Design of Ad Hoc Filtration Beds for Treating Contaminated Waste Waters – 18385

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ABSTRACT

We employed the Gold Sim Contaminant Transport model to design a system of reactive filtration beds that may be used to selectively remove radioactive cesium and other radionuclides from contaminated waters. These filtration beds can be rapidly constructed ad hoc by using common materials that permit timely, in-field treatment of incoming contaminated waters generated during the response and recovery phase of a radioactive release event. Sensitivity analyses on the system parameters revealed simple relationships between the bed design and the expected breakthrough of cesium, allowing us to generate a concise set of look-up tables to aid the early responders.

INTRODUCTION

The probability of a terrorist attack with a radionuclide dispersal device (RDD) is unknown, but response and recovery plans should be created beforehand to minimize the detrimental economic, social, and psychological effects of any widespread nuclear contamination event. Recovery plans/guidance have been published (e.g., [1,2]), but most have been limited in scope to the European and UK communities [3]. The ongoing recovery effort following the disaster at the Fukushima Daiichi nuclear power station serves as a reminder of the difficulties in developing a response effort post-event. Earlier studies have evaluated the known strategies in the event of widespread radiological dispersal [3,4] and their application to the United States recovery landscape. Two high-priority gaps are the lack of site-specific response/recovery plans and waste management strategies.

At WM2016, the Integrated Wash-Aid, Treatment, and Emergency Reuse System (IWATERS) was reported as a quick, non-destructive wide-area remediation technique suitable for the urban environment [5]. With this system, an ionic wash solution (“Wash Aid”) is applied by fire hose to the contaminated surfaces (buildings, roadways, or vehicles). To improve decontamination efficacy [6], simple potassium, ammonium, or sodium-based salts and low concentrations of surfactant can be added in-line to fire hose operations by use of common eductors employed to incorporate foaming agents. The contaminated runoff is contained in reservoirs made from flood control barriers and passed into filtration basins containing active sorbents for the radioactive species in solution. Ideally, the sorbents would be chosen from locally available options such as common clays, soils, or crushed stone and gravel. The effective partition coefficient of the mixture will dictate the dimensions of the bed, the volume of water that can be processed before breakthrough, and the rate at which water can be processed (Fig. 1). Several beds may be needed. For final waste clarification, a mobile trailer can be employed before sending the treated water to a portable tank or reservoir for reuse via common firefighting drafting methods.

