

Summary of the Transport of Cesium in the Environment

INTRODUCTION

The U.S. Environmental Protection Agency's (EPA's) Homeland Security Research Program (HSRP) is assessing strategies and methodologies for remediation of areas contaminated with radionuclides. Most of EPA's research efforts have focused on Cesium (Cs), an extremely mobile and difficult to clean up radionuclide, with the ^{137}Cs and ^{134}Cs being the predominant isotopes. Cs is a likely contaminant that would result from its use (probably in the form of cesium chloride [CsCl]) either in a radiological dispersal device, from its release or generation through decay processes from nuclear power plant accidents, or from its release from an improvised nuclear device. The EPA would likely be involved with remediation of areas contaminated after one of these types of radiological incidents. The resulting contamination could be potentially widespread, involving many square miles urban, agricultural, and forested areas.

Because of its initial mobility right after deposition, knowledge of the transport of Cs in the environment is crucial to inform cleanup and sampling strategies. EPA's Homeland Security Research Program has conducted a series of experimental studies and has acquired field data from the Fukushima and Chernobyl accidents focused on assessing how Cs moves within the environment following an intentional or accidental release. This information on the mobility of Cs (see Figure 1) falls into several areas, all of which are important to simultaneously consider following a radiological incident, including:

- Cs interaction with urban surfaces under ambient conditions
- Effect of weathering on Cs-contaminated surfaces
- Cs movement within soil
- Cs movement in bodies of surface water
- Cs uptake into vegetation
- Cs movement within water and wastewater infrastructure
- Cs movement due to wildfires in contaminated forest land
- Cs movement due to precipitation events.

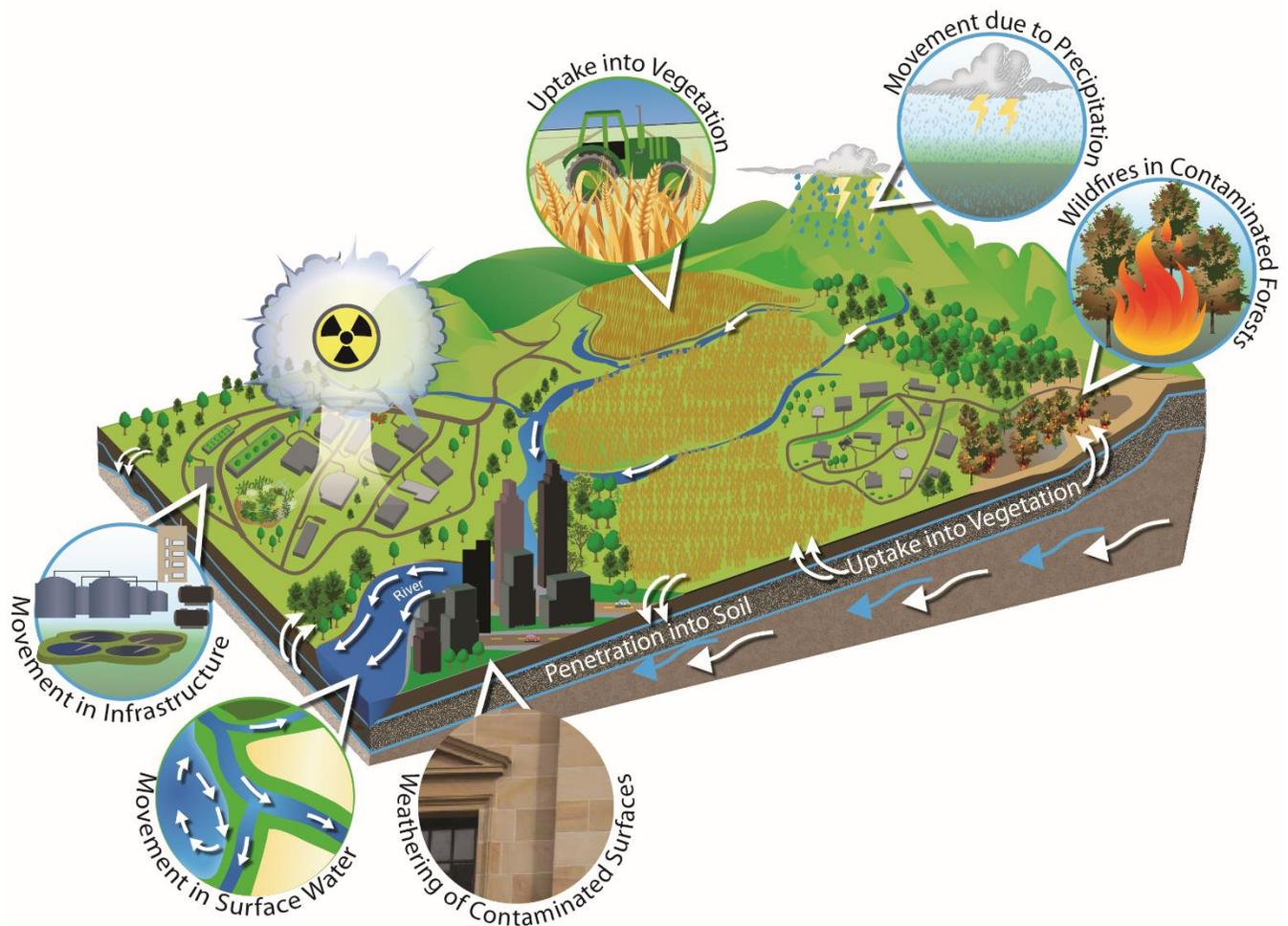


Figure 1: Mobility of cesium.

CESIUM INTERACTIONS WITH URBAN SURFACES UNDER AMBIENT CONDITIONS

CsCl, even if it is deposited as a dry particle, will eventually become aqueous when exposed to relative humidity (RH) above 67% or to rain. Cs-containing aqueous droplets can easily be transported through the pores of porous materials or through surface cracks. Once Cs ions (Cs hereafter) have migrated into these materials, Cs can be difficult to efficiently extract without removal of the affected surface. The adsorption of the Cs to the porous materials also hampers its ability to be extracted using existing chemically based decontamination technologies.

The ability to remove Cs from surfaces depends on the ability to reach the contamination. This ability is partially governed by the penetration depth of the Cs contamination. To experimentally assess the migration of Cs in its aqueous form into urban materials, water solutions of Cs were aerosolized onto the surfaces of five study materials: asphalt, brick, concrete, limestone, and granite.

The migration was determined by measuring Cs penetration depths as a function of high and low RH. Results from the 87% RH experiments are shown in Table 1 (U.S. EPA, 2014b).

Table 1. Penetration of Cs into Urban Surfaces at 87% Relative Humidity

Contaminant-Surface Interaction Time (days)	Penetration Depth* (mm)				
	Asphalt	Limestone	Granite	Concrete	Brick
1	0.2	0.7	2.0	0.5	0.8
7	0.4	1.7	0.2	0.7	0.8
14	0.4	1.3	3.5	0.5	0.7
28	0.6	1.4	2.3	2.6	1.1

Sources: Gusarov et al., 2011; Maslova et al., 2013; Full references are at the end of the brief.

*Penetration depths (mm) indicate where 90 % of the cesium was found

** Average of two data points – standard deviation not available.

From these studies, it was concluded that the Cs penetration depth profile primarily depends on the type of building material and the heterogeneity of the sample surface. These studies also demonstrated that the penetration depth was not strongly affected by the contaminant-surface interaction time for up to 28 days; operationally this would be similar to the time between contamination and cleanup. The ability to remove Cs from these surfaces also depends on the ability of external forces (either man-made such as decontamination technologies, or natural, such as precipitation events) to desorb the Cs from the material or to extract the Cs before it develops strong binding to the material. This ability is partially governed by the Cs adsorption. The Cs sorption characteristics were determined by spiking water solutions of Cs onto the urban surfaces. These studies showed that sorption of Cs increased with time (from 1 to 28 days) for the five materials studied. The maximum degree of sorption was seen after 24 hours of interaction for concrete, limestone and brick, while for asphalt and granite it occurred after 6 days. These sorption test results also suggested that asphalt is the material with the highest sorption capability. Lastly, to assess the properties to desorb the Cs, the ability to extract Cs with competing ions was tested. This extraction ability increased in the following order, from greatest to least: brick > granite > limestone > concrete > asphalt.

EFFECT OF WEATHERING ON CS-CONTAMINATED SURFACES

It is known that weathering can reduce the contamination on surfaces, although the depth of penetration of the Cs into the surface may impact the ability of weathering to reduce contamination. Based on limited experimental studies, penetration depth of Cs into the building materials was in the following order, from greatest to least: limestone > brick > concrete = asphalt = granite. To assess the impacts of the weathering process on Cs contamination, the amount of Cs removed from urban surfaces (asphalt, brick, concrete, limestone, and granite) and the amount of Cs that penetrated the building materials after a simulated rain event (average 2 cm per hour for 30 min) was determined. The coupons were contaminated by spiking them with water solutions of Cs. The percent of Cs removed is shown in Figure 2. These results suggest that it would be more difficult to remove Cs from some materials such as brick, concrete, and limestone after a rain event than prior to the event due to greater subsurface penetration.

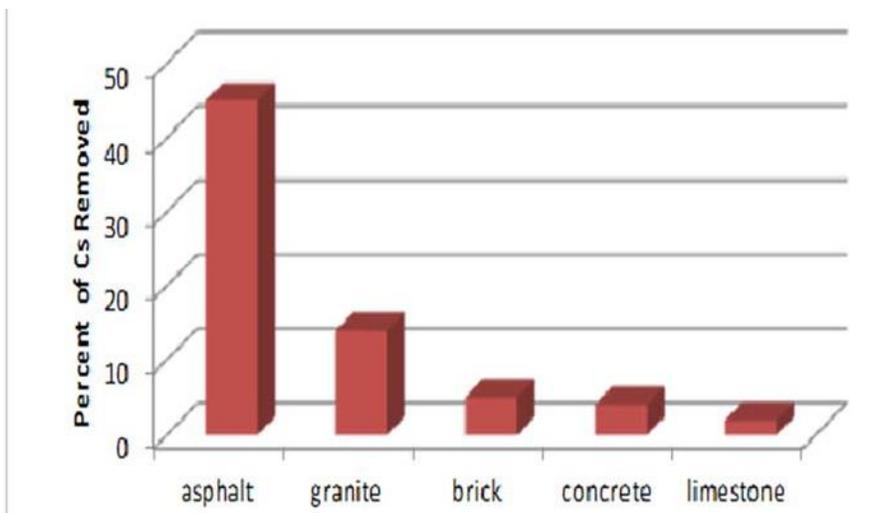


Figure 2: Percent of initial Cs contamination removed from study materials after simulated rainfall (2 cm/hour) (U.S. EPA, 2012).

Cs MOVEMENT WITHIN SOIL

In Fukushima, Japan, it was found that most of deposited radioactive Cs (more than 90%) remained within 2 inches (5 cm) of the ground soil surface at most sites (Matsuda et al., 2015). After the Fukushima incident, Cs was deposited in vegetative surfaces and the Cs was transported to the litter layer due to natural activities such as wind, precipitation, plant shedding, etc. The recent study in Fukushima suggested that Cs was rapidly leached from the forest-floor litter layer to soil layer. Then Cs was immobilized in the upper (0–5 cm) mineral soil layer through its interaction with clay minerals (Koarashi et al., 2016). Another Japanese study suggested that the transfer rate of Cs in litter layer to soil surface depended on the litter layer decomposition rate, which is a function of activity and biomass of microorganisms in the soil (Kurihara et al., 2018). The Cs-soil penetration depth and time may vary depending on various factors including soil clay content, litter decomposition rate, weather, precipitation, etc. However, based on the observations in Fukushima, the soil penetration depth will be less than 4 inches (10 cm) and the time to transfer the initial Cs on vegetation to the soil layer will take a few months to a year depending on the region.

Cs MOVEMENT IN BODIES OF SURFACE WATER

Runoff from contamination sources to surface waters is an important pathway for aiding in the transport of contaminants (Comans et al., 1989). The migration of contaminants is typically facilitated by decontamination efforts (i.e., wash water) and precipitation events. Although groundwater contamination resulting from the migration of radiological contaminants is still a concern, the spreading of contaminants at the surface level increases the odds of exposure through various routes, including direct exposure, ingestion (both direct and indirect) and possibly inhalation.

Once introduced to aquatic environments, radionuclides begin accumulating in bottom sediments and organic matter (Thiessen et al., 1999). Factors influencing aquatic contamination levels include ground-surface contamination levels, volumetric flow rate, and time since initial deposition (Monte et al., 2006). These levels are known to drop relatively quickly by means of flushing, burying, and radioactive decay. This process is accelerated in oceans, rivers, and lakes that are supplied by tributaries.

Cs UPTAKE INTO VEGETATION AND FUNGI

There has been some work to examine the uptake of Cs into food plants, particularly to identify where the Cs partitions within the different parts of the plant. A review article (Tensho et al., 1961) reported that Cs uptake into the edible portions of some plants was much lower than the amount of Cs that partitioned into the other parts of the plants. Uptake into the edible portions of plants ranged from 8.3%, 9.9%, 16.8%, and 16.9% for lowland rice, upland rice, broad beans, and wheat, respectively. It was suggested that the transport mechanisms for potassium (K) largely control the transport of Cs (Zhu and Smolders, 2000). This paper suggested that modeling the uptake of Cs by plants to assess a given plant species' ability to take Cs out of the soil is possible as a means to develop potential phytoremediation approaches, although based on the plants that were evaluated in these studies, the potential for those plant species to be operationally useful for phytoremediation is minimal due to the excessively long time (decades) that it would take to achieve meaningful reductions in soil-bound Cs, and this approach would potentially generate large volumes of waste. Some work (Kammerer et al., 1994; U.S. EPA, 2004) has suggested that fungi are potentially capable of accumulating higher quantities of Cs than vascular plants, which indicates that mushrooms may offer a more attractive option for phytoremediation, but additional work needs to be performed.

Forests are known to greatly reduce the overall mobility of radionuclides. Forests (including plants and trees) act as ecological sinks with a majority of Cs residing within the first few inches of soil. Though forests are effective at limiting the spread of contamination, they are difficult to decontaminate. Therefore, presence of nearby forests may influence decisions when considering decontamination of non-forested areas, especially when adjacent to populated or recreational areas, because the forests could act as either a sink for radionuclides or a source of further radionuclide releases.

Contaminants are either captured or absorbed by leaves and needles or migrated by means of root uptake (Shaw, 2007). The process of capturing (i.e., scavenging) through the forest canopy greatly varies by species. For example, conifers have a greater interception capacity than broad-leaved deciduous trees. A majority of the contamination eventually migrates to the litter layer by means of runoff during periods of rain or leaf shedding (Tikhomirov et al., 1993). Root uptake has been described as limited due to the retention qualities of soil. Contamination that is absorbed typically resides in the needles, twigs, and leaves versus the core wood (Nakanishi et al., 2013). The retention rate of Cs in trees and plants is a function of water availability (i.e., more water, higher retention) (Nakanishi et al., 2013).

Cs MOVEMENT DUE TO WILDFIRES IN CONTAMINATED FOREST LAND

Initial cleanup priorities after an incident would likely focus on populated areas, leaving the forested areas to pass for one or more seasons, where the overhead canopy materials (where initial deposition of radionuclides would occur) would fall to the forest floor. The nuclear power plant accidents in Chernobyl and Fukushima showed that the contaminated forests were not remediated on a large scale due to lack of fast and cost-effective mitigation methods. Forest decontamination is difficult because modification of forest via decontamination (e.g., top soil removal) may disturb forest ecosystems via erosion, sedimentation, disruption of habitat and loss of nutrients.

There is a potential for emissions of radionuclides such as Cs from a wildfire in a radionuclide-contaminated forest. EPA conducted a laboratory simulation study (Hao et al., 2018) of a wildfire

involving pine needles and peat doped with non-radioactive Cs. In this study, emissions of Cs were measured as a function of aerodynamic particle size and compared to Cs content of the residual ash. In these tests, only 1-2.5 % of the Cs on the pine needles burned was emitted as Cs in the airborne particulate matter. Peat fires did not emit Cs into the air. Most of the Cs was concentrated on particle sizes larger than 10 micrometers (μm). Additional work is ongoing to evaluate different forest fire conditions and different types of biomass.

Cs MOVEMENT WITHIN WATER AND WASTEWATER INFRASTRUCTURE

For Cs in water as a soluble ion, its movement through water and wastewater infrastructure primarily follows the flow of the water, relentlessly spreading contamination wherever it goes ((U.S. EPA, 2013). However, spread of Cs is also related to the ability of the Cs ion to interact with construction materials and other substances found in the urban environment (Kaminski et al., 2015b), including water and wastewater systems (U.S. EPA, 2014a). For many common construction materials (U.S. EPA, 2018), such as iron, copper (see Figure 3), and plastic pipes, interaction in water and wastewater system is minimal, occurs slowly, or is inhibited by other substances found in water and wastewater systems (e.g., other naturally occurring ions that out-compete Cs ions for available adsorption sites) (U.S. EPA, 2014d). However, adsorption to cement-mortar pipes and concrete infrastructure components (like channels, tanks, etc.) can occur under some conditions (Szabo and Minamyer, 2014), retarding movement and increasing persistence of Cs as described above. In fact, several other urban surface materials, like brick, are also used in construction of water and wastewater infrastructure (Kaminski et al., 2016). Hence, they are subject to the same Cs persistence as described above.

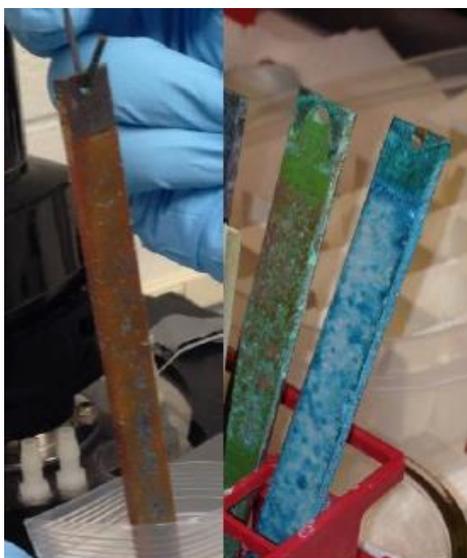


Figure 3. Laboratory experiments to investigate the persistence of Cs on copper surfaces. The different colors result from varying water quality parameters.

Cs strongly adsorbs to many clay and clay-containing substances (Jolin and Kaminski, 2016), such as soils, which may be found in water systems as sediments. Such sediments may be found in both drinking water and waste water systems. For instance, drinking water storage tanks can accumulate sediments that can act as reservoirs of Cs (U.S. EPA, 2014c). Sometimes these sediments result from fine particles arising from river/lake sediments containing clay that adsorb Cs. For waste water

systems, examples include stormwater, which can wash clay/sediments/soils into collection systems, changing the movement of Cs in several ways. Examples include: (1) transport of dissolved Cs into receiving waters; (2) concentration of adsorbed Cs when solids are removed from storm water via engineering controls; and (3) partitioning of Cs into wastewater collection and treatment plant components, especially when the stormwater enters into a combined stormwater-sewer system (as are found in many cities) (Kaminski et al., 2015a). In the wastewater collection system and plant, Cs can adsorb to construction materials (such as concrete, clay tiles, and brick). It can also be concentrated by unit operations designed to remove solids to which the Cs is coincidentally adsorbed (see Figure 4).



Figure 4. Close-up view of the collection trench in a drained aeration grit tanks showing potential for Cs contaminated sediment accumulation and concentration.

Cs MOVEMENT DUE TO PRECIPITATION EVENTS

Contaminants can be remobilized due to precipitation and flooding (James et al., 2005; Ueda et al., 2012). Contamination levels following these events have been closely correlated with increasing volumetric flow rates. River banks and flood plains near large rivers are of particular concern. These areas are known to collect high concentrations of Cs following significant precipitation events for many years following initial deposition (Japan Atomic Energy Agency, 2015). The EPA's Homeland Security Research Program is currently working to address the data gap associated with transport of radionuclides into and out of surface water.

CONTACT INFORMATION

For more information, visit the EPA Web site at <http://www2.epa.gov/homeland-security-research>.

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