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Review of technologies for preventing secondary transport of soluble and particulate radiological contamination from roadways, roadside vegetation, and adjacent soils

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Abstract

Transport of contaminants from roadways to the environment is well known, although studies of technologies for preventing and managing this appear infrequently in the literature. This paper reviews technologies studied for radiological contaminants. In addition to nuclear facility decommissioning, nuclear power plant accidents at Chernobyl (Former Soviet Union), Fukushima (Japan) and elsewhere have provided real world situations to both develop and test technologies to remediate radiological contamination and to return roadways, along with adjacent vegetation and soil, to prior use. From publications arising from these efforts, technologies were reviewed for radioactive material with two distinct properties (water-soluble and insoluble radioactive contaminants). The reported characteristics and capabilities of technologies are summarized in this review. This review also presents logistical considerations of implementation of the technologies, including waste management which can be an extreme impediment to rapid remediation if generated quantities of hazardous waste exceed local handling capacity. The summarized literature review suggests future avenues of work, chiefly for insoluble particulates, focused on technologies which may be mechanistically applicable to their remediation. While the underlying chemical and physical mechanisms that contribute to transport differ among contaminants, the studies reviewed here might also be applicable to non-radioactive contaminants, because the presence of radioactivity is largely independent of the underlying mechanisms.

Keywords

Roads; Street cleaning; Street sweeping; Radioactivity; Vegetation; Soils

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Supplementary materials

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1. Introduction

Adopting mitigation strategies to reduce road dust emissions is important for managing transport of contaminants through the environment. One example is the goal of reducing atmospheric particulate matter (PM). Even though this is a topic of wide spread interest, a review about technologies to accomplish this noted “a general dearth of information about their effectiveness in reducing ambient PM concentrations” and also related that most of the information comes not from the scientific literature but from local authorities (Amato, 2010). In this case, resuspended roadside dust is composed of: 1) a bulk mineral from soil, pavement abrasion, construction by-products, etc.; 2) metal loading (e.g., copper, antimony, iron, manganese, zinc) resulting from vehicle wear (e.g., brakes, tires, catalysts); and 3) organic loading from leaked automotive fluids (e.g., fuels, oils, coolants). In addition to atmospheric transport of roadway contaminants, secondary contamination of soils and vegetation adjacent to roadsides occurs (Werkenthin, 2014).

Common control processes for transport of roadway dust are sweeping, washing, and dust suppressants (Amato, 2010) with few new studies for each appearing in the literature. To expand this limited knowledge base, it is useful to consider another scenario in which reducing environmental transport is important, namely when radiological contaminants become deposited on roadways and their surrounding areas. This can occur from erosion of natural radioactive materials and movement of contaminated sediments onto roadways (Muminov, 2010), as well as anthropogenic activities such as nuclear power plant (NPP) accidents, radiological dispersion device (RDD) releases, nuclear detonations, and other various accidents. While wide area radiological contamination is a less frequent occurrence, great public awareness, interest, and sensitivity to radiological contamination has resulted in studies regarding technical effectiveness surrounding the implementation of technologies and methodologies to manage the environmental transport of radionuclides from their initial deposition on roadways, including roadside vegetation and soils. Aside from minimizing public health impacts, timely implementation of these technologies and methodologies is very important because roadways, both urban and rural, are vital to response and recovery operations for impacted communities. While fueled by the interest in minimizing public health impacts and maximizing community resiliency, these technical studies have arisen because it is scientifically recognized that if the radionuclides are not adequately contained in the roadway and roadside areas, there is a great potential for spread of radioactive contamination through airborne (e.g., particulate) and especially waterborne (e.g., soluble and particulate) routes. Namely, roadways are designed to drain into local stormwater systems which in turn can ultimately drain into regional, tribal, national, and international water systems.

One reason why radiological contamination of roadways can serve as a source of data for managing roadway emissions from other types of contaminants stems from the magnitude of historical incidents. Radiological contamination resulting from NPP accidents, intentional release through an RDD, or an improvised nuclear device (IND) can contaminate large areas requiring remediation, on the order of dozens of city blocks (each typically a fraction of a square kilometer) to hundreds of square kilometers. Due to the size of these areas, a variety of roadways are impacted, both urban and rural, and may present a significant challenge

and technical gap. The Fukushima NPP accident produced an immediate contamination area the size of the United States (US) state of Maryland. In US National Planning Scenarios, an example RDD could contaminate 36 city blocks (tens of square kilometers), while an example IND may contaminate ~8000 km² (GAO, 2013). A prior report by the US Government Accountability Office (GAO) (GAO, 2010) found that remediation technologies selected simply because of prior use in an unrelated/untested situation may generate waste types and volumes that are more difficult to dispose, subsequently increasing remediation time and cost. Additionally, a US GAO investigation (GAO, 2013) found that while Federal guidance (DHS, 2009) would direct State and local governments to initiate decontamination procedures, it provided only limited information on the capabilities needed to complete such actions. Moreover, some State and local governments may not have expertise, prior knowledge or available technologies to initiate wide area remediation, and this may negatively impact subsequent remediation efforts and waste management. The complexity and difficulty escalate when multiple nations are involved, as was the case for the Chernobyl NPP incident.