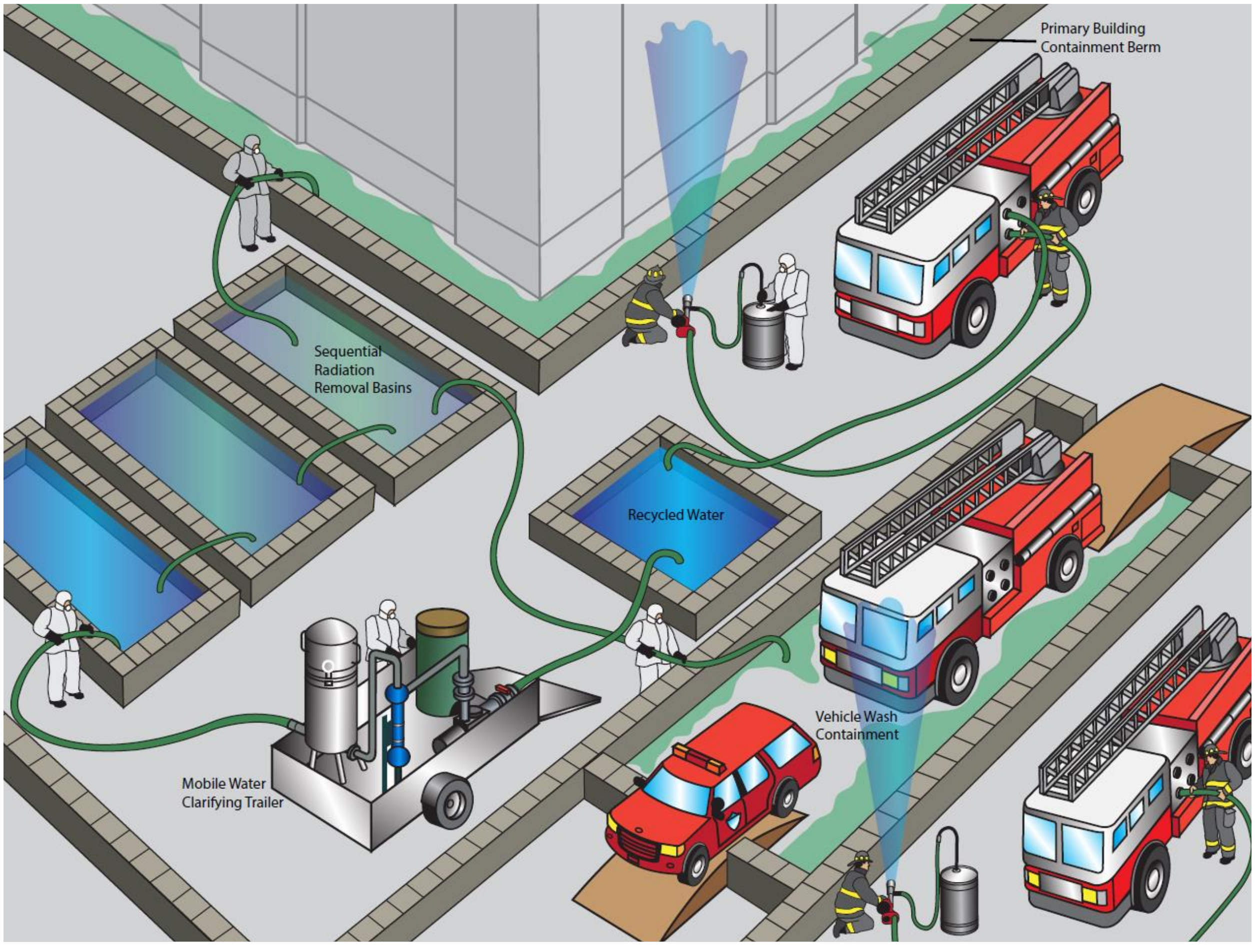

Figure 1. Artist's rendition of potential deployment of the IWATERS for large (building) and small-scale (vehicle) nuclear contaminations. Additives are drawn via an eductor into a firehose to promote removal of contaminants from the surfaces. This contaminated water (green) is contained and transferred through several filtration beds containing solid sorbent materials (i.e., sequestering agents) before being clarified in a mobile filtration trailer and recycled for continued decontamination operations.

A key aspect of the IWATERS is design of the reactive filtration beds. GoldSim is a reactive-transport model that is capable of probabilistic and deterministic simulations with an easy-to-use interface. It is used by a variety of research institutions and commercial groups across more than ten countries such as DOE, NRC, and NASA [7]. We used the GoldSim Contaminant Transport Model to design these filtration beds and demonstrated their utility as a means of treating contaminated waters on site. By extension, GoldSim can be used to generically design ad hoc filtration beds for a variety of scenarios, including chemical and radioactive contamination, and can be coupled to external programs (e.g., decontamination efficacy, dosimetry, and supply chain analysis), as part of a much broader effort to develop realistic simulations and virtual exercises to compress the recovery phase timeline.

METHODS

Column flow experiments

For this study, we focused on a mixture of sand and vermiculite clay as a bed designed for cesium contaminations [6]. Masses of vermiculite (250 mg) and sand (100 g) were mixed and dry packed into a column (2.54 cm ID x 25.4 cm length), and a solution containing 0.14 μCi/L of both ^{137}Cs and ^{85}Sr in tap water was passed continuously. Samples were collected every 20-30 min and counted by using a NaI gamma detector as described elsewhere. These experiments are detailed in a prior publication [8]. The resulting eluent profile was compared to the GoldSim model results that were based on the hydraulic properties of the media and the batch sorption coefficient (K_d) measured in our laboratory [8]. The agreement was good in each case and allowed us to extrapolate the bed sizes and water volumes described herein.

