODS

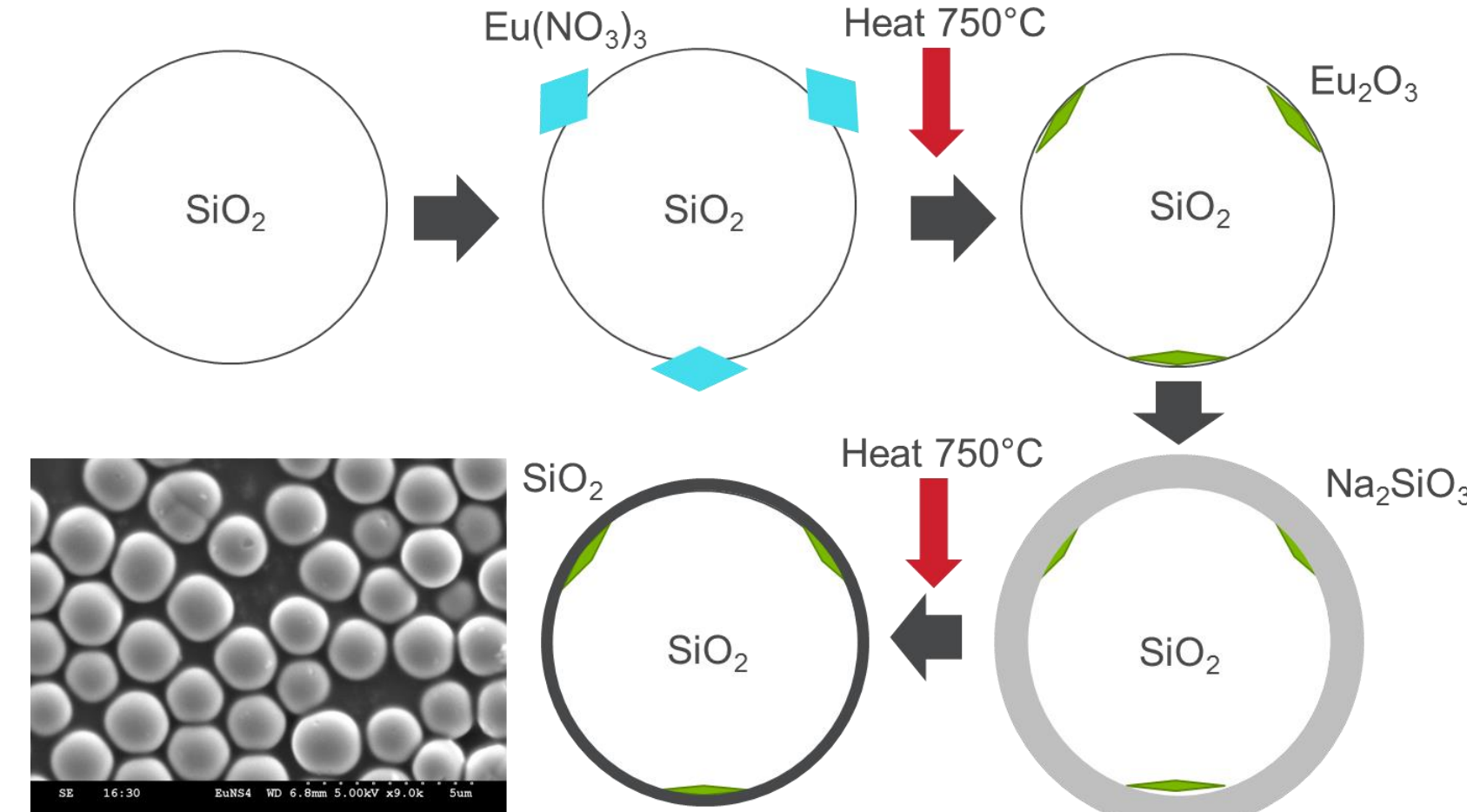
- Dissolved Cs-137 applied to surface and allowed to dry for 24 hours
- For strippable decon agents: polymer applied to surface, allowed to dry, and stripped off (left)
- Solutions
 - Static tests: test surface suspended on glass bead within dish, 10 mL of solution applied and allowed to interact for 1 hour (center)
 - Flow tests: decon solution flowed at low pressure over surface (right) for 5 mins



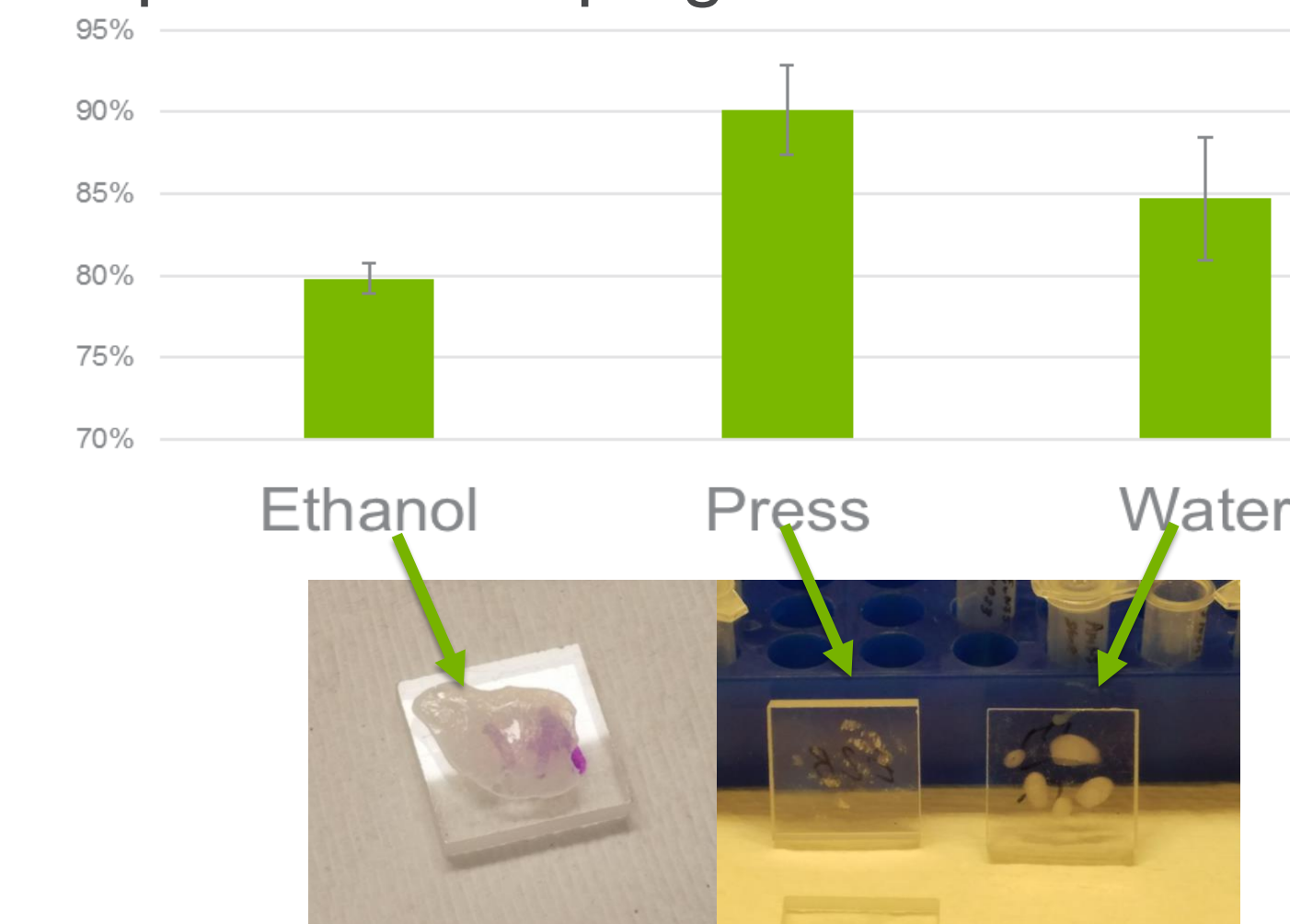
DEVELOPING PARTICULATE SURROGATES

Target was to radiolabel 2 and 0.5 μm silica particles without the radiolabel leaching in solution. Over 20 methods were tried, and in the final method tried, Eu-152 or Am-241 were deposited on the particle surface as trioxides, then coated with sodium silicate. Using ethanol to deposit particle on surface minimized clumping leading to more accurate removals.

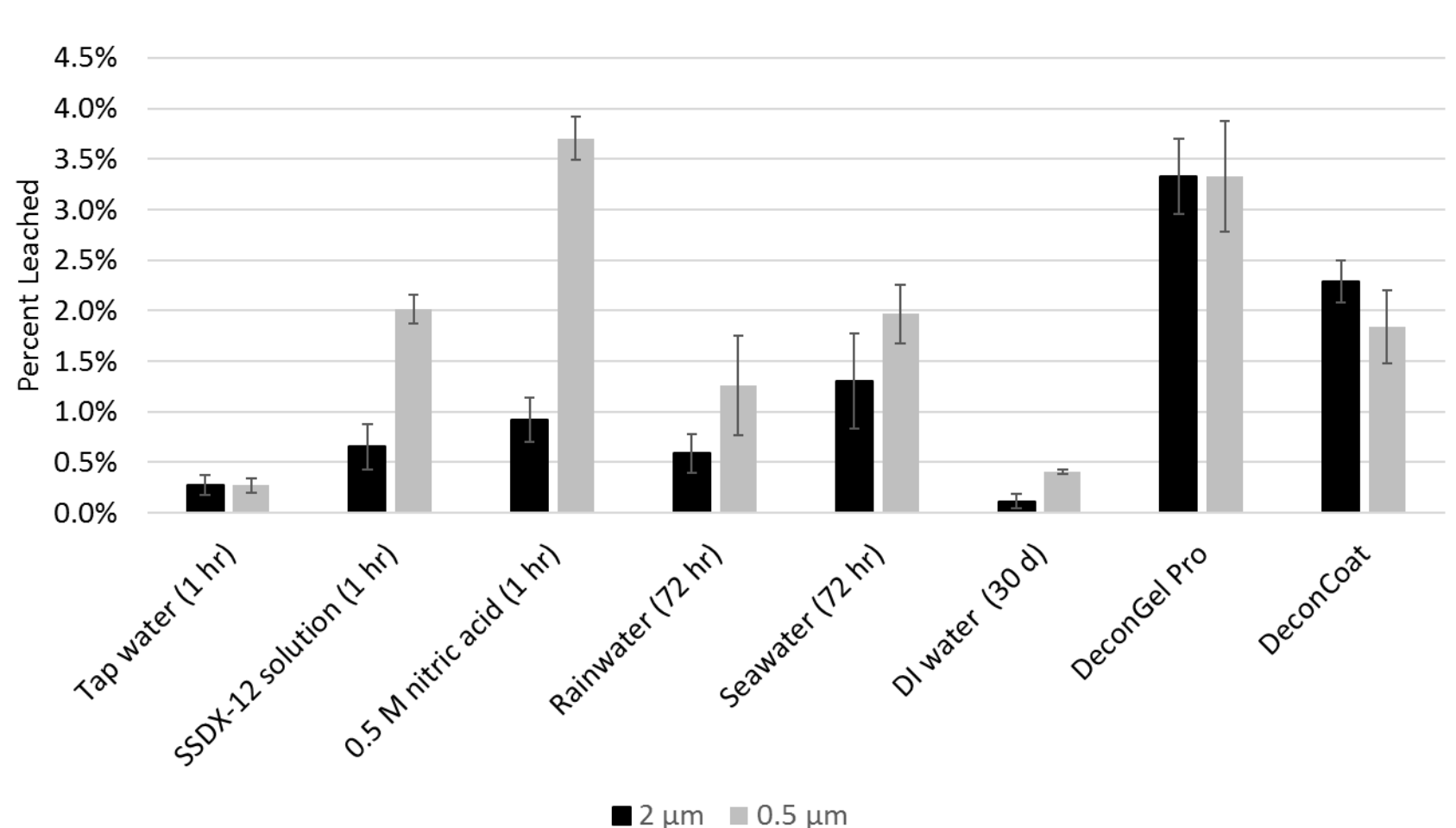
(Right) Visualization of radiolabeling method: First depositing the radiolabel as a nitrate salt, decomposing the salt to a trioxide, and coating with sodium silicate, which decomposed to silica. SEM imaging displayed particles of the correct size.



(Below) Small differences in removals of particles from acrylic surface between spiking in ethanol, pressing the surface into a powder, and spiking in water were the result of particle clumping on the surface.



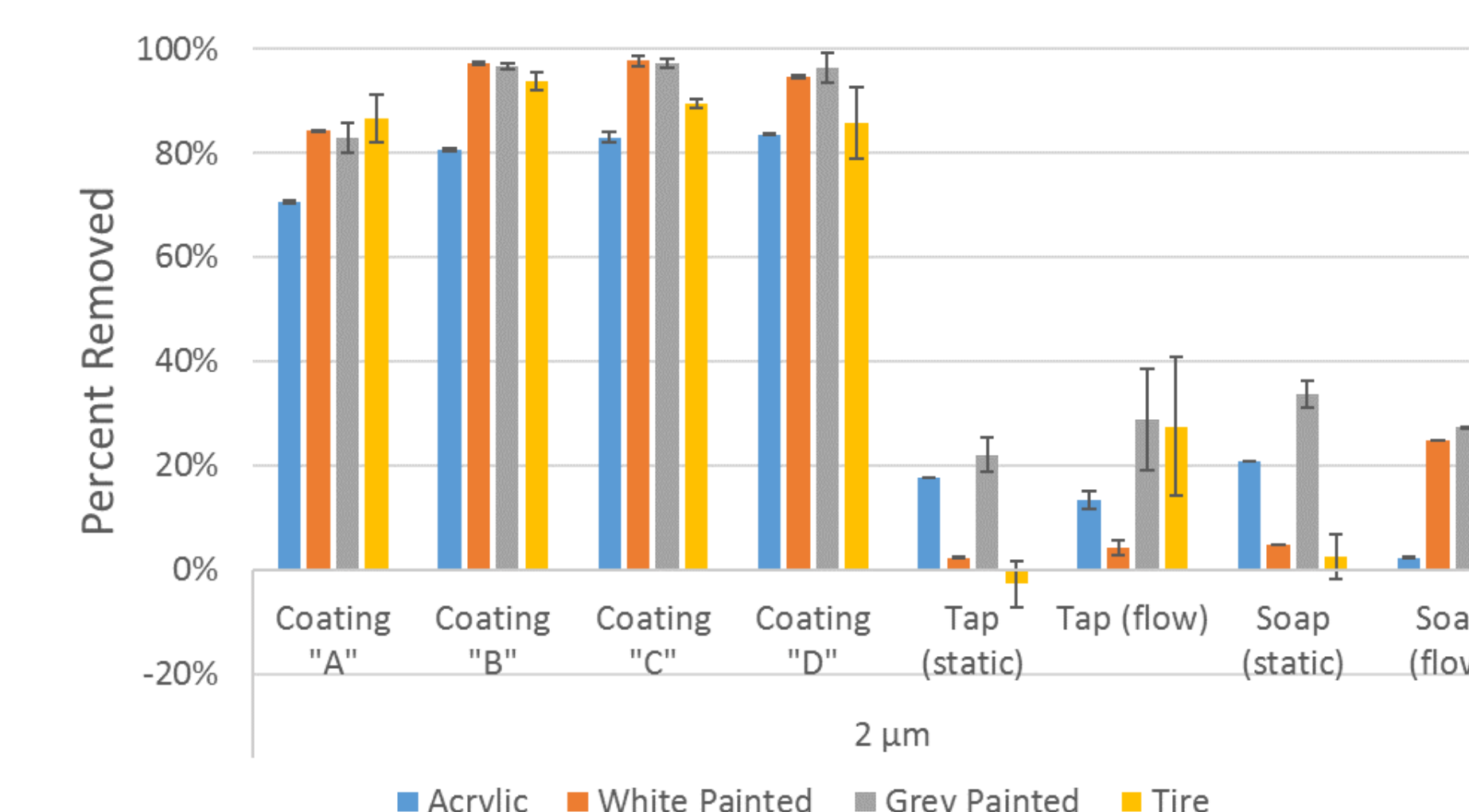
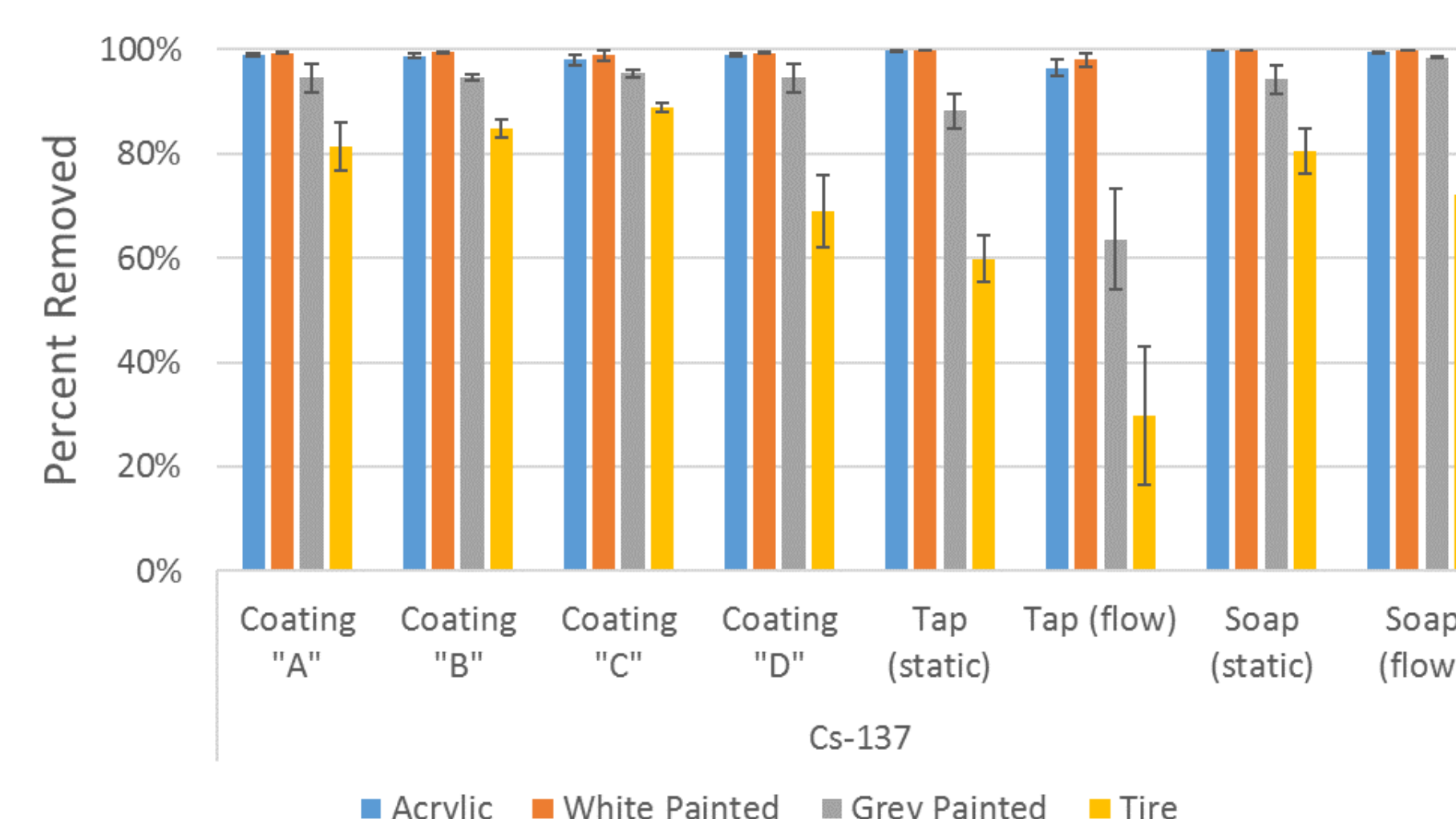
(Below) Leaching of radiolabel in solution was minimal as even leaching in aggressive 0.5 M nitric acid was below 4%



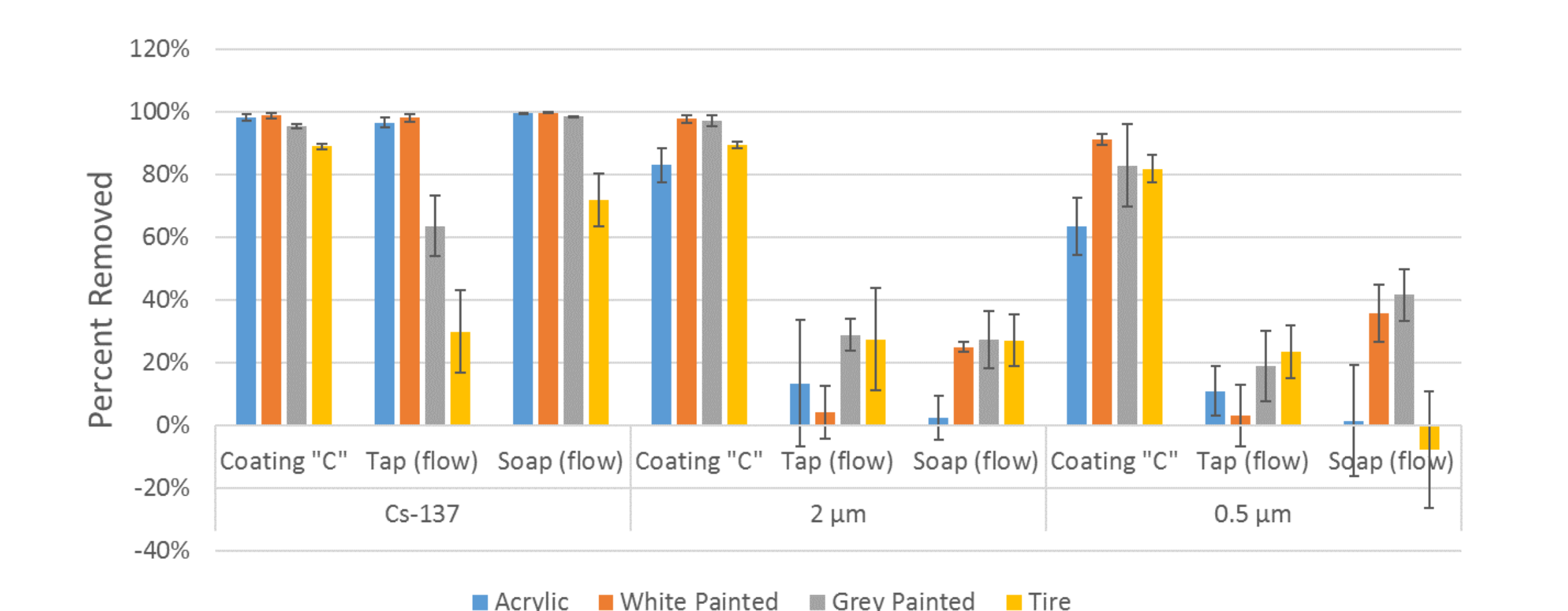
DECONTAMINATION: SOLUBLE VS PARTICULATE

Comparing particle removals with those of Cs-137 reveals that particle-surface interactions are stronger than the electrostatic Cs⁺ ion-surface interactions.

(Right) Initial removals of Cs-137 from all surfaces except tire are high for each of the decontamination agents

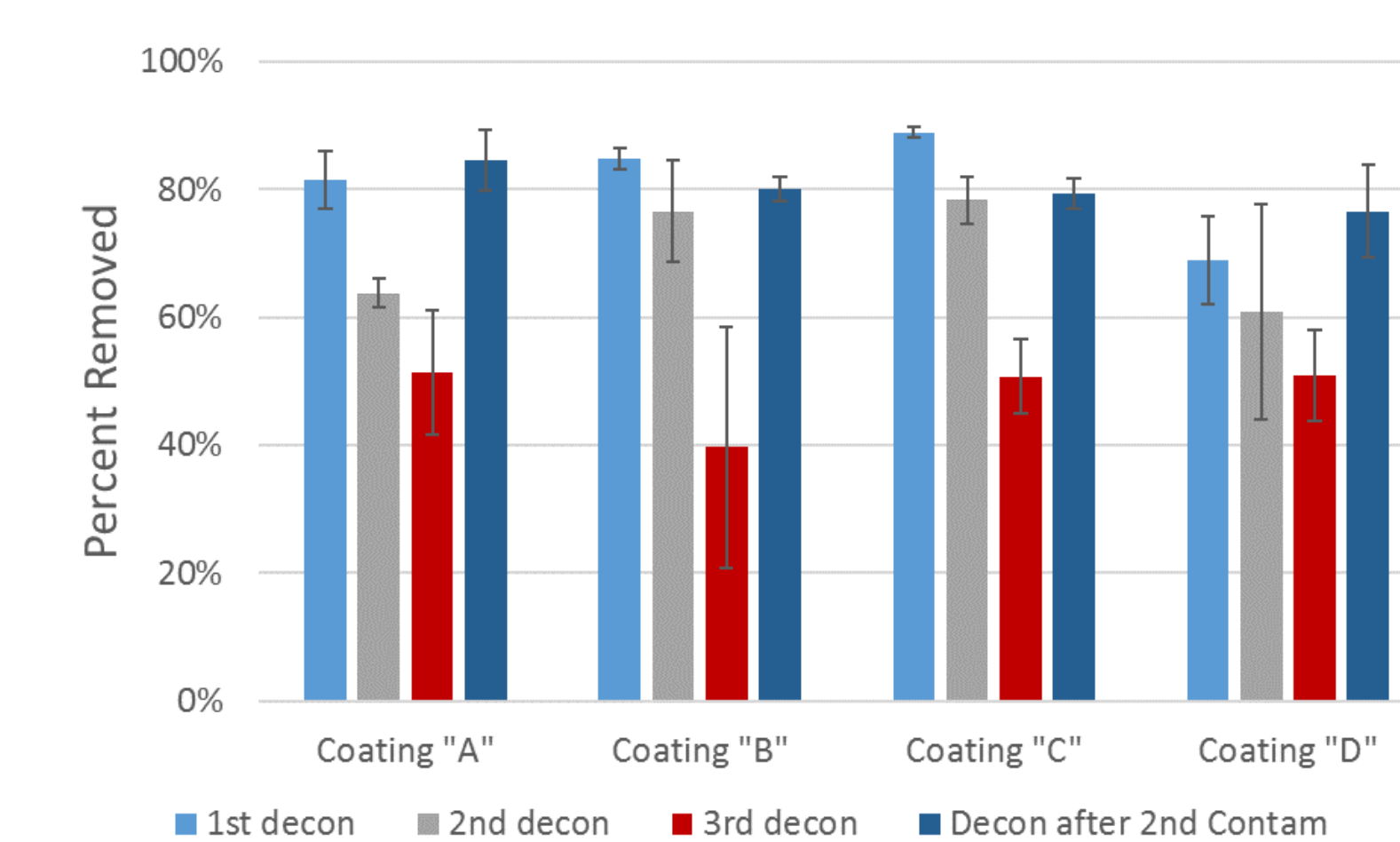
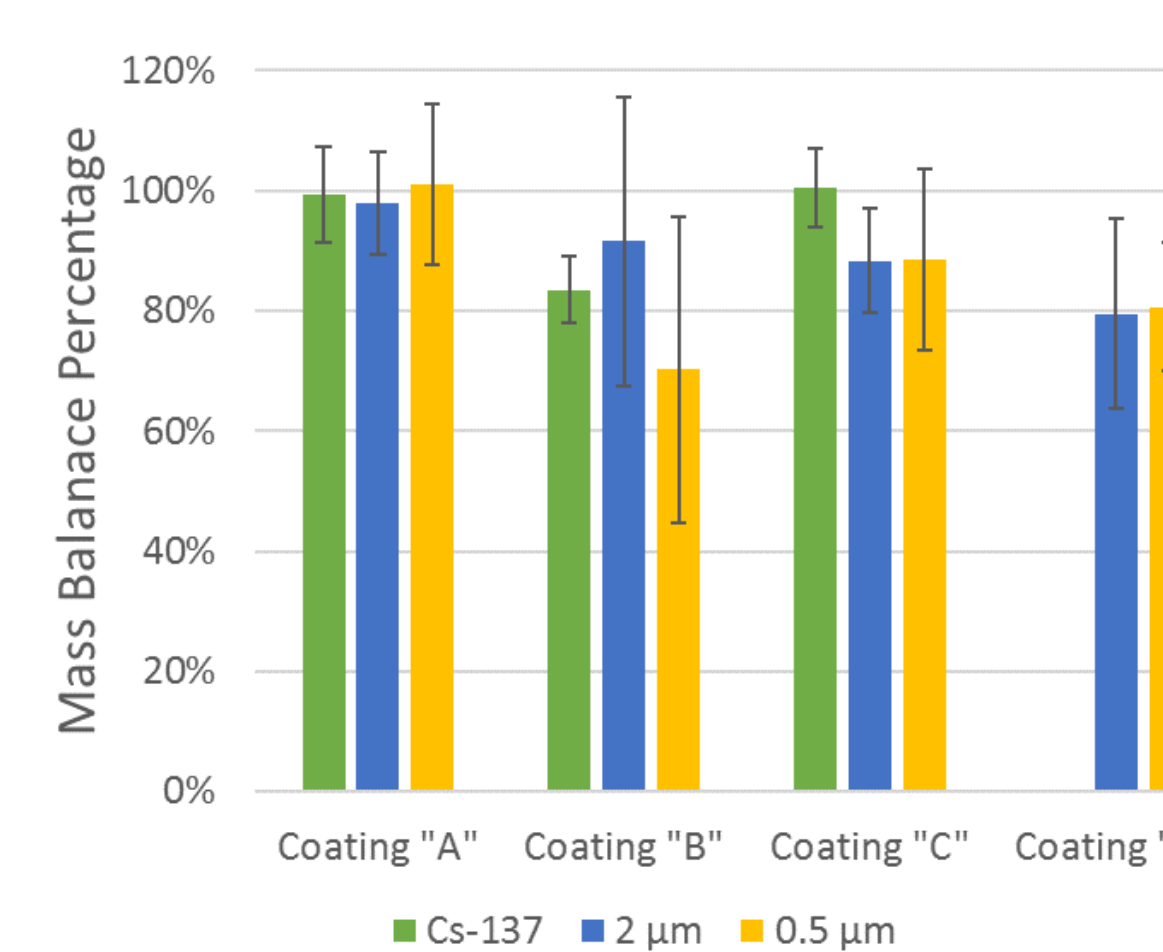


(Left) Particle removals of 2 μm silica particles are distinctly lower when using either soap or tap water compared to the four strippable coating



(Above) Comparing removals by coating "C", flowing tap water, and flowing soap solution highlight the drastic difference between Cs-137, 2 μm, and 0.5 μm interactions with surfaces. Particle attraction likely due to a combination of electrostatic and van der Waals integrations between the particle and the surface.

(Right) Removals of Cs-137 from tire samples decrease after the initial decontamination while differences are seen between the agents after a second contamination



(Left) Mass balance for tire coupons, determined from counting the removed films, revealed difference in radioactivity retention across the strippable decontamination coatings

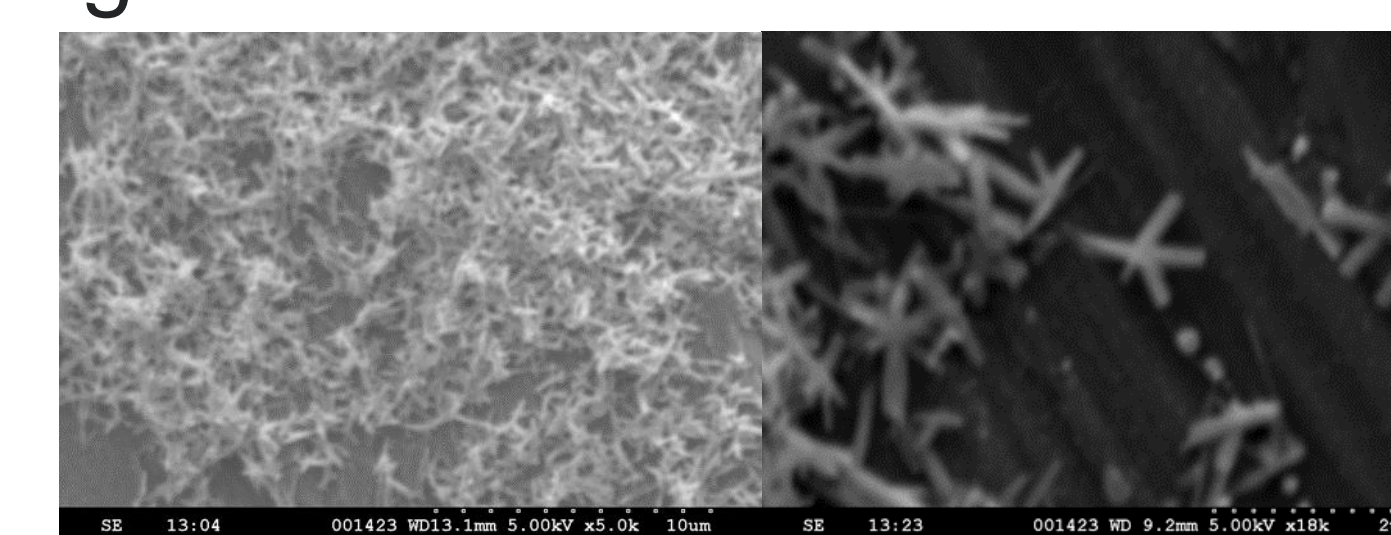
CONCLUSIONS

- Established a method to inertly radiolabel silica particles in the size range of far-field fallout without changing particle size
- Cs-137 removals were high with all decontamination agents
- Lower removals for particles indicated surface interactions and raise concerns for proper decontamination
- SOPs currently being developed

NEXT STEPS: OTHER SURROGATES

- Determining surface properties that influence particle-surface interactions (electrostatic, vdW)
- Mineralogical and radiation effects:
 - Electrostatic and vdW differences between particle types
 - Electrostatic charge buildup due to radioactive decay
 - Particle size: radioactive aerosols can range from nanometers to microns

(Right) Radiolabeled iron oxides provide drastically different coverage and particle geometry



ACKNOWLEDGMENTS

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