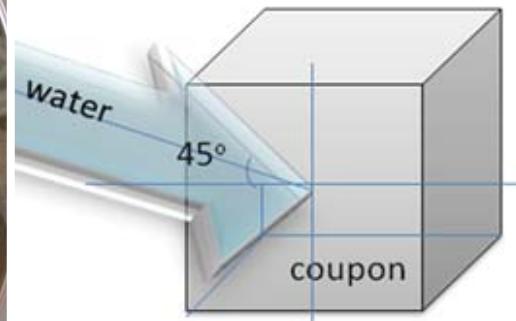
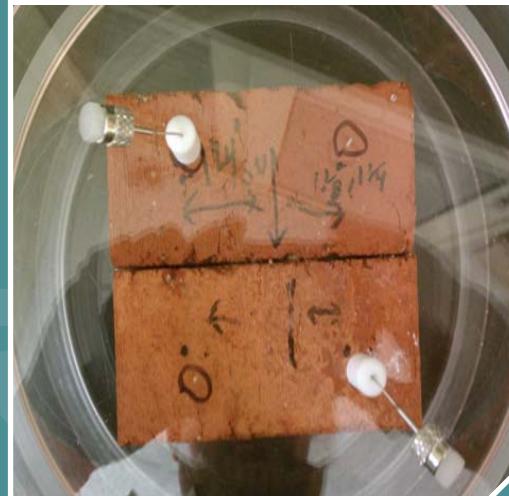


# Water Wash Down of Radiological Dispersal Device (RDD) Material on Urban Surfaces: Effect of Washing Conditions on Cs Removal Efficacy



## **Notice**

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## **Acronyms and Abbreviations**

A	ampere(s)
amu	atomic mass unit(s)
Ce	Cerium
Co	Cobalt
Cs	Cesium
CsCl	Cesium Chloride
DI	Deionized
EPA	U.S. Environmental Protection Agency
ft	foot
Ge	Gallium
GPM	Gallon per minute
He	Helium
ICP-MS	Inductively Coupled Plasma - Mass Spectrometry
In	Indium
L	liter(s)
Li	Lithium
mA	milliampere(s)
Mg	Magnesium
mg	milligram(s)
µg	microgram(s)
µL	microliter
min	minute(s)
mL	milliliter(s)
M <sub>pc</sub>	Average Cs amount (□g) from positive controls
M <sub>r</sub>	Average Cs amount (□g) in replicate rinsate samples
NHSRC	National Homeland Security Research Center
Pb	Lead
ppm	Parts per million
psi	Pounds per square inch
QA	Quality Assurance
QC	Quality Control
RDD	Radiological Dispersal Device
Rh	Rhodium
RH	Relative Humidity
rms	root mean square
Sc	Scandium
sec	second(s)
Tb	Terbium
U	Uranium
V	volt(s)

## Executive Summary

The U.S. Environmental Protection Agency (EPA) holds responsibilities associated with homeland security events: EPA is the primary federal agency responsible for decontamination following a chemical, biological, and/or radiological (CBR) attack. The EPA's Homeland Security Research Program (HSRP) was established to conduct research and deliver scientific products that improve the capability of the Agency to carry out these responsibilities. As part of this program, the EPA's National Homeland Security Research Center (NHSRC) carries out performance tests on homeland security technologies. This current study investigated the impact of water wash down conditions to decontaminate urban surfaces contaminated with cesium (Cs). The contaminated surfaces were prepared and maintained at constant relative humidity (RH) and washed in a chamber simulating the delivery of water from a fire hose. Various water wash down conditions (e.g., duration, water pressure, angle, and pattern) were evaluated for their efficacy at removing Cs from the test coupon surfaces. This removal was determined by the measurement of the amount of Cs in the water rinsate samples of a function of each individual wash condition.

**Experimental Procedures.** Coupons were pre-conditioned at 33% RH at  $21 \pm 3^\circ\text{C}$  for five weeks before being dosed with nonradioactive cesium chloride (CsCl). The CsCl particles were deposited as aerosols onto the pre-conditioned urban surfaces of the test coupons using CsCl-methanol solution. Twenty-four hours after contamination, the coupons were decontaminated using a simulated wash down by water from a fire hose. The contaminated coupons were washed under various wash conditions: wash duration (5,

15, and 20 seconds), water nozzle pressure (40, 80, 120, and 160 pounds per square inch (psi)), wash angle (45 and 90 degrees), wash patterns (from bottom to top and from top to bottom). To trace the amount of decontamination resulting from these wash down tests, the rinsate from each wash down was collected and analyzed for amount of Cs using Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

**Results.** Various conditions of water wash down using a simulated fire hose chamber were tested to investigate the impact of water wash on Cs removal from three different urban surfaces. Overall (asphalt, brick, and concrete coupons combined), there was a statistically significant (Student's *t*-test,  $p=0.0098$ ) effect of washing duration. The five second time period exhibited a consistent lower efficacy than the 20 second time period. There was no statistically significant effect of the wash angle among asphalt samples, but the  $45^\circ$  angle gave lower efficacy than the  $90^\circ$  angle among brick ( $p=0.0595$ ) and concrete samples ( $p=0.0350$ ). Water volume was found to have a significant positive effect on efficacy among asphalt ( $p=0.0080$ ) and concrete samples ( $p=0.0325$ ). The effectiveness of Cs removal increased with higher water pressure per applied water volume for asphalt and concrete samples but not for brick samples. Wash pattern tests were conducted with the brick and concrete coupons. The results showed no difference in Cs removal efficacy for the two different patterns tested.

## 1.0 Introduction

An explosive radiological dispersal device (RDD), also called a dirty bomb, is the combination of a conventional explosive device with radioactive materials that can be obtained from industrial, commercial, medical, or research applications.<sup>1</sup> An RDD attack can impact a society in various ways, including creation of casualties, disruption of the economy, and potentially desertion of the contaminated area.<sup>2-5</sup> Fast and cost-effective decontamination strategies are critical to minimize the social and economic damage resulting from an RDD event.

One of the major processes for remediation of the radioactively contaminated surfaces is wash-off via water application. Weather phenomena such as rain also play a significant role in the remediation process. Numerous studies have been conducted to assess the impact of water exposure on contaminated surfaces resulting from nuclear accidents.<sup>6-9</sup> These studies have demonstrated the removal of radionuclides from various types of surfaces via rain run-off and water wash-off after the Chernobyl accident. The study by Roed<sup>6</sup> showed that the first rain run-off from the contaminated surfaces removed a higher portion of radioactive contaminants compared to the subsequent rain or water application on the same surfaces. In addition, Andersson et al.<sup>9</sup> showed that the radioactive contaminant removal rates by rain or water application varied depending on the surface type. These results are consistent with more recent studies completed by the U.S. Environmental Protection Agency (EPA). US EPA has conducted a test to investigate the fate and transport of Cs on urban surfaces after rain exposure.<sup>10</sup> The results from the study showed that the fate of Cs on

surfaces was dependent upon contaminant deposition conditions and surface types.

Rain or water application onto porous surfaces may result in increasing difficulty with decontamination. The US EPA's rain study showed extended subsurface penetration of Cs through porous materials when the surfaces were exposed to rain.<sup>10, 11</sup> This subsurface penetration of radionuclides can increase the difficulty of decontamination by limiting the mass transfer of Cs ions to the surface. This subsurface penetration occurs by wet deposition of water soluble particles onto porous surfaces or rain or high RH exposure of dry deposited water-soluble particles on porous surfaces. An optimized application of water may remove radioactive materials more effectively than weathering itself and would result in reducing the amount of subsurface penetration.