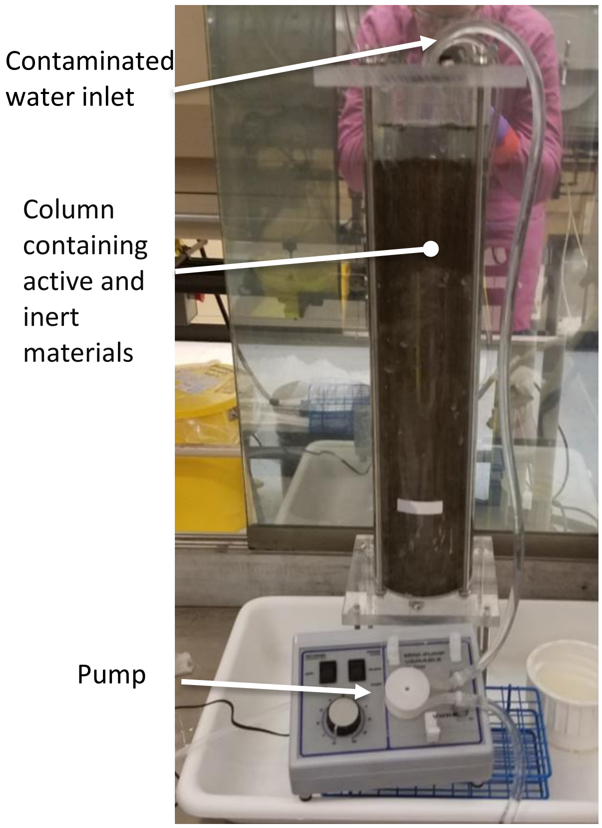

Figure 2. Photograph showing experimental setup where ^{137}Cs spiked water was gravity fed through a column of mixed sorbents. The pump supplies a fixed head of water above the column.

Modeling the treatment of contaminated water

In GoldSim, "elements" when linked together can model complex scenarios. In a 2016 study, experiments were performed to validate a GoldSim model for a sand/clay bed system with ^{137}Cs, ^{90}Sr, and ^{152}Eu [8]. The key element in the model is the Aquifer, which models flow in pipes, rivers, and saturated/unsaturated vertical columns [7]. For the recycling of a used wash solution, the model partitions the radionuclides between the sand/clay bed and the fluid. Aquifer is used to create a series of cells connected by mass flux links based on user inputs [9]. This connection of cells is referred to as a "cell net pathway". The starting mass balance equation for a cell net pathway (including the Aquifer) is represented as:

$$m'_{is} = -m_{is}\lambda_s + \sum_{p=1}^{NP_s} m_{ip}\lambda_p f_{ps} R_{sp}\left(\frac{A_s}{A_p}\right) + \sum_{c=1}^{NF_i} f_{cs} + S_{is} \ , \qquad \text{(Equation 1)}$$

which can be simplified to

$$m'_{is} = \sum_{c=1}^{NF_i} f_{cs} + S_{is} \qquad \text{(Equation 2)}$$

where

m'_{is} is the rate of mass increase in cell *i* of species *s* (e.g., $^{137}Cs^{+}$ or $^{90}Sr^{2+}$),
f_{cs} is the mass rate into cell *i* of species *s* through link *c* from all mass flux units *NF* linked to cell *i*, and
S_{is} accounts for "external" source inputs [9].

The second term on the right-hand side of Eqn. (1) accounts for decay of parents *p* into species *s*, but because the chemical or radiological decay constants λ for the species of interest is negligible in the time scale of the simulation, all other terms in Eq. (1) are ignored. Our model (see snapshot of graphic user interface in Fig. 3) requires an input of the "ExperimentalFactors" (head height of water), "Columnspecs" (height and surface area of the sorbent bed), SolidCharacteristics (composition of bed and the bulk density, permittivity, and porosity of its components), "Radionuclide" (sorbent-dependent sorption coefficients K_d), "Water" (diffusivity), the "Species" to be tracked (^{137}Cs), and the concentration of incoming species ("ContinConcentrations" for continuous feed at a given concentration or "SpikeConc" for a single injection of a concentrated bolus). With this input, the model can calculate the gravity-fed flow velocity and the time-dependent concentration of radionuclides in the bed and effluent.

By varying these input factors using the sensitivity analyses option within the software, GoldSim produces output plots that allow us to model the output dependencies on these input parameters. From these plots, we can generate look-up tables that are easy to decipher by the non-specialist and can be further developed to provide in-field guidance tables or phone applications that will allow the user in the field to instruct responders on how to construct the filtration beds.