Pre-planning for remediation can greatly reduce the time to respond, and subsequently can minimize further contamination and reduce cleanup costs, allowing responders to minimize the impact to both public health and the environment. US Department of Homeland Security (DHS) updated the National Infrastructure Protection Plan (The White House, Office of the Press Secretary, 2013) in support of Presidential Policy Directive 21, Critical Infrastructure Security and Resilience (DHS, 2013). Of interest in the Protection Plan to this review paper is the transportation system—including road, rail, airports—providing ingress and egress routes from urban areas, which indirectly impact plans for power, water, communications, health, security, and emergency service.

The purpose of this review paper is to summarize the most applicable available information on technologies and methodologies for managing transport of radiological contaminants on roadways and roadsides. This information mainly comes from both laboratory studies of radiologically contaminated sites/facilities, as well as reports from catastrophic radiological releases following NPP disasters. This information may be applicable to non-roadway related radiological releases, as well as non-radioactive contaminants on roadways, which may also result in environmental transport through air- and water-borne routes.

2. Methodological approach

A broad range of expertise and capabilities has been tried and tested for removing radiological contamination from buildings and sites at commercial and research nuclear facilities and the US Department of Energy (DOE) complex sites. However, remediation of nuclear/radiological facilities differs greatly from a wide-area urban remediation effort in many ways, including magnitude, timeframe, urgency, cost, and stakeholders, as well as psychological, public perception, and economic impacts. Despite nuclear complex decommissioning experiences and remediation of widespread contamination following NPP accidents at Windscale (United Kingdom [UK]), Chernobyl (Former Soviet Union [FSU]) and Fukushima (Japan), remediation technologies that can be quickly and effectively deployed to address RDD and IND contamination can be greatly improved. Such

technologies need to be widely and quickly available; easy to use; efficient; safe for operators, public and the environment; and have minimal waste management concerns.

This review summarizes the technologies and their available performance characteristics that can be used for remediation of wide area surfaces following RDD, IND, and NPP accident contamination, with preference given to commercial off-the-shelf technologies that can specifically be applied to remediation of contamination on roadway and roadsides. This report leveraged prior technology reviews from five major resources: 1) US Environmental Protection Agency (USEPA) reports; 2) International Atomic Energy Agency (IAEA) reports; 3) Reports related to major NPP accidents, such as Fukushima and Chernobyl accidents; 4) Literature related to nuclear weapons tests and accidents sites; and 5) Publications from 2012 to present, including review articles describing older publications and other information such as technical support and vendor information. To the extent possible, the purpose of this review is to provide quantitative metrics for comparing available technology costs, application time, labor and equipment requirements, waste type/volume, availability, and technical performance that would be utilized in a wide area incident response.

3. Remediation of radiological contamination from roadways and roadside vegetation/soils

3.1. Background on radiological particle characteristics observed at contamination sites

To help understand remediation approaches, it is necessary to discuss the physical and chemical characteristics of radionuclides historically observed from their release. Limited literature was found characterizing identified radioactive materials from the release scenarios (NPP accidents and nuclear detonation) while none was available for RDDs. The literature points to a wide radioactive particle size range (<10 μm [micrometers] to fragments) and possibly thin surface films formed during release in some cases.

In general, an RDD refers to a device that intentionally disperses radioactive material. An RDD can be a conventional bomb or a method designed to disperse radioactive material to cause destruction, contamination, and radiation injury - an RDD does not produce a nuclear yield like a conventional or improvised nuclear device (CIA, 2003). While a criminal act that dispersed radioactivity—but perhaps not with the intent to do so—a cited “benchmark” for the consequences of an RDD is the 1987 Goiania incident in Brazil involving the human dispersion of the radioactive ^{137}Cs chloride powder from a stolen and broken radiotherapy radiation source, leading to 4 deaths and 244 people exposed (Magill, 2007). Two potential sources of materials for RDDs due to their prevalence include commercial radioactive sources and nuclear fuel. Commercial sealed radiation sources often contain powders like cesium chloride, radium-226 bromide/chloride, sintered solids such as ^{90}Sr fluoride, and metals like cobalt-60 (Peterson, 2007). Powders are generally considered to be widely dispersible, and solid forms like metals are considered to be much less dispersible. However, a RDD’s radioactive material could be purposefully altered physically and chemically as part of the device design. Physical or chemical alteration could also result from the explosion of a “dirty bomb” to potentially include oxides and/or nitrates (from the explosive)