The current study investigated the impact of various water wash down conditions on Cs removal efficiency from porous surfaces using a simulated fire hose water delivery system. In this study, the nonradioactive CsCl particles were deposited by spraying a CsCl-methanol solution onto the test surfaces. This method kept particles closer to the surfaces and in the form of CsCl compared to the particles from a CsCl-water solution; this is because most of the methanol in the droplets evaporates before the droplets touch the surface (compared to the water-based solution). The study focused on the assessment of various conditions of water wash down parameters including wash down duration, water pressure, wash angle, and wash patterns. Nonradioactive CsCl particles were chosen as an RDD surrogate materials. Three

materials (asphalt, brick, and concrete) were selected as the urban surface materials to be used as test substrates in this study. The results of this study provide insight into

parameters influencing the effectiveness of water wash down methods on urban surfaces contaminated through a release from an RDD.

## **2.0 Materials and Methods**

### **Test Overview**

The amounts of deposited cesium removed from urban surfaces via water wash down were studied under specific conditions of water application. Wash down conditions included wash down duration, water pressure, wash angle, and wash patterns. Three different building materials were contaminated with Cs particles via aerosolization, and the contaminated surfaces were washed simulating water delivery from a firehose. The wash down rinsate samples were collected and analyzed for the amounts of Cs that were removed from the surface.

Test coupons were prepared and contaminated at the EPA facility located in Research Triangle Park, NC. Test coupons were conditioned at a constant 33% RH for five weeks before contamination. The coupons were then dosed (contaminated) with Cs and conditioned at 33% RH for 24 hours before wash down. All coupons were washed in a chamber in which a wash down using a firehose was simulated. This simulation is based on the use of a reduced distance and small surface area with less

water delivered through the use of a conventional garden hose-type nozzle that sprays water to a smaller coupon area while maintaining the same pressure on the surface (normalized to material surface area) as observed in a realistic fire hose water wash down application. The Cs amounts from the collected wash down rinsates were analyzed using Inductively Coupled Plasma with Mass Spectrometry (ICP-MS).

### **Building Materials**

Three different building materials were used in this study and the material information is described in Table 1. Coupons (3.0 cm x 3.0 cm x 3.0 cm (W x L x H)) for the wash down duration, water pressure, and wash angle tests were prepared using a diamond saw with distilled water as the lubrication fluid. For the wash pattern test, larger concrete and brick coupons (12.5 cm x 10.0 cm x 2.5 cm (W x L x H)) were used. Each coupon was inspected visually to find any defects, cracks, or stains. Coupons with any defects discovered by visual inspection were discarded.

**Table 1. Building material description and source.**

Material	Description	Locality	Source
Brick	Red, fine-grained	Made from NC red Triassic clay	Triangle B. Company, Durham, NC
Concrete	Cement with sand aggregate (prepared within six months of tests and not weathered)	Concrete premix (QUIKRETE® Atlanta, GA)	Home Depot, NC
Asphalt	Laboratory Pressed Asphalt (prepared within two years of tests and not weathered)	NC	NC Department of Transportation

Freshly cut coupons were stored and soaked in deionized water overnight (at least 12 hours). These coupons were dried in an oven at 80 °C at negative pressure (~10" Hg) for 24 hours. Coupon dimensions and weight were recorded. Five sides of each coupon were sealed with water-impermeable sealant (Stonelok™ E3, Richard James Specialty Chemicals Corp., Hastings on Hudson, NY). The top surface remained unsealed for deposition of the Cs. The sample identification and the top face designation were marked on the two opposite sides of each coupon.

### Coupon RH conditioning

Coupons were placed into a chamber that was held at 33% RH for at least five weeks before surface contamination. The coupons were stored in RH-controlled chambers and were opened only when coupons needed to be added or taken out. The contaminated coupons were washed down using a water delivery system simulating a firehose washdown after 24 hours of contamination.

Coupons were stored in the same 33% RH chamber during the 24 hours after contamination. RH and temperature in the constant RH chambers were monitored and recorded every 10 minutes throughout the test periods using a temperature/RH data logger (HOBO U10-003, Onset Computer Corporation, Pocasset, MA). Triplicate coupons were prepared for each water wash down testing condition. After a wash down, coupons were dried in laboratory air for 24 hours and then stored in the 33% RH chamber.

Prior to particle deposition, the top surfaces of the larger coupons (12.5 cm x 10.0 cm) were thoroughly cleaned with a 2550 psi/2.3 gallons per minute (GPM) pressure washer (Troy-Bilt Gas Pressure Washer, Lowe's, Durham, NC) to remove any loose pieces of building materials. After cleaning, the large coupons were dried in laboratory air for at least five weeks. After deposition, large coupons were stored in laboratory air (23 ± 2 °C and 40 ± 2% RH) for 24 hours before the wash down test.

## Cesium Particle Deposition

A Cs-containing methanol solution was deposited onto coupons using a metered syringe (MicroSprayer® Aerosolizer, Model 1A-1C and FMJ-250 High Pressure Syringe, Penn-Century, Inc., Windmoor, PA). The deposition liquid volume was 25 microliters ( $\mu\text{L}$ ) per coupon with 200 parts per million (ppm) of CsCl (99.99%, Fisher Scientific, Pittsburgh, PA) solution. For small coupons, the deposition chamber was designed to center a coupon on the bottom of the chamber and to slide the syringe needle to spray aerosols as shown in Figure 1 through a centered hole in the top lid. This procedure yields a nominal 3.7 microgram ( $\mu\text{g}$ ) deposition of CsCl onto a 3 cm x 3 cm coupon surface.

Four different locations were contaminated on the large coupons using the deposition apparatus shown in Figure 2. Each deposition volume was 25  $\mu\text{L}$  and contained 200 ppm of CsCl solution. The large

coupon was contaminated with approximately 15  $\mu\text{g}$  of CsCl per coupon. Four of the coupon sides were covered with painter's tape to prevent the potential deposition of CsCl on the sides, and this tape was removed after deposition.

The deposition amount was calibrated (positive control) by depositing CsCl solutions onto clean polyethylene plastic sheets held at the same distance from the tip of the syringe and with the same surface dimensions as the building test coupons. The five positive control samples for Cs were transferred to clean 50 mL tubes for individual extraction. The tubes were filled with 5% ultrapure OPTIMA HNO<sub>3</sub> (Sigma-Aldrich, St. Louis, MO) in deionized water until the solution covered the plastic surface entirely. The plastic sheets were extracted by sonication for 20 minutes. After removal of the coupon, the tubes were filled up to 50 mL with 1% nitric acid solution and analyzed by ICP-MS.

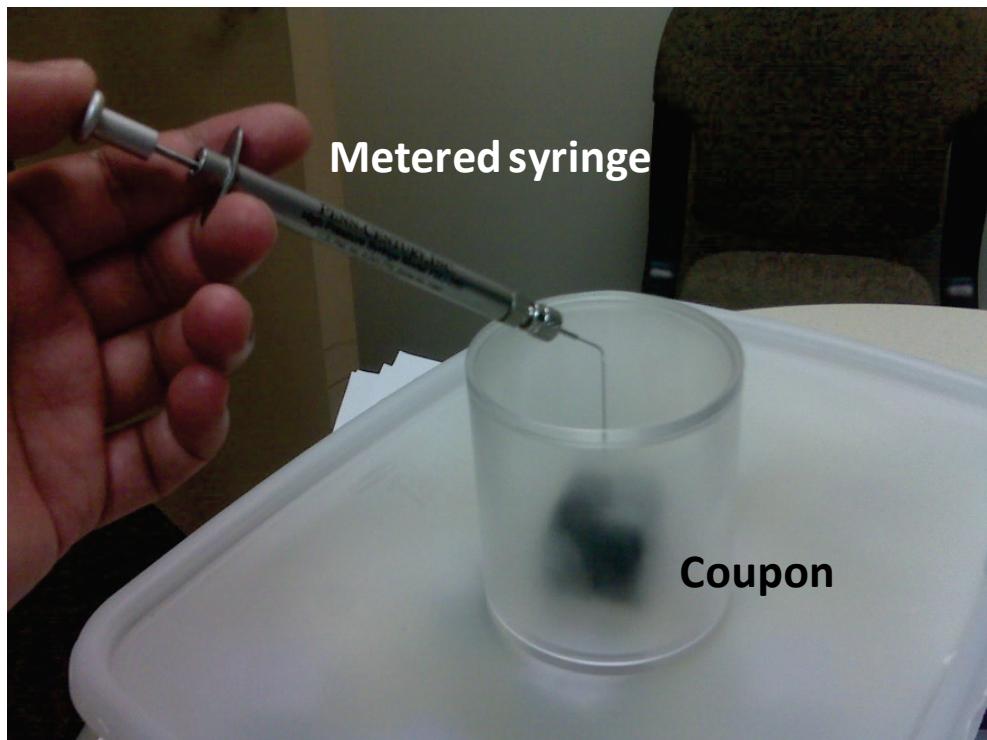


Figure 1. Illustration of particle deposition onto a small coupon

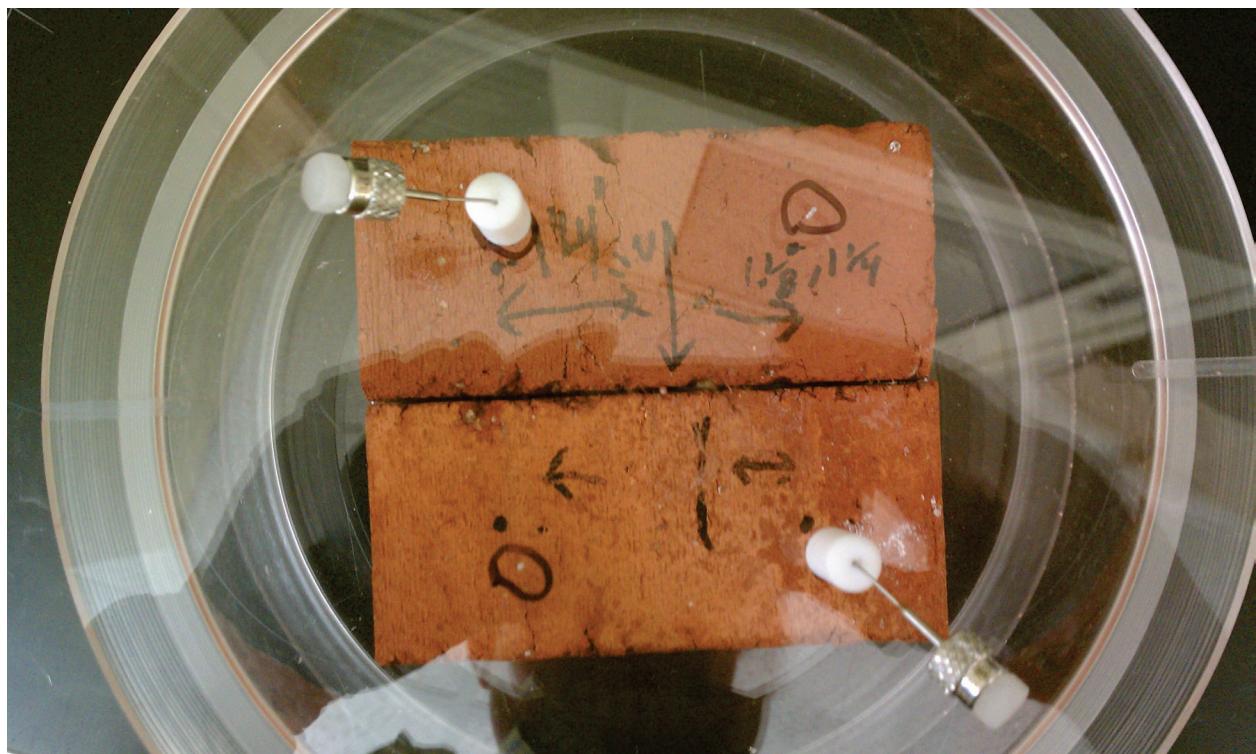
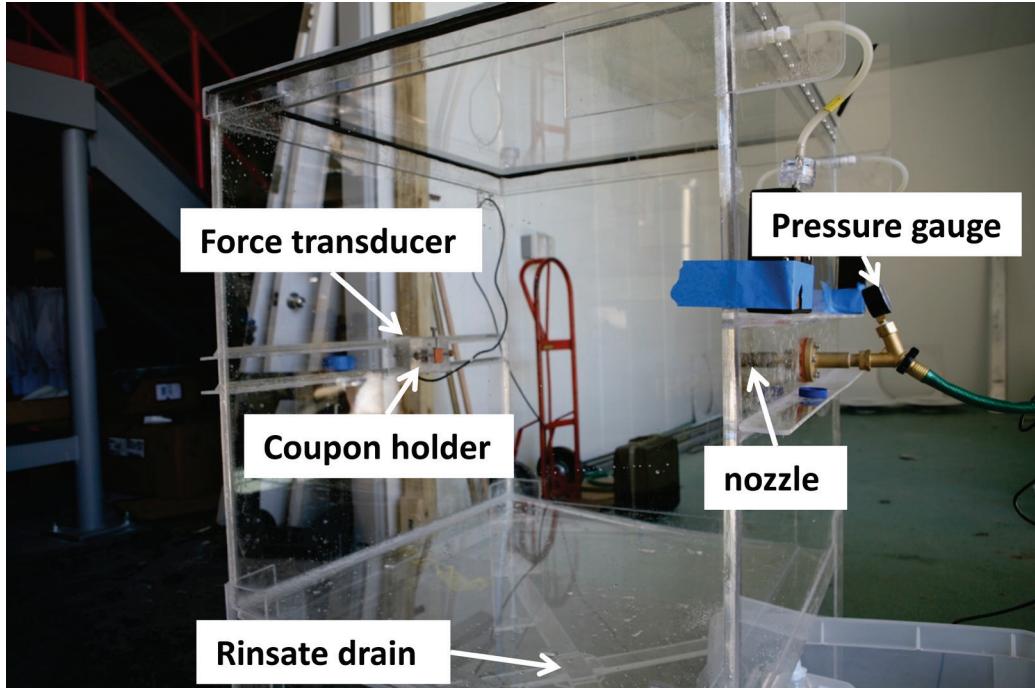


Figure 2. Illustration of particle deposition onto a large coupon

## **Firehose System**

All coupons were washed in a chamber using a simulated firehose water delivery system. Water delivered from a firehose as used for washing down building surfaces was simulated based on an experiment performed at a Durham, NC, Highway Fire station. A force transducer that was used to measure the pressure showed that water from a 50 psi, 2.4 cm diameter smooth bore tip firehose nozzle held 30 feet (ft) from the target generated a 1.6 psi pressure on an 8" diameter target surface with a ~6.5 inch water jet diameter. This information was used as the initial guide for this study. The nozzle, target dimensions, and distance from nozzle to the target were scaled down in the experimental setup to mimic the 1.6 psi pressure at the target surface. A Plexiglas chamber (90 cm x 90 cm x 90 cm) was built and is shown in Figure 2. The nozzle was fixed in the center of a chamber side, and the coupon holder was positioned on the opposite side of the nozzle. The distance between the nozzle and the coupon surface was kept constant as 23" for all test conditions. The chamber top was designed to be able to open and close for cleaning the inside of the chamber after each test. The chamber bottom was slanted slightly to collect the rinsate water. A force transducer

was located on the back of a coupon in the coupon holder to measure the applied pressure by the water jet emitted from the nozzle. The nozzle and water pressure were adjusted to create a water jet with the same cross section as the small coupon surface area (3 cm x 3 cm) at the point of impact and with a 1.6 psi pressure on the surface. A water pressure of 120 psi was required to generate this pressure at the coupon surface. The water pressure was monitored using an inline pressure gauge (Watermaster 200 PSI Pressure Gauge, 91130, Orbit, Bountiful, UT) to ensure reproducible wash down conditions. Deionized water was used for the simulated firehose test and the water was applied for a specified time. The rinsate water was collected in plastic containers (Cubitainer, Fisher Scientific, Pittsburgh, PA) located under the bottom drain. The rinsate containers were weighed pre- and post-experiment to measure the amount of water used during each wash down test. After the wash down, the coupons were removed from the chamber coupon holder and the chamber walls were cleaned and dried before the next test. The rinsate collecting containers were thoroughly cleaned with Triton-X (Fisher Scientific, Pittsburgh, PA) solution and deionized water prior to each test.



**Figure 3. Simulated Firehose System**

## Wash Down Conditions

### Wash down duration

The coupons were washed for three different durations: 5, 15, and  $20 \pm 1$  seconds. For the wash down duration tests, the other wash down conditions were 120 psi water pressure and a 90 degree angle between water jet direction and exposed coupon surface. Water from the hose nozzle covered the entire coupon surface. The collected rinsate volume for the 5-, 15- and 20-second washes was collected into one-gallon Cubitainers. The bottom of the chamber was capped prior to washing and the volume of rinsate collected was split into two samples. The rinsates were analyzed for Cs amount to determine whether the decontamination efficacy was dependent on the wash down duration.

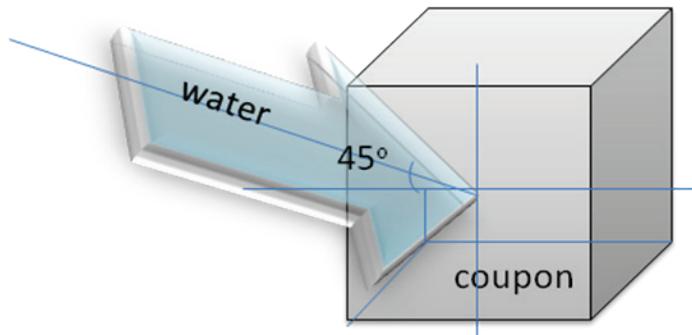
### Water pressure

Four different water pressures were applied to the coupons from the same distance and for the same duration. The applied nozzle water pressures were 40, 80, 120, and  $160 \pm 5$  psi. Pressure was adjusted by changing the nozzle orifice size and also varying supply water pressure while keeping the same size orifice in the nozzle. The rinsates from various water pressure tests were analyzed for Cs amount to determine the dependence of water pressure on the decontamination efficacy. For these tests, the wash duration was 20 seconds and wash angle was 90 degrees. The pressures applied to the coupons during wash down were monitored using a pressure transducer (S Beam Load Cell, LCCA-100, Omega, Stamford, CT) attached to the coupon holder.

### **Wash angle**

The previous wash down tests were conducted with a 90 degree angle between the water jet direction and the coupon

surface. The water application angle was adjusted to 45 degree as shown in Figure 4. The other conditions during this test were 120 psi water pressure for 20 seconds.

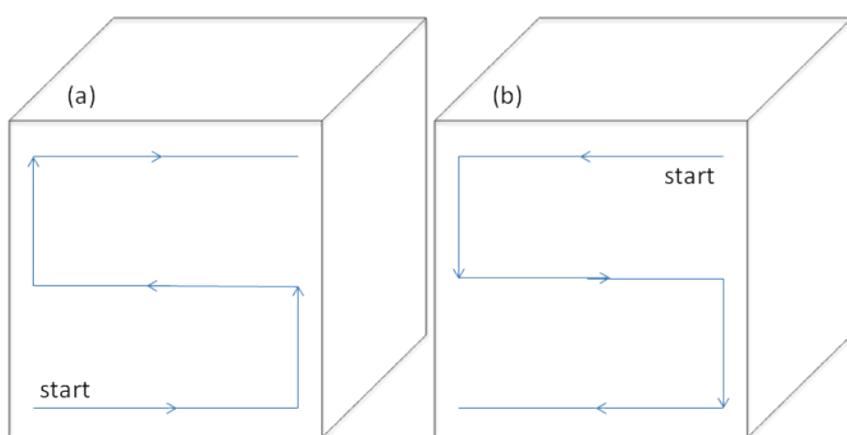


**Figure 4. Water wash down with 45° angle to coupon surface**

### **Wash pattern**

Two different wash patterns were tested with 12.5 cm x 10.0 cm x 2.5 cm (W X L x H) coupons. The two different patterns are shown in Figure 5. One pattern (Figure 5(a)) involves washing from bottom to the top, and the other pattern was the opposite (from top to bottom) as shown in Figure 5(b). The wash down angle was 90 degrees and the

water pressure was 120 psi for 20 seconds. The rinsates were analyzed for the effect of wash pattern on decontamination efficacy. This test was conducted with concrete and brick coupons only, as these surfaces are often used as vertical surfaces. Asphalt surface was not tested since it is usually used for horizontal surfaces.



**Figure 5. Water wash down test patterns**

## Test Matrix

The coupon test matrix is shown in Table 2. Three replicate samples were prepared for each surface material. As shown in Table 2, a total of three blank coupons were prepared for all three surface materials. Blank coupons were the coupons without Cs

deposition. These blank coupons were conditioned at 33% RH, and after wash down they were placed back in the 33% RH chamber. These three blank coupons were washed down and the rinsates were collected for baseline cesium concentration determination.

**Table 2. Test Matrix for Simulated Firehose**

Substrate	Wash Duration	Water Pressure	Wash Angle	Wash Pattern	Small Blanks	Large Blanks
Concrete	9	9	3	6	3	3
Brick	9	9	3	6	3	3
Asphalt	9	9	3	0	3	0

Positive controls were created by depositing cesium particles on a clean cut Ziploc® surface using the same conditions as the coupon deposition. A total of five positive control samples were prepared before deposition of Cs onto the building material test coupons, and the extracted solution was analyzed by ICP-MS using EPA Method 200.8.<sup>13</sup> A total of five positive controls for large coupons were also prepared using clean cut Ziploc® surfaces, and cesium particles were deposited in the same manner as for the test coupons. Sets of positive controls were prepared for each of the test parameters. The extracted solution was analyzed using ICP-MS.

## Analysis of Wash Down Rinsates

The rinsate samples in the plastic containers were transferred to three 50 mL clean vials and the rest of the rinsate samples were discarded. The samples were stored in a refrigerator until they were analyzed. Before analysis, the rinsate samples were

filtered using a syringe filter, and 5 mL of each sample was transferred to a clean 15 mL tube labeled with the sample identification. The rinsate samples from the wash down tests were analyzed for Cs using EPA Method 200.8. A model ELAN 6000 ICP-MS (Perkin Elmer, Waltham, MA) was used for Cs analysis. The operating conditions of the ICP-MS are summarized in Table 3.

A 10 µL aliquot of internal standard reference solution was dispensed into every sample vial before analysis. The internal standard solution contained 100 mg/L of various elements (Ge, In, Li, Sc and Tb). Rinsates for the blank samples were also collected and processed in the same manner as the test coupons prior to analysis using EPA Method 200.8. All containers used for dilution, extraction, and analysis were cleaned using 1% Triton X-100 (Fisher Scientific, Pittsburgh, PA) solution in deionized (DI) water followed by multiple rinses with DI water and dried in a Class 100 clean bench for 24 hours.

**Table 3. Operating conditions for ELAN 6000**

Parameters	Values
Rf power	1200 Watts
Carrier Gas Flow Rate	0.87 liters (L)/min
Lens Voltage	9 Volts (V)
Analog Stage Voltage	-2600 V
Pulse Stage Voltage	1850 V
Discriminator Threshold	70 mV
AC Rod Offset	-8 V
Integration Time	2000 sec
Scanning Time	4.120 minutes
Replicates	3
Sweeps	20
Sample Uptake Rate	~0.10 mL/min
Plate Voltage	3347 V DC
Plate Current	0.50 A DC
Grid Current	94 milliamperes (mA) DC
Filament Voltage	6.18 V root mean square
Dwell Time per atomic mass unit (AMU)	100 min  He (3.016 amu)= 2089 Mg (23.985 amu)= 2065 Rh (102.905 amu)= 1998 Ce (139.905 amu)= 1995 Pb (207.977 amu)= 2113 U (238.050 amu)= 2223
Resolution	

## **Removal Efficacy**

The efficacy of a firehose wash down was assessed by determining the amount of Cs in the water wash rinsate samples. The Cs amount in the rinsate water samples was

compared to the Cs amount in the positive control rinsate. Removal efficacy of Cs from the coupon material was calculated as the ratio of  $M_r$  and  $M_{pc}$ :

$$\text{Removal Efficacy (\%)} = M_r / M_{pc} \times 100$$

where  $M_r$  is the average Cs amount ( $\mu\text{g}$ ) in replicate rinsate samples, and  $M_{pc}$  is the average Cs amount ( $\mu\text{g}$ ) from five positive controls.

## **4.0 Quality Assurance/Quality Control**

QA/QC procedures were performed according to the quality assurance project plan for this test (available upon request). All equipment (balance), monitoring devices (e.g., pressure gauge, relative humidity, temperature) and an analyzer (ICP-MS) used at the time of evaluation were verified as being within calibration. QC samples generated during testing included use of positive control coupons, blank coupons, and wash down water samples. The average recoveries for the Cs positive controls were

between 70% and 120%. The relative standard deviation of Cs amounts from positive control recovery results were less than 14%. The analysis results of blank coupon rinsate samples showed the Cs amount to be below the minimum quantification limit (< 0.025 µg/L) for all three materials. The clean deionized water before wash down application was analyzed for Cs and the results showed that Cs amounts were below minimum quantification limit (< 0.025 µg/L).

## 5.0 Results

### Cs Removal Efficacy

Cs removal efficacy results are listed in Tables 4 for asphalt, brick, and concrete materials, respectively. The complete data are listed in the Appendix. Overall, there was an effect of the variation of the material ( $p < 0.0001$ ). Asphalt samples resulted in higher decontamination efficacy values than

brick samples ( $p < 0.0001$ ) with a mean difference of 28% or concrete samples ( $p < 0.0001$ ) with a mean difference of 21%. There was no statistically significant difference between brick and concrete samples ( $p=0.1549$ ). Analysis of variance (ANOVA) and student's  $t$ -tests were conducted using statistical software (SAS version 9.2, SAS Institute, Cary, NC).

**Table 4. Effect of material types on wash efficacy**

Paired Comparison	Method	p-value	Mean difference	Sample size
Overall	ANOVA	<.0001	Not applicable	102
Asphalt vs. Brick	$t$ -test	<.0001	Asphalt > Brick, 28%	30 vs. 36
Asphalt vs. Concrete	$t$ -test	<.0001	Asphalt > Concrete, 21%	30 vs. 36
Brick vs. Concrete	$t$ -test	0.1549	Not sig. diff., 5%	36 vs. 36

### Wash Down Duration

The average Cs removal efficacy results as a function of wash duration is shown in Table 5. The coupons were washed in the

simulated fire hose test chamber under the following conditions: water pressure 120 psi and wash angle 90 degrees. The error ranges in Table 5 are one standard deviation of triplicate sample results.

**Table 5. Average Cs removal efficacy as a function of wash duration**

Duration (sec)	Asphalt	Brick	Concrete
5	50 ± 7%	34 ± 19%	31 ± 8%
15	61 ± 9%	43 ± 26%	42 ± 14%
20	71 ± 5%	46 ± 8%	55 ± 7%

The data in Table 5 show gradual increase of average Cs removal efficacy with increased wash duration for all three materials. The positive correlations between removal efficacy and wash duration were statistically tested. Student's  $t$ -tests for each material were conducted using statistical software (SAS version 9.2, SAS Institute, Cary, NC). The results are listed in Table 6. Initially, the statistical tests were conducted with 5%

significance, and many test resulted in significance at the 5 or 10% confidence level. To identify the potentially significant wash conditions, two test statistical significances are indicated by asterisks on the p values: \*\* for 5% and \* for 10% significance. The results show that there was an effect of duration of decontamination within asphalt samples. For asphalt, the five second duration yielded lower efficacy

values than either a 15 second duration ( $p = 0.0855$ ) or a 20 second duration ( $p = 0.0067$ ), and a 15 second duration yielded lower efficacy values than a 20 second duration ( $p = 0.0926$ ). For concrete, there was also an effect of duration, with the 5 second duration yielding lower efficacy values than

a 20 second duration ( $p = 0.0166$ ), but there were no statistically significant results between 5 and 15 and 15 and 20 seconds. Brick samples showed no statistically significant duration effect for all three durations.

**Table 6. Student's *t*- test results for the effectiveness of wash duration on Cs removal efficacy**

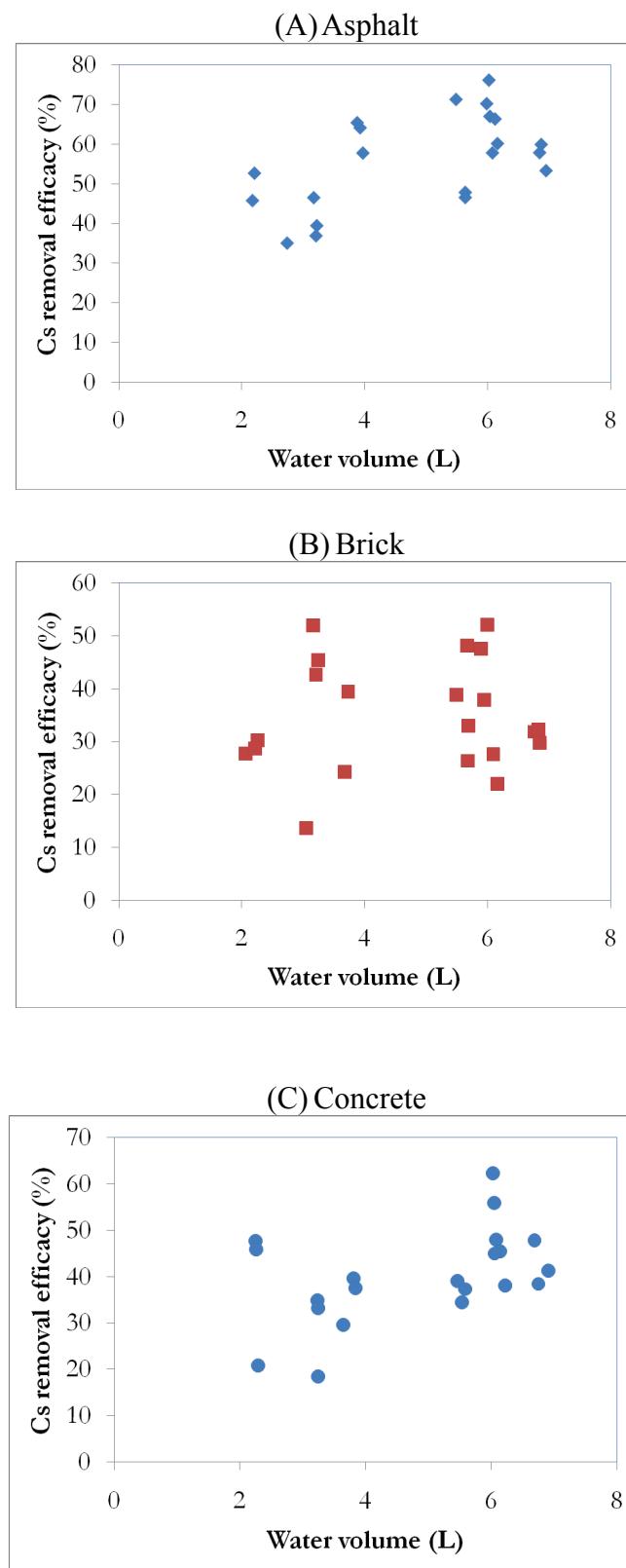
Testing	p-Value	Mean Difference	Sample Size
Asphalt			
5 sec vs. 15 sec	0.0855*	5 < 15, 11%	3 vs. 3
5 sec vs. 20 sec	0.0067**	5 < 20, 21%	3 vs. 3
15 sec vs. 20 sec	0.0670*	15 < 20, 8%	3 vs. 3
Brick			
5 sec vs. 15 sec	0.5664	Not sig. diff., 9%	3 vs. 3
5 sec vs. 20 sec	0.4672	Not sig. diff., 11%	3 vs. 3
15 sec vs. 20 sec	0.8709	Not sig. diff., 2%	3 vs. 3
Concrete			
5 sec vs. 15 sec	0.1862	Not sig. diff., 11%	3 vs. 3
5 sec vs. 20 sec	0.0166**	5 < 20, 25%	3 vs. 3
15 sec vs. 20 sec	0.1222	Not sig. diff., 13%	3 vs. 3

\*\* for 5% and \* for 10% significance

## Water Pressure

The average Cs removal efficacy as a function of wash pressure was tested, and the results are plotted versus the total water volume applied to the coupons (Figure 6). This water volume per unit time represents the water pressure applied to the coupon surface. The coupons were washed in the simulated firehose test chamber under the following conditions: wash duration 20 seconds and wash angle 90 degree. Each test was executed in triplicate. Some tests were conducted by restricting the nozzle orifice so that the total applied water volume per coupon surface area was less with the

higher water pressure. This lower coverage of water on the coupon surfaces was observed with 160 psi tests; the surface coverage was approximately 60 to 70% by visual inspection. Other tests were conducted by adjusting the input water pressure and keeping the same orifice size in the nozzle. Because the total applied duration is fixed at 20 seconds, the efficacy data were plotted as a function of total applied water volume. As seen in Figure 6, there is a clear tendency of increased efficacy with increased water volume for asphalt. However, the plots for brick and concrete samples are not clear for exhibiting this trend.



**Figure 6. Cs removal efficacy as a function of total applied water volume**

A linear regression analysis (SAS, version 9.2, SAS Institute, Cary, NC) for the Cs removal efficacy as a function of water volume was conducted for each material, and the results are listed in Table 7 with the test significance indicated by asterisks on the p-values: \*\* 5% and \* 10%. Water

volume was found to have a statistically significant positive effect on efficacy within “Asphalt” ( $p = 0.0080$ ) and “Concrete” ( $p = 0.0325$ ). However, there was no significant effect of water volume (pressure) within “Brick”. The slope was higher with the asphalt samples than the concrete samples.

**Table 7. Effect of water volume on Cs removal efficacy**

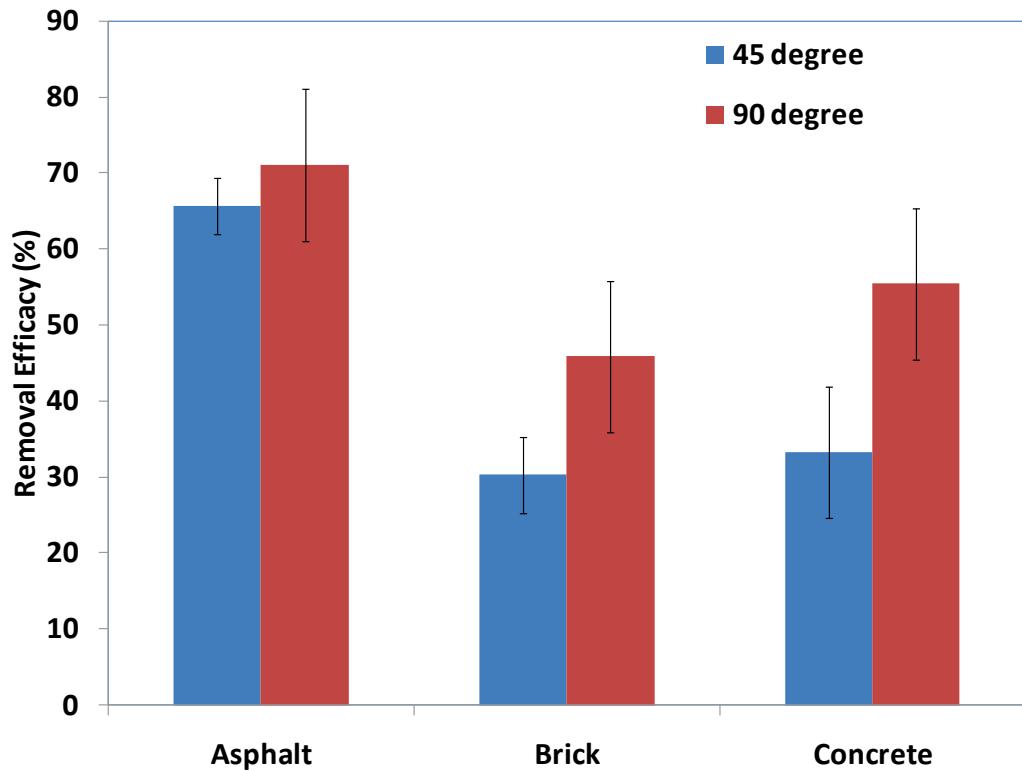
Material	p-Value	Slope (%/L)	95% Confidence Interval	Sample Size
Asphalt	0.0080 **	4.04	(1.18,6.90)	21
Brick	0.7255	0.50	(-2.46,3.48)	21
Concrete	0.0325 **	2.93	(0.27,5.59)	21

\*\* Test significance: 5%.

## Wash Angle

The average Cs removal efficacies from washing at 45 degree and 90 degree angles are shown in Figure 7. The error ranges in Figure 7 are one standard deviation for triplicate sample results. The coupons were washed in the simulated firehose test chamber under the following conditions: water pressure 120 psi for 20 seconds. A paired t-test (SAS, version 9.2, SAS Institute, Cary, NC) was used to compare average Cs

removal efficacy for 45 and 90 degree wash down tests. The test results showed that there was no statistically significant effect of wash angle within asphalt sample sets ( $p=0.3329$ ), but the  $45^\circ$  angle gave lower efficacy than the  $90^\circ$  angle with brick ( $p = 0.0595$ ) and concrete sample sets ( $p = 0.0350$ ). The mean difference of efficacies from 45 and 90 degree wash down were 13 and 18% for brick and concrete sample sets, respectively.

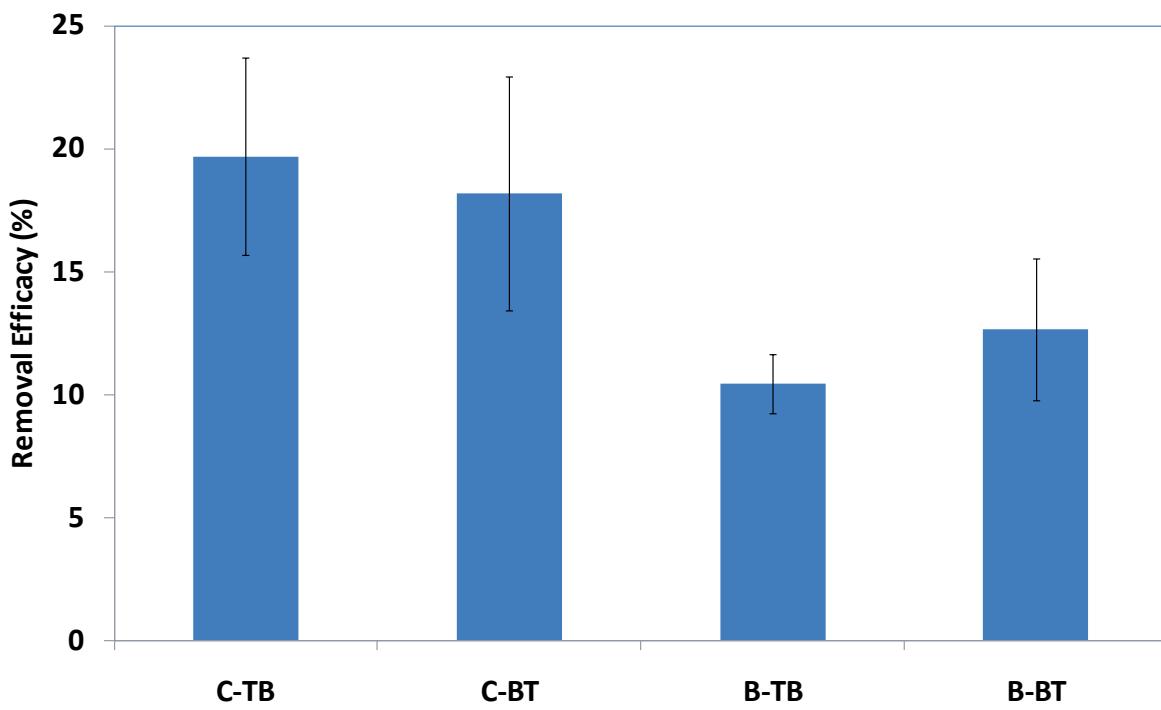


**Figure 7. Comparison of average Cs removal efficacy at wash angles 45 and 90 degrees**

## Wash Pattern

The Cs removal efficacy was studied using two different wash patterns with large (5" x 4" x 1") coupons. The coupons were washed in the simulated fire hose test chamber under the following conditions: water pressure 120 psi for 20 seconds at 90 degree wash angle. The coupons were washed in two different patterns: from bottom to top or from top to bottom. The wash pattern tests were used to evaluate the potential impact of pattern on vertical surface wash down efficiency. The results from the testing of brick and concrete samples are shown in Figure 8. A paired *t*-

test was used to compare the average Cs removal efficacies for two different wash patterns. Brick (two sample *t*-test,  $t = -1.2$ , 4 degrees of freedom,  $p=0.14$ ) and concrete samples (two sample *t*-test,  $t = 0.42$ , 4 degrees of freedom,  $p=0.35$ ) showed no statistical difference in their mean Cs removal efficacies for the two different wash patterns. The removal efficacy results from wash pattern tests were less than the small coupon tests. This is because wash pattern tests were conducted with coupons that were ~10 times larger than the ones used in the other tests; this resulted in wash water volume per unit area that was 10 times less than for the small coupons.



**Figure 8** Average Cs removal efficacy at different wash pattern: top to bottom wash (TB) and bottom to top wash (BT) for concrete (C) and brick (B) samples

## 6.0 Discussion

Different wash conditions for removal of Cs from three different urban surfaces using a simulated fire hose water delivery system were evaluated. Nonradioactive CsCl particles were aerosolized from a CsCl solution (dissolved in methanol) onto three different materials, asphalt, brick, and concrete. The tested parameters included wash duration, water pressure, wash angle, and wash pattern. The test results were analyzed to determine whether the variations in the wash down conditions have an impact on Cs removal efficacy.

The removal efficacies improved by increasing the wash duration (from 5 seconds to 20 seconds) from 50% to 70% for the asphalt samples, 34% to 43% for the brick samples and 31% to 55% for the concrete samples. Longer wash duration from three different wash duration tests showed a statistically significant increase in Cs removal efficacy for the asphalt samples with increase of wash duration. The concrete samples showed statistically significant increase for 20 seconds compared to 5 seconds, but the results for brick samples were not statistically significant for all three durations. The longest wash duration in this test was 20 seconds. Increasing the wash duration further may increase the effectiveness. However, increased water duration also increases the volume of water applied, which is one of the most important factors in the logistical requirements for this type of decontamination. In terms of the removal amount as a function of applied water volume, the five-second test showed the highest efficacy among three test durations for all three materials. The amount removed with the 5 second wash down was more than

50% of the total amount removed with a 20 second wash down.

For the water pressure test, Cs removal efficacies were measured as a function of water volume with fixed duration (20 sec). This water volume per unit time represents the water pressure applied to the coupon surface. Applied water volume and Cs removal efficacies were statistically tested for their correlations. Because some tests (tests with 160 psi) restricted the water nozzle orifice for pressure control, the water stream diameter decreased with increasing water pressure. As a result, the reduced water stream diameter may have resulted in less removal efficacy due to reduced coupon surface coverage. The impact of the stream diameter might be minimal because the Cs contamination is concentrated on the coupon center and the visual inspection during wash down confirmed that the water stream coverage area was centered and approximately 60 to 70% of the entire coupon surface. The results in Table 7 showed that the effectiveness of Cs removal increased with higher water pressure per applied water volume for asphalt and concrete samples, but not for brick samples. The study results imply that Cs is dominantly removed by water wash down from the test surface without physical removal of surface material itself. If the water pressure increases (e.g. by use of a pressure washer), the Cs removal efficacy may increase due to removal of surface material which Cs is binded.

Two different wash angles (45 and 90 degree) were tested for the impact on Cs removal efficacy with three materials (asphalt, brick, and concrete). The statistical analysis showed higher efficacy with 90

degree than 45 degree wash angles for the brick and concrete samples. The difference in average removal efficacy for the two different angles was approximately 15% and 21% for the brick and concrete samples, respectively. This result is related to the water pressure amount applied directly to the coupon surface. The angled water application reduced the applied pressure to a coupon surface. The pressure transducer read the pressure level at the 45 degree wash angle as approximately 32% of the pressure at the 90 degree wash angle. As seen in the water pressure test results, the reduced water pressure on coupon surfaces resulted in the low removal efficacy at the 45 degree wash angle. For asphalt coupons, the statistical results showed that the Cs removal difference from these two different wash angles was not significant. This asphalt coupon result does not follow the results from the water pressure tests. It is uncertain why asphalt samples did not show the higher removal efficacy with 90 degree wash angle.

Wash patterns were studied for impact on removal efficacy for the brick and concrete samples. The wash patterns evaluated were moving the hose from bottom to top and from top to bottom of a coupon. The removal efficacies from two different

patterns were tested statistically, and the results showed no effects on Cs removal for both materials. This result could be an artifact of using the small coupons, and this explanation needs to be confirmed with operational scale tests.

The current study was conducted as an exploratory test to probe any potential improvement on Cs removal efficacy from porous urban surfaces by adjusting water wash down conditions. The study demonstrated the effect of water wash down conditions on efficacy of Cs removal from porous urban surfaces. The Cs removal efficacy showed a positive correlation with wash duration and water pressure for the asphalt and concrete samples. The 90 degree wash angle was more effective than the 45 degree wash angle for the brick and concrete samples. Wash patterns (bottom to top and top to bottom) did not affect Cs removal efficacy for the materials tested. These findings are applicable only to the condition where Cs particles have limited penetration into porous surfaces. If Cs particles have penetrated into the subsurface via high RH or rain exposure prior to a water wash down, then the improvement by adjusting wash conditions may be reduced.

## Appendix

**Table A-1. Test conditions and efficacy results for asphalt coupons**

Sample ID	Wash Duration (sec)	Nozzle Pressure (psi)	Applied Water Volume (L)	Wash Angle (degree)	Wash Pattern	Wash Area (cm <sup>2</sup> )	Removal Efficacy (%)
A1	5	120	1	90	NA	9	45
A2	5	120	1	90	NA	9	58
A3	5	120	1	90	NA	9	48
A4	15	120	5	90	NA	9	60
A5	15	120	5	90	NA	9	54
A6	15	120	5	90	NA	9	69
A7	20	120	6	90	NA	9	67
A8	20	120	6	90	NA	9	76
A9	20	120	6	90	NA	9	70
A10	20	40	7	90	NA	9	53
A11	20	40	7	90	NA	9	60
A12	20	40	7	90	NA	9	58
A13	20	80	6	90	NA	9	60
A14	20	80	6	90	NA	9	66
A15	20	80	6	90	NA	9	58
A16	20	160	4	90	NA	9	64
A17	20	160	4	90	NA	9	58
A18	20	160	4	90	NA	9	65
A22	20	40	2	90	NA	9	46
A23	20	40	2	90	NA	9	53
A24	20	40	2	90	NA	9	35
A25	20	80	3	90	NA	9	47
A26	20	80	3	90	NA	9	40
A27	20	80	3	90	NA	9	37
A28	20	140	6	90	NA	9	48
A29	20	140	6	90	NA	9	71
A30	20	140	6	90	NA	9	47
A19	20	120	6	45	NA	9	66
A20	20	120	6	45	NA	9	64
A21	20	120	6	45	NA	9	72

**Table A-2. Test conditions and efficacy results for brick coupons**

Sample ID	Wash Duration (sec)	Nozzle Pressure (psi)	Applied Water Volume (L)	Wash Angle (degree)	Wash Pattern	Wash Area (cm <sup>2</sup> )	Removal Efficacy (%)
B1	5	120	2	90	NA	9	54
B2	5	120	2	90	NA	9	31
B3	5	120	2	90	NA	9	19
B4	15	120	5	90	NA	9	27
B5	15	120	5	90	NA	9	32
B6	15	120	5	90	NA	9	72
B7	20	120	6	90	NA	9	38
B8	20	120	6	90	NA	9	48
B9	20	120	6	90	NA	9	52
B10	20	40	7	90	NA	9	32
B11	20	40	7	90	NA	9	32
B12	20	40	7	90	NA	9	30
B13	20	80	3	90	NA	9	14
B14	20	80	6	90	NA	9	22
B15	20	80	6	90	NA	9	28
B16	20	160	6	90	NA	9	48
B17	20	160	4	90	NA	9	24
B18	20	160	4	90	NA	9	39
B28	20	40	2	90	NA	9	28
B29	20	40	2	90	NA	9	30
B30	20	40	2	90	NA	9	29
B31	20	80	3	90	NA	9	52
B32	20	80	3	90	NA	9	45
B33	20	80	3	90	NA	9	43
B34	20	140	6	90	NA	9	26
B35	20	140	6	90	NA	9	39
B36	20	140	6	90	NA	9	33
B19	20	120	6	45	NA	9	36
B20	20	120	6	45	NA	9	35
B21	20	120	6	45	NA	9	27
B22	20	120	7	90	TB	129	11
B23	20	120	7	90	TB	129	9
B24	20	120	7	90	TB	129	11
B25	20	120	7	90	BT	129	16
B26	20	120	7	90	BT	129	11
B27	20	120	7	90	BT	129	11

**Table A-3. Test conditions and efficacy results for concrete coupons**

Sample ID	Wash Duration (sec)	Nozzle Pressure (psi)	Applied Water Volume (L)	Wash Angle (degree)	Wash Pattern	Wash Area (cm <sup>2</sup> )	Removal Efficacy (%)
C1	5	120	1	90	NA	9	29
C2	5	120	1	90	NA	9	24
C3	5	120	2	90	NA	9	39
C4	15	120	5	90	NA	9	51
C5	15	120	5	90	NA	9	28
C6	15	120	5	90	NA	9	47
C7	20	120	6	90	NA	9	62
C8	20	120	6	90	NA	9	56
C9	20	120	6	90	NA	9	48
C10	20	40	7	90	NA	9	48
C11	20	40	7	90	NA	9	41
C12	20	40	7	90	NA	9	38
C13	20	80	6	90	NA	9	45
C14	20	80	6	90	NA	9	38
C15	20	80	6	90	NA	9	45
C16	20	160	4	90	NA	9	30
C17	20	160	4	90	NA	9	40
C18	20	160	4	90	NA	9	38
C28	20	40	2	90	NA	9	21
C29	20	40	2	90	NA	9	48
C30	20	40	2	90	NA	9	46
C31	20	80	3	90	NA	9	18
C32	20	80	3	90	NA	9	35
C33	20	80	3	90	NA	9	33
C34	20	140	6	90	NA	9	39
C35	20	140	6	90	NA	9	37
C36	20	140	6	90	NA	9	34
C19	20	120	6	45	NA	9	45
C20	20	120	6	45	NA	9	33
C21	20	120	6	45	NA	9	34
C22	20	120	7	90	TB	129	17
C23	20	120	7	90	TB	129	24
C24	20	120	7	90	TB	129	18
C25	20	120	7	90	BT	129	13
C26	20	120	7	90	BT	129	20
C27	20	120	7	90	BT	129	22

## References

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- <sup>1</sup> Gonzalez, A.J. (2003) Security of radioactive sources: threats and answers. In *International Conference on Security of Radioactive Sources*. pp.33-58. Vienna, Austria: International Atomic Energy Agency.
- <sup>2</sup> NRC (May 2007) Backgrounder on Dirty Bombs: U.S. Nuclear Regulatory Commission.
- <sup>3</sup> Zimmerman, P.D. and Loeb, C. (2004) Dirty Bombs: The threat revisited. *Defense Horizons*, pp.1-11.
- <sup>4</sup> Karam, P.A. (2005) Radiological terrorism. *Hum Ecol Risk Assess*, **11**, 501-523.
- <sup>5</sup> Rosoff, H. and von Winterfeldt, D. (2007) A risk and economic analysis of dirty bomb attacks on the ports of Los Angeles and long beach. *Risk Anal* **27**, 533-546.
- <sup>6</sup> Roed, J. (1987). Run-off from the weathering of roof material following the Chernobyl accident. *Radiation Protection Dosimetry*, **21** (1/3), 59-63.
- <sup>7</sup> Roed, J. (1988). The Distribution on Trees of Dry Deposited material from the Chernobyl Accident. Paper NKA/AKTU-245 988)2, presented at the Joint CEC/OECD (NEA) Workshop on Recent Advances in Reactor Accident Consequence Assessment, Rome, Italy, 25-30 January 1988.
- <sup>8</sup> Karlberg, O. (1988). Weathering and Migration of Chernobyl Fallout in Sweden. Studsvik, Sweden, Report NP 216 EA.
- <sup>9</sup> Andersson, K.G., J. Roed, and C.L. Fogh. (2002b). Weathering of radiocaesium contaminationon urban streets, walls and roofs. *Journal of Environmental Radioactivity*, **62**, 49-60.
- <sup>10</sup> U.S. Environmental Protection Agency (U.S. EPA). 2012. Fate of Radiological Dispersal Device (RDD) Material on Urban Surfaces: Impact of Rain on Removal of Cesium, Washington, D.C., EPA/600/R/12/569.
- <sup>11</sup> U.S. Environmental Protection Agency (U.S. EPA). 2010. Radiological Dispersal Device Outdoor Simulation Test: Fate of Cs on Limestone, Washington, D.C., EPA/600/X/10/005 (FOUO).
- <sup>12</sup> Gusarov, A., Il'icheva, N., Konoplev, A., Lee, S.D., Maslova, K., Popov, V., and Stepina, I. (2011) Fate and Transport of radiocesium in urban building materials. International Conference on Radioecology & Environmental Radioactivity: Environment & Nuclear Renaissance **46**(6), S265-S269.
- <sup>13</sup> U.S. Environmental Protection Agency (U.S. EPA). 1994. Determination of Trace Elements in Waters and Wastes by Inductively Coupled Plasma-Mass Spectrometry, Washington, D.C. Method 200.8 revision 5.4.

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