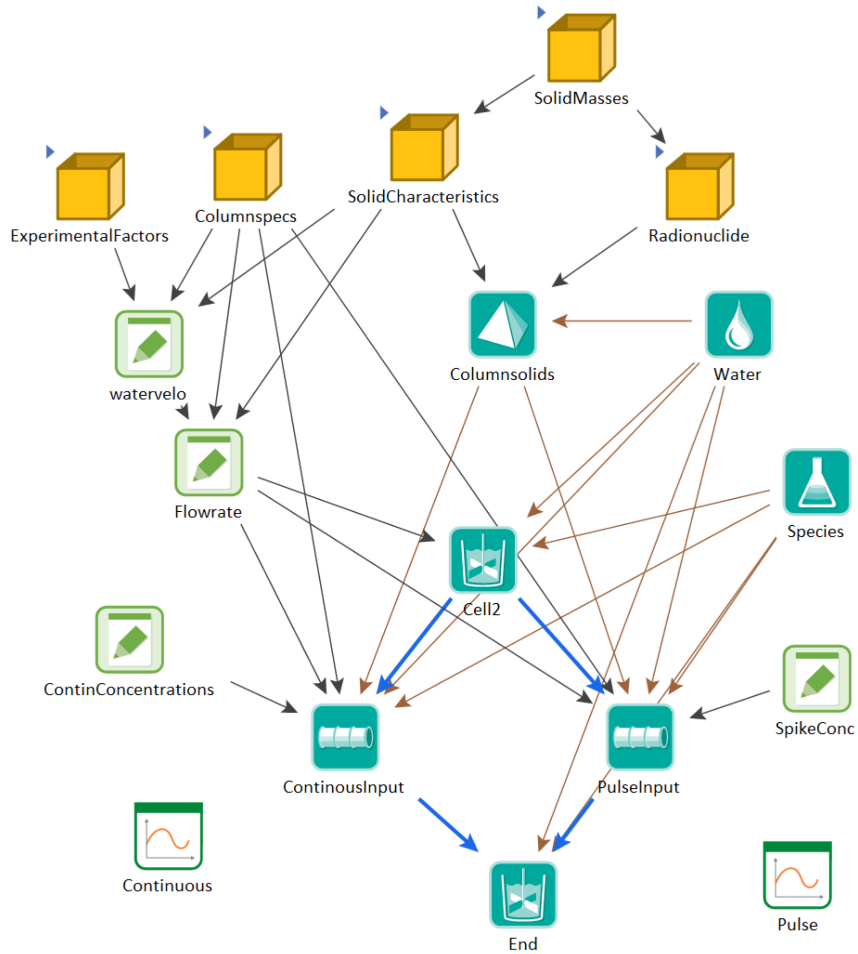

Figure 3. Flowsheet for GoldSim CT, which provides a probabilistic and deterministic simulation with an easy-to-use interface.

DISCUSSION

We initially performed a check of the model output to the experimental data to verify the accuracy of the model algorithm for a variety of sorbents and flow conditions [8]. Having established good agreement between data and model, we began exploring the design of scale-up filtration bed designs.

Generating look-up tables

We focused on removal of cesium contamination into beds composed of a mixture of vermiculite clay (as the active sorbent) and sand (to provide enhanced water permeability). From the model outputs, we performed sensitivity analyses to determine the effects of changing the composition of the bed (sand:clay ratio), the flow velocity of water through the column, the sorption coefficient of the active sorbent (in this case, the vermiculite clay), etc. The relationships were linear (example shown in Fig. 4). This facilitated the generation of look-up tables using linear extrapolations in most cases from a baseline run of the program.

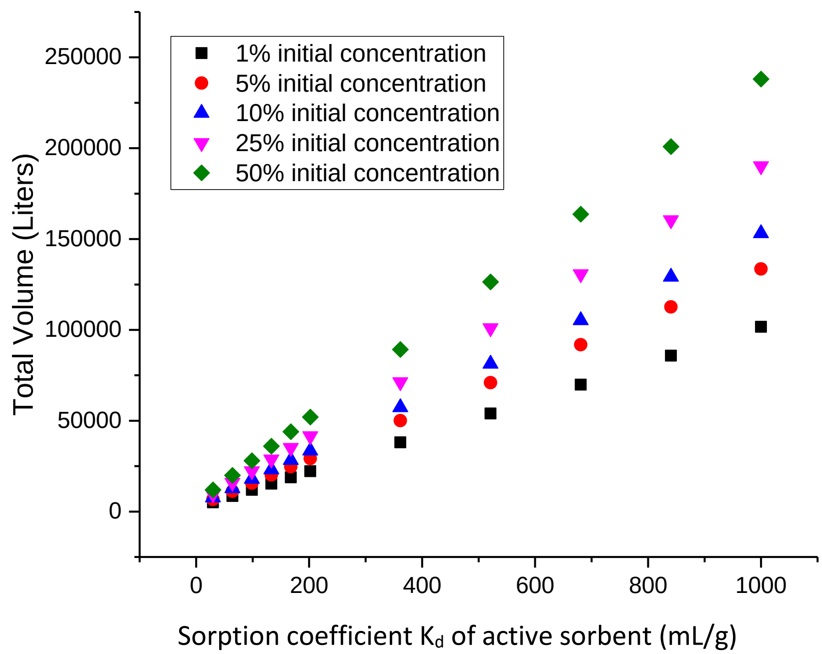

Figure 4. Total treated volume of wash solution vs. effective K_d of the active sorbent for a target breakthrough concentration expressed as a percentage of initial concentration of ^{137}Cs.

Look-up Tables 1 and 2 show important output values for processing cesium in tap water and salty water, respectively, for the baseline case of a 9.29 m^2 (100 ft^2) bed containing a sand/clay mixture with depth of 0.305 m (1 ft). For a 50:50 mixture, processing of 540,000 L (143,000 gallons) will take 92 hours, which will deposit 54 mCi of cesium into the bed, before breakthrough of the bed occurs (breakthrough value set at the drinking water standard of 1 pCi/L). A 90:10 sand:clay mix increases the permeability of the bed so that 129,000 L (34,000 gallons) can be processed in 12 hours, depositing 13 mCi of ^{137}Cs before breakthrough. Breakthrough occurs much earlier in a bed processing the potassium salt water (Table 2) due to competition for sorption sites by the K^+ ions. For example, a 90:10 mixture in this case can process 7600 L (2000 gallons) in under an hour, sorbing less than a millicurie before breakthrough.

Table 1. Baseline technical look-up table for constructing ad hoc filtration beds for processing cesium-contaminated fresh (tap) water.

Wash Water: Tap				
Sand:Clay				
Ratio ↓	Hours	mCi	Gallons	Gal/min
50:50	91.9	53.95	142571	25.9
60:40	64.0	45.26	119602	31.1
70:30	42.9	35.53	93897	36.5
80:20	26.1	24.75	65414	41.8
90:10	12.1	12.93	34206	47.2

Table 2. Baseline technical look-up table for constructing ad hoc filtration beds for processing cesium-contaminated salt (0.1 M KCl) water.

Wash Water: 0.1 M KCL				
Sand:Clay				
	Hours	mCi	Gallons	Gal/min
50:50	5.1	3.005	7965	25.8
60:40	3.6	2.534	6726	31.0
70:30	2.4	2.001	5322	36.3
80:20	1.5	1.408	3762	41.6
90:10	0.7	0.768	2076	47.0

Using the linear relationships derived from the sensitivity analyses, we can construct another table (Table 3) that allows the user to estimate the bed size needed. In this example, we start with the baseline case for salty water (0.1 M KCl) shown in Table 2. The main attributes that can be changed are the size of the bed in units of feet squared and the depth of the bed in units of feet. Row 1 in the upper part of Table 3 represents the mathematical factor that must be applied for every 100 ft^2 of surface area added. Row 2 represents the mathematical factor that needs to be applied for each foot of depth that is increased. The lower part shows an example calculation for a bed with a surface area of 400 ft^2 (37.16 m^2) and a depth of 3 ft (0.91 m). In this example, the bed has a four times larger surface area than the base case and, therefore, can retain four times the quantity of ^{137}Cs, can process four times the total gallons of water, and can process this solution at four times the rate. However, the depth is also increased from one to three feet. Therefore, the hours that the bed can be used increases by 4 x (3 - 1) = 8. The activity of ^{137}Cs that can be retained increases by another factor of 2.5 (3 - 1) = 5. The total gallons that can be processed in that time increases by another factor of 2.5 (3 - 1) = 5, and the rate increases by another factor 1/(0.8 (3 - 1) = 0.625. Overall, the time before breakthrough increase by a factor of 8, the activity retained in the bed and total volume by a factor of 20, and the rate by a factor of 2.5.

Table 3. Technical look-up table for constructing a 400’ x 3’ ft ad hoc filtration bed for processing different volumes of total quantities of cesium in contaminated-salt (0.1 M KCl) water.

	Hours	mCi	Gallons	Gal/min
SA ↑	No Effect	multiply by Total Sqft/100		
Depth ↑	x4(Depth-1)	x2.5(Depth-1)	x2.5(Depth-1)	/.8(Depth-1)

Wash Water: 0.1 M KCl	400 x 3			**Table 2**
Sand:Clay Ratio ↓	Hours	mCi	Gallons	Gal/min
50:50	41.1	60.1	159301	64.4
60:40	28.8	50.68	134516	77.6
70:30	19.5	40.02	106435	90.8
80:20	12.0	28.16	75236	104.1
90:10	5.9	15.36	41517	117.5

We are further simplifying these tables into an Excel spreadsheet that asks the user simple questions about how much water they expect to process over what period of time and what sorbent materials are available. It will then provide the output specifications for the bed. We expect that such an algorithm can be adapted easily into a phone application that could be implemented in the field.

With these tables, we present example scenarios and describe the types of beds and expected performance (volume of water treated, breakthrough concentrations, and total treatment times) for small filtration beds that might be used to clean runoff from contaminated roofs to those suitable for city-wide wash-down activities that we demonstrated in the past.

Field demonstrations

IWATERS was demonstrated in two full-scale events to demonstrate the logistics of construction and operation (no radionuclides were used). The first was at the Wide Area Recovery and Resiliency Program (WARRP) in Denver (2012), and the second was at the Environmental Protection Agency's and Department of Homeland Security's Technology Demonstration in Columbus (2015). In both cases, we focused on the design for radioactive cesium applications. Key components were demonstrated:

- Used commercial-off-the-shelf equipment and reagents
- Provided wash water additives in-line
- Captured contaminated water with flood control barriers
- Simulated removal of radionuclides with ad-hoc filtration basins
- Reused the processed water using drafting techniques.

We also demonstrated the use of rain barrels to clean contaminated water generated during roof wash-down activities. The 55-gallon drum (Fig. 5) can process >7000 gallons (26,500 L) of tap water or >400 gallons (1500 L) of 0.1 M KCl in tap water at a rate of up to 80 gal/h (300 L/h) before breakthrough occurs.

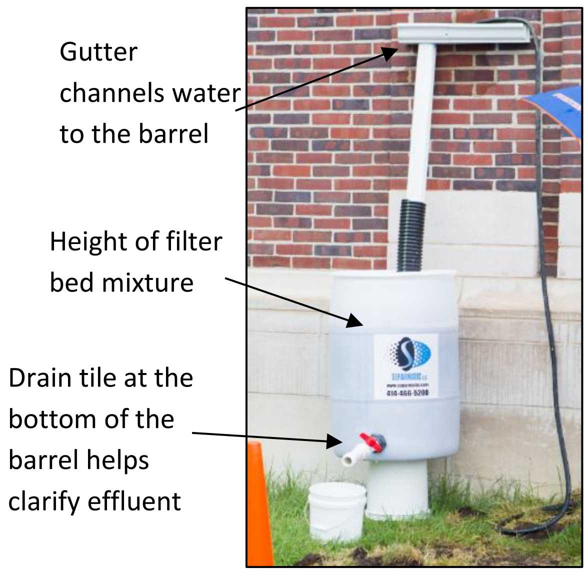

Figure 5. Rain barrel filter design for treating roof runoff. This barrel contained a 70:30 mixture of sand:clay (2.75 ft^2 x 2 ft depth) capable of processing >7000 gallons of tap water with cesium, or >400 gallons of 0.1 M KCl in tap water.

A vehicle-accessible reservoir (Fig. 6) was used to collect the contaminated wash from egressing vehicles in a contaminated zone. The water flowed and mixed with the sorbent (or passed through the bed into a drain tile buried under the sorbent mixture). This 70:30 sand:clay bed can process >500,000 gallons (1.9 x 10^6 L) of tap water or >31,000 gallons (117,000 L) of 0.1 M KCl in tap water at a rate of 13,000 gal/h (49,000 L/h).

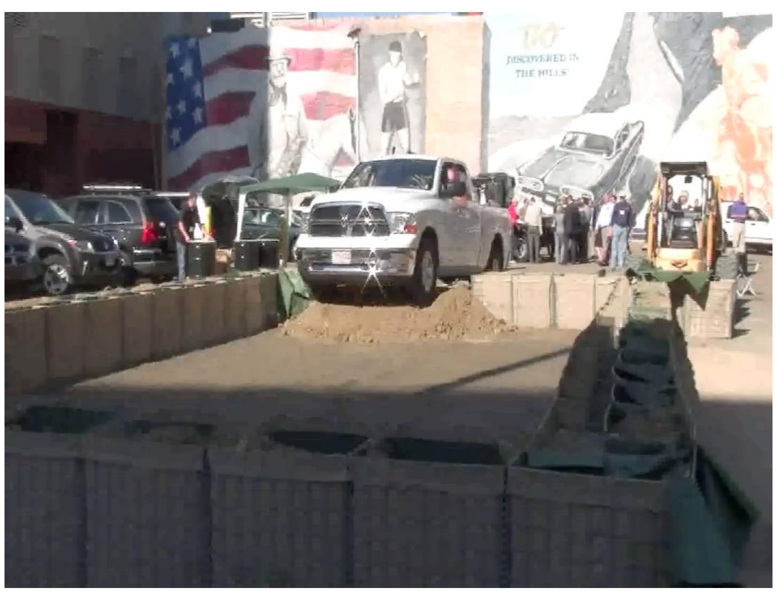

Figure 6. Vehicle decontamination basin. This reactive filtration basin can be used to treat contaminated runoff from vehicle decontamination activities. The flood barriers are designed to be driven over to allow vehicle entry. This bed, containing a mixture of 70:30 sand:clay (600 ft^2 x 1 ft depth), can treat >500,000 gallons of tap water with cesium or >31,000 gallons of 0.1 M KCl in tap water

before experiencing breakthrough.

A building reservoir (Fig. 7) was designed to capture contaminated runoff from building wash-down activities. A tarp was fastened to the façade to direct runoff into the reservoir. This bed can process >1.4 million gallons (5.3×10^6 L) of tap water or >80,000 gallons (300,000 L) of 0.1 M KCl in tap water at a rate of 15,000 gal/h (57,000 L/h).

Figure 7. Building runoff basin. By attaching tarps to the building façade using wood strips and an powder actuated fastener, the runoff from building wash-down activities can be collected in the basin. This basin was filled with 70:30 sand:clay (540 ft^2 x 1 ft depth), which can treat >1.4 million gallons of tap water with cesium or >80,000 gallons of 0.1 M KCl in tap water before breakthrough.

CONCLUSIONS

IWATERS can decontaminate buildings, roadways, vehicles, aircraft, runways, etc., using tap water and tap-water-containing mild additives, depending on the type of contamination. The resulting decontamination waste water can be treated on-site by a combination of natural sorbents and off-the-shelf filter systems for a variety of nuclear and radiological contaminants. Our modeling results suggest that ad hoc IWATERS designs could treat millions of gallons of contaminated waters suitable for reuse. Simple ad hoc systems can also be used to increase the resilience of recovery operations by providing immediate means of decontaminating and treating contaminated waters, even in water-scarce areas.

The approach we describe can be applied generically to all types of radionuclide contaminants, provided the sorption coefficient (K_d) is known. Then, look-up tables can be used to provide immediate specifications on the design of suitable filtration beds. In this way, the user can design filtration beds to effectively remove radionuclides in contaminated waters based on currently available materials, such as clays, soils, gravel, crushed lime, and specialized sorbents. With such information, we hope to mitigate the effects of generating large volumes of contaminated waters and provide a means of rapidly initiating decontamination activities to restore critical infrastructure and the urban environment. We continue evaluating the IWATERS for contaminants to suggest options for

- Increasing the decontamination of various radionuclides and surfaces.
- Treating the wastewater with readily available reactive materials, given the geographic or logistical constraints of a particular situation.
- Optimizing designs of the entire system and providing guidance on site-specific implementation.
- Integrating the treatment of biological and chemical threat agents to IWATERS using a similar methodology with an effective sorption coefficient.

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