over a wide particle size range. Nuclear reactor fuels can come as uranium dioxide (UO₂), mixed-oxides (a mixture of uranium and plutonium oxides), uranium metals or their alloys, and microspheres (uranium or thorium oxide), as well as less common forms (Bodansky, 2004).

The most extensive characterization of nuclear-incident-produced radiological particulates found were those resulting from nuclear weapons detonation tests by the US and UK. Nuclear detonations of American weapons containing both uranium-235 (²³⁵U) and plutonium-239 (²³⁹Pu) at Enewetak Atoll (DOE, 1982) created objects near “ground zero” that were plated with thin films of plutonium and fission products (e.g., ²⁴¹Am, ¹³⁷Cs, ⁹⁰Sr) while radioactive particles rose to great heights before settling back to Earth or being washed down by rain. These particulates range from occasional millimeter-sized Pu metal pieces to sizes similar to particles naturally found in soil. Plutonium-239, ²⁴⁰Pu, ²⁴¹Am (produced by beta decay of plutonium-241, a small part of weapons-grade Pu), and ²³⁸Pu were alpha emitters seen in significant amounts. Contaminated metal debris was also found in one area when the device failed to yield fission. Millimeter-sized and larger radioactive fragments, as well as Pu-contaminated beryllium, were detected on-site from plutonium-device safety tests (Hamilton, 2009). United States land-based Bikini Atoll nuclear weapons tests (DNA, 1981) produced dry white, opaque, and irregularly shaped particles (–15–1000 μm diameter, many particles flaky), as well as non-crystalline spheroidal particles (likely nuclear device components and fission products). One site sample suggested 33% of the activity from >225 μm particles, and ~20% of the activity in <10 μm particles, with specific activity decreasing with increasing particle size. Fallout decay activity also included beta disintegrations, gamma photons, and gamma ionization. UK weapons tests at Maralinga in south Australia (Burns, 1995) also created plutonium-coated pieces (metal, plastic, wire, etc.), fine fragments/particles sometimes not visible to the eye, and very finely divided material in the inhalable range, similar to soil particles. A portion of the Pu contamination in the soil existed as < 1 mm particles of high specific activity.

Among accidents involving nuclear weapons, later forensic analysis of the 1960 Boeing Michigan Aeronautical Research Center (BO-MARC) “broken arrow” (non-nuclear explosion of/fire involving a nuclear weapon) missile incident found generally crystalline/smooth Pu particles with 15–65 μm lateral dimensions (Bowen, 2013). This contrasted with the particulates collected from the 1966 Palomares, Spain and 1968 Thule, Greenland hydrogen bomb incidents which were fluffy amorphous or agglomerated grains resembling popcorn.

Nuclear reactor melt-down-produced radioactive particulates are also characterized in the available literature. In the case of Chernobyl (Pollanen, 2002), >95% of the identified particulates were UO₂ reactor fuel particles generated by mechanical fuel disintegration, and <3% were attributed to condensation particles (<1 μm) produced from volatiles such as cesium (¹³⁴Cs, ¹³⁷Cs), iodine-131 (¹³¹I), and rubidium-87 (⁸⁷Rb) within 10 km of the nuclear plant. Fine condensation particles made up ~60% and 98% at 25 and 65 km, respectively, from the reactor, while >65% of the total activity came from nuclear fuel particles in the 30 km around the NPP. Chernobyl-produced particles were reported to be crystalline due to their release at high temperatures (Krekling, 1998). Identified particulates

from the Fukushima NPP accident were aerosols (~0.1–2 μm) volatilized due to the reactor meltdown that collected radioactive cesium and iodine while airborne and came down by dry deposition (Kristiansen, 2012). Some isolation and characterization of particulates (up to ~6.4 μm diameter) from the Fukushima accident have recently begun to be described (Sato, 2016), including cesium-rich microparticles (Furuki, 2017; [moto, 2017; [kehara, 2018; Ochiai, 2018)

3.2. Overview of remediation of radiological contamination from roadways and roadsides

The primary objectives for wide-area remediation are to minimize human radiation exposure and to reduce the risk of environmental contamination. Early remediation (decontamination performed as soon as possible after an incident) can support response and recovery by restoring the ingress and egress routes (e.g., road, rail, air), power, water, and communications, along with health, security, and emergency services. Non- and minimally-destructive remediation technologies may reduce the response cost by generating less waste volume and may support recovery efforts because of return to usability of the impacted structures. A variety of remediation technologies exist, ranging from merely waiting for natural decay, to simple physical removal through vacuuming, to complete surface removal or demolition. Most of the remediation technologies described in this review paper, except for repaving, some coatings, and reverse tillage, involve the removal of surface contaminations.

Many commercial entities exist with expertise and capabilities to perform remediation of contaminated facilities and localized sites. However, after a site's contamination characteristics have been assessed and required decontamination efficiencies determined, an evaluation is required to determine which technologies could be utilized to respond quickly in each *wide area* remediation effort with minimal destructive impact (corresponding to minimal waste volume and costs). This decontamination technology summary is intended to be a tool to help guide these evaluations, with the "best available" published data from past applications to actual radiologically contaminated sites. In some cases, methodologies may not be traditional nuclear/radiological decontamination tools. For example, retrofitted street-sweepers along with other municipal and commercial equipment may prove useful in cleaning large areas of roadway (while being easy to operate and widely available) if adapted to the particular technical needs associated with the specific radiological release (Kaminski, 2018).

The USEPA identified several surveying, vegetation removal, surface removal and cleaning, waste stabilization, waste water cleanup, and volume reduction technologies (USEPA, 2013b). This USEPA report provides good qualitative information, but no quantitative details such as cost or efficacy. The summary of surface decontamination methods is reproduced in Table 1.

Several reviews of remediation and waste management technologies have previously been performed (e.g., Brown, 2007; IAEA, 1989, 1999; Lehto, 1994; Nisbet, 2015; USEPA, 2006a2013a2013b 2013c), in which categories are broadly compared using key factors (health and safety, time to implement, performance, availability, costs, process waste, and throughput) and vendor information is documented. These reviews can reflect radiological

incidents (e.g., Chernobyl and Fukushima NPP accidents) that happened since prior reviews that lead to practical information and renewed interest in scientific research in the field. They can be specific to the country or region that developed them, such as the UK Recovery Handbook (Nisbet, 2015) and the EURANOS Handbook (Brown, 2007). These reviews can also contain summaries of technologies, arranged in various formats such as datasheets/tables, that reflect the nature, focus, and purpose of the review. This review paper focuses on the specialized topic of roadways and roadsides, key infrastructure upon which many activities following a radiological incident are dependent.

In some cases, methods and technologies are expected to provide good results but are not well documented in the literature (USEPA, 2014a). Limitations that cannot be closed by technology development are identified (USEPA, 2013a2013b 2013c), such as the time taken to implement or limited availability. However, technological capabilities including performance, throughput, and waste generation can be improved to provide better solutions to radiological remediation. An example is lawn mowing, which scored moderately overall (USEPA, 2013b) but could score highly if improvements were made to environmental, safety and health aspects, and the capturing of contaminated cuttings. By combining this technology with others (e.g. fixatives or filtration) from the technology *toolbox*, both low-scoring factors associated with mowing can be addressed to improve overall performance. Similarly, improving dust collection and filtration associated with municipal equipment, such as street sweepers and vacuum leaf collectors, could reduce both waste residuals and the risk of resuspension.

RDDs and INDs, depending on their makeup and design, have the potential to generate water-soluble or -insoluble contamination, if not both within a single incident, like the Chernobyl NPP accident (Pollanen, 2002). Radioactive sealed sources often expected to be used in RDDs can come in both water-soluble (e.g., cesium chloride) and insoluble (e.g., americium oxide) forms (Peterson, 2007). If traditional nuclear weapons are used as the model for INDs, insoluble films and particulates/pieces such as plutonium (Pu), as well as soluble ^{137}Cs and ^{90}Sr , would be generated (DOE, 1982). Plutonium-238 and ^{241}Am have half-lives of 88 and 430 years, respectively, and are inhalation/ingestion hazards despite being alpha-particle emitters that do not pose a significant external health hazard. Cesium-137 (half-life of 30.17 years) is a fission product in nuclear reactors, and it has medical and industrial applications. Cesium-137 is identified as a principal contaminant of concern, among several other radionuclides, due to the combination of its intermediate half-life, decay by high-energy pathways, high chemical reactivity, and high mobility in the aqueous phase (Hardie and McKinley, 2014; IAEA, 1999; JAEA, 2015a). Therefore, the majority of surface remediation technology research, development, and demonstrations are based on removal of radioactive cesium (USEPA, 2013b; USEPA, 2014b; JAEA, 2015a; Kaminski, 2016).

A discussion of remediation technologies appears below in Sections 4 and 5, and an important concept for comparing remediation technologies is the Decontamination Factor (DF). This is the ratio of initial contamination level to the contamination level after decontamination, and it can be translated to removal efficiency using the following definitions:

$$DF = \frac{C_{init}}{C_{final}} \quad (3-1)$$

$$\% \text{ Removal} = \frac{C_{init} - C_{final}}{C_{init}} = 1 - \frac{C_{final}}{C_{init}} \quad (3-2)$$

where C_{init} and C_{final} are the pre- and post-treatment contaminant concentrations. By substituting Eq. (3-2) into (3-1), the final relation (Eq. (3-3)) is as follows:

$$\begin{aligned} \% \text{ Removal} &= 1 - \frac{1}{DF} \\ \% \text{ Removal} &= 1 - \frac{1}{DF} \\ \% \text{ Removal} &= \frac{DF - 1}{DF} \end{aligned} \quad (3-3)$$

For purposes of discussion below in terms of the removal of radionuclides, Table 2 illustrates a range of conversions of DF to percent removal. As Table 2 suggests, it is especially good to keep in mind that lower DF values correspond to numerically wider ranges of percent removal, and the reverse at high DF values. In practice, the remediation technologies (Sections 4 and 5) can result in a range of DF values depending on the exact situation to which they are applied, so DF values can be thought of as illustrating the potential of a technology, rather than defining it.

As noted above, the primary objectives for wide-area remediation are to minimize human radiation exposure and to reduce the risk of environmental contamination, so in some circumstances, if a DF reduction does not result in a suitable minimization of radiation exposure, remediation via resurfacing or repaving (as opposed to decontamination) may be necessary to reduce dose from radionuclides remaining after application of remediation technologies. Even for remediation technologies with high DF but which have slow implementation rates, the application of repaving provides additional options for recovery and opening routes to key infrastructure locations while significantly reducing the dose to those who travel on the contaminated roads or freeways. Similar techniques could be applied to parking lots, public open spaces, and airport runways and taxiways. Long term concerns about exposure and migration of radionuclides should be considered as the repaved surfaces ages, potentially re-exposing the contaminated surface. In this regard, the emergence of potholes in the repaved surfaces due to weather conditions should also be monitored.

In addition to repaving, resurfacing a roadway may provide a significant reduction in dose, while stabilizing the contamination and preventing migration. This technique was used following the Chernobyl NPP accident, with 25,000 miles of road washed daily and most resurfaced with asphalt, concrete or stone resulting in a 3-fold reduction in dose (IAEA, 1991). Note that it is possible to apply asphalt over vegetated areas and land next to roadways to accomplish the same purpose, although the mechanical instability of these areas may quickly cause the applied asphalt layer to degrade. The method, which typically involves adding a layer of stabilizing gravel followed by asphalt and using a road roller to consolidate, can be applied on a large scale over topsoil or existing roads, and results in a

50–75% of dose reduction from ^{137}Cs through the application of 5–6 cm of asphalt over a large area (Andersson, 2003). Typically, a 5–10 cm thick layer (asphalt density typically 1.6 g/cm^3 , with a maximum of 2 g/cm^3 depending on pebble type) was applied. Traces of contamination have been observed in the repaving material.

Repaving and resurfacing techniques are not discussed below because the required equipment will depend on the specifics of the impacted roadways, and there is great familiarity and availability of such equipment. The type and nature of contaminated dusts produced may differ depending on the type of radiological contamination and should be factored into health and safety plans. However, this approach could be widely performed by road and construction crews, with equipment that is readily available and minimal additional training of workers.

4. Remediation technologies for water-soluble radiological contamination of roadways and roadsides

The bulk of the recent wide-area radiological decontamination literature has focused on the removal of mobile water-soluble species, principally ^{137}Cs and ^{90}Sr , due to both the long half-lives of these radionuclides and difficulties in their removal due to surface or ground penetration and movement caused by natural waters (e.g., rain, streams, groundwater). Soluble contaminants may be transported via precipitation and water application, and the contaminants will be distributed horizontally and vertically in the environment. Vertical transport was observed in Fukushima with a few millimeters into concrete and several (up to 30) centimeters (cm) into soil. The transported soluble contaminants can further react with surfaces.

The following subsections discuss the key elements of decontamination technologies for soluble surface contaminations, with separate subsections for either physical or chemical removal technologies. The discussion below is intended to be descriptive, including minimal details like DF and cleaning rates, when reported. Additional details, including operational considerations) of each technology, including implementation with specific equipment, can be found in the “Operational Information” sheets in Appendix A of the Supplemental Information. These operational information sheets include entries for environmental health and safety considerations for workers. These entries are intended to alert the reader to the types of potential hazards that should be addressed in Health and Safety plans required by local authorities. In the operational information sheets, some cost information is provided for reference purposes only, because equipment, labor, and other logistics costs can vary widely by location, even within a particular region. Costs may also be significantly impacted during a wide scale incident by market forces because the equipment, while often commercially available, is routinely used for other purposes and not stockpiled for radiological emergencies.

Some surface decontamination processes use a combination of both chemical and physical removal technologies. Chemical surface removal technologies generally dissolve the contaminants on surface materials to form soluble species, which can be removed with either liquid or solid carriers. Contaminants with varying chemical properties may have

to be decontaminated by a combination of different chemical technologies. Therefore, chemical methods are chemical compound specific. However, physical methods such as high-pressure washing, machining, and abrasion physically remove a thin surface layer of the contaminated substrate and therefore, are not dependent on the contaminant's chemical form.

Recently gained experiences and knowledge from remediation of contaminated areas in the aftermath of Fukushima NPP accident suggest that physical technologies were operationally easier and effective to apply to wide-area decontamination (IAEA, 2013; JAEA, 2015b). However, the operational summary sheets in Appendix A (and also Appendices B and C) were written to allow tailoring to the characteristics of the cleanup site (e.g., size including wide area) and available resources (e.g., manpower, equipment). While performed for other types of cost representations, no attempt was made to apply them even to a model city, town, and rural area, as each site was expected to vary significantly. The available information compiled in the operational summary sheets is primarily intended to inform practitioners about technologies that may be useful for their goals—while accounting for local constraints such as equipment availability, desired schedule, manpower, budget, etc.

4.1. Physical removal technologies for roadways

For technologies described below that physically remove water-soluble contamination, the contaminant is assumed to physically or chemically bind to substrates, such as roadway construction material (e.g., concrete, asphalt), dust and debris on the roadside, vegetation adjacent to the roadway, etc. Thus, removal of the substrate, along with the bound contaminant, removes the radioactivity. These technologies generally remove not only the contaminants on the surface, but also a layer of the surface in order to remove the surface bound-contaminants. Instead of liquid wastes, mechanical removal of hard surfaces may generate dusts and airborne hazards, also risking worker exposure and producing secondary contamination. High-efficiency particulate air (HEPA) filtration technology, if operated properly, can be incorporated with many of the mechanical surface removal technologies to eliminate airborne hazards.

The physical removal technologies specifically discussed below are all variants of a two-step process: a) apply a mechanical force to a surface and b) remove what is loosened from the surface. The sections below tend to discuss specialized equipment for doing this, which, while not necessarily the original intent, seem applicable to large areas like roadways. Other variations of this “brush/wipe/rinse/vacuum” strategy are available, especially for small areas within roadway infrastructure that are inaccessible to the equipment below. In addition, toll booths or roadway maintenance garages may require remediation, and can be considered to be remediated similar to buildings, not as roadways.

4.1.1. Road sweeping (“Street Sweeping”) (Appendix A.1 operational information sheet)—Many models of vehicle-based road sweepers (sometimes referred to as “street sweepers” but more generally refers to different sizes of equipment depending on the intended application, e.g., parking lot sized sweepers) are equipped with high-speed rotating hard steel-wire brushes, a conveyor for transporting debris, and a trash container.

The sweepers can sweep and collect debris, litter, and soils (to which water-soluble radiochemicals have attached) from roads (both paved and unpaved) and other paved surfaces (e.g., sidewalks) of suitable size. The high-speed rotating brooms can exert an abrasive action on the pavement and sweep clean rough surfaces.

Road sweepers with the following features – either supplied by the manufacturer or through aftermarket modification – could be beneficial to address the safety and technical needs of radiological decontamination operations. These features include, but are not limited to, the following: a) enclosing the brushing action in a hood which is connected to HEPA filtration vacuum system; b) a cab enclosure with a filtered air supply and shielding against strong beta and gamma radiation; c) a radiation monitoring system to scan the surface and provide feedback on the extent of decontamination; d) a remote operations system for highly contaminated (i.e., high-radiation dose) areas; and e) installing removable covers or coatings on portions of the road sweepers, especially the re-entrant surfaces, to prevent heavy contamination and facilitate sweeper decontamination prior to maintenance work (Barbier and Chester, 1980; USEPA, 2014b).

A regenerative air sweeper, e.g., TYMCO Model DST-6 Dustless Street Sweeper, uses a controlled blast of air to dislodge debris from the surface. All debris swept up by the pick-up head is directed up a large diameter heavy duty suction hose into the hopper. The sweeper removes trash, dirt, and fine particles from the entire area beneath the full-width pick-up head and cleans the diverted air to 99.999% of 0.5-micron sized particles. 0–50% removal efficiencies (DF= 1–2) with cleaning rates of 219 and 438 m²/h for medium and large sizes road surface cleaning vehicles, respectively, have been reported. (JAEA, 2015b).

Some street sweepers are capable of spraying water onto the road surface. For radionuclides in chemical forms that do not react with surfaces, washing with water can be considered a form of physical removal of the dissolved radiochemical. Water can be applied at low or high pressures, and, in the case where the soluble radionuclide has dried, the application time should allow for re-dissolution. Water generated from the washing operation should be appropriately managed. In addition to street sweepers, other apparatus and equipment are discussed in detail below for more complex cases than simple water washing.

4.1.2. Vacuuming (Appendix A.2 operational information sheet)—Industrial-grade vacuums are commercially available and operation requires minimum training. Vacuums equipped with HEPA filters that are designed for radiological decontamination can retain, if verified to be functioning properly, nearly 100% of particles larger than 0.3 microns. Vacuuming is usually recommended as the final cleanup of remediation areas after materials have been dried and contaminated materials removed. Although it works best on smooth surfaces, vacuuming also reduces loose contamination on porous and rough surfaces. A wide range of vacuum systems are commercially available. This technology generates debris wastes. The reported DFs range between 1.1-1.2 (Bossart and Blair, 2003; USEPA, 2013b; Gates-Anderson, 2012; Heiser and Sullivan, 2009; JAEA, 2015b; Kaminski, 2016). Reported decontamination rates were 12 m²/h (JAEA, 2015b).

4.1.3. High-pressure and ultra-high pressure washing (Appendix A.3 operational information sheet)—The technology uses high water pressure to remove not only the removable and/or soluble contaminants, but also some of the contaminated surface at sufficiently high pressures. Pressures up to 3000 pounds per square inch (psi) or 20 megapascals (MPa) are known as high-pressure washing, and higher pressures up to ~26,000 psi (180 MPa) are commonly known as ultra-high pressure washing, hydroblasting, hydrolasing, and hydraulic blasting (USEPA, 2014b; Fisher and Kohler, 2011; Heiser and Sullivan, 2009). These technologies can be useful for some difficult contamination scenarios but are labor-intensive. Perhaps one of the most significant drawbacks to the technology has been that it generates large quantities of wastewater, which requires secondary waste handling and treatment (Fisher and Kohler, 2011). Japan's Decontamination Pilot Project has demonstrated that pressure washing is able to treat large areas and a variety of surfaces quickly, with relatively quick mobilization and set up times. High-pressure washing operations require skilled workers, and published DFs range from 1.2–5 depending on the surface type, operation pressure, and speed (USEPA, 2014b; JAEA, 2015b; Kaminski, 2016). Lower pressure can remove paint from concrete while leaving the concrete intact. Higher pressure can be used to remove 0.5–1 cm or more of the concrete from the surfaces. The cleaning speed of a water-jet vehicle can be as high as 125 m²/h (JAEA, 2015b). Manual brushing and wiping have been used in combination with high-pressure water jet washing (10–20 MPa) to increase DF (to 1.3–3.3) (JAEA, 2015a).

4.1.4. Hot water pressure washing and steam vacuuming (Appendix A.3 operational information sheet)—As a variation on pressure washing, but requiring different equipment, hot water washing and steam vacuuming utilize pressures up to 1000 psi (7 MPa) and heated water to enhance decontamination performance. Cleaning rates are 28–33 m²/h for hot water pressure washing, and 9–14 m²/h for steam vacuuming (Heiser and Sullivan, 2009).

4.1.5. Blasting (Appendix A.4 operational information sheet)—This technology uses high pressure to propel a stream of abrasive particles against a surface to remove a thin layer of the surface. The kinetic energy of the media abrades and cleans the surface. Compressed air or a spinning wheel can be used to propel the blasting media. The most common blasting is grit blasting, also known as sandblasting, for which sand or synthetic grits are used. HEPA filter vacuums are used to collect wastes and recycle the grit. Grit can be recycled a number of times before wearing out and requiring replacement. The waste stream consists of the removed surface material and spent grit. Blasting uses a large amount of grit, so cost depends on the grit chosen. Most surface types and shapes, including complex surface geometries and intricate surfaces, can be treated with the proper choice of grit. The reported DF range was 2.5–10 depending on the abrasive media materials and the type of blasting (JAEA, 2015b). Treatment rates are on the order of 5.5–9 m²/h, depending on the method of propelling of abrasives (Heiser and Sullivan, 2009; JAEA, 2015b; Kaminski, 2016).

There are two types of grit blasting: air and airless (or centrifugal shot) blasting. The major advantages of a centrifugal shot blasting are its high production rates, its high efficiency

with low cleaning cost, and no moisture concerns during operations. This technology is better used for decontamination of large areas of open space. While decontamination efficiency is lower, traditional compressed air blasting is simple to operate, has low initial cost, is extremely flexible, and can be used to clean deep holes and cavities. The two types of blasting are complementary for different decontamination tasks.

4.1.6. Dry ice (carbon dioxide), sponge, and soda blasting (Appendix A.4 operational information sheet)—These technologies use compressed air with less aggressive/soft pellets as the blasting media. They are best used for more delicate surfaces or surfaces with low-level contamination. The dry ice pellets vaporize after they strike the surface, so there is less secondary waste generated compared to blasting with other media. The absorptive soft sponges can be wetted with cleaning agents to enhance decontamination efficiency or impregnated with abrasives to tailor to a specific surface. In soda blasting, sodium bicarbonate is applied against a surface using compressed air. It is a very mild form of abrasive blasting and a non-destructive method for many surface cleaning applications (Benson, 1995; Heiser and Sullivan, 2009; JAEA, 2015b). A 60–90% decontamination efficiency (DF range of 2.5–10) and ~9 m²/h decontamination rate was reported for dry ice blasting (JAEA, 2015b).

4.1.7. Shaving (Appendix A.5 operational information sheet)—Shaving technology uses diamond/metal blades on electrically powered equipment to shave off a thin layer of the concrete surface with fixed and penetrated contamination. Among the core dry decontamination technologies, shavers are used primarily in open flat areas. They effectively remove a uniform layer of surface by milling it. Typically, the shaver employs a rotating drum and a series of blades that are spaced closely to remove a consistent layer of material. A vacuum recovery system should be integrated to collect the shaving debris and control dust. The advantages of shaving technology are that it leaves behind a relatively smooth and uniform surface while creating a minimal volume of waste. The technology is most effective for smooth surfaces and surfaces contaminated with thin depositions of contaminants. A DF of 10 and higher is reported to be attainable. Reported shaving depth are from 0.1–13 millimeter (mm), and removal rates are 15–120 m²/h (DOE/EM, 1998a; JAEA, 2015b).

4.1.8. Scabbling (Appendix A.6 operational information sheet)—This technology is based on a series of impact heads that aggressively beat the target surface, break it up, and chip it away. The chipping action removes a concrete layer in 1.5–4.5 mm increments. A recovery HEPA vacuum system should be used in conjunction with a scabber to control dust and debris. Compared to shaving technology, scabbling is slower and leaves a flat but roughly finished surface. Very high DFs are attainable (i.e., DF of 10 or higher or >90% efficiency) by controlling the surface removal depth (Heiser and Sullivan, 2009). Reported removal rates were 23–42 m²/h for a concrete layer removal thickness of 0.15 cm (DOE/EM, 1998b; DOE/EM, 1998d; DOE/EM, 2001).

4.1.9. Grinding (Appendix A.7 operational information sheet)—This term refers to coarse grained abrasive in the form of either water-cooled or dry diamond grinding wheels, metal wire brushes, or multiple tungsten carbide-coated discs or sanding heads

to remove thin layers or coatings from the contaminated surface. A grinder is an electric-powered tool with a vacuum port for dust extraction and is suitable for flat or slightly curved surfaces, resulting in a smooth surface. Literature DFs range from 1.6 using iron brushes to 14 with a diamond grinding wheel. Increasing dwell times and number of passes also increase achieved DFs (Heiser and Sullivan, 2009). Wastes generated were minimal, and a reported decontamination rate using a portable grinder with a 12.7 cm diamond grinding wheel was 1–3 m²/h at 1.5 mm removal depth (DOE/EM, 1998c).

4.2. Chemical treatment of roadways

4.2.1. Low pressure washing, including additives and foams (Appendix A.8 operational information sheet)—The technology uses chemical reagents dissolved in water or as a foam, which are sprayed on contaminated surfaces, allowed dwell times to take effect, and then removed by vacuuming and/or rinsing steps (Bossart, 2003; USEPA, 2013a 2013c; Kaminski, 2016; Lear, 2007). The application of decontamination liquid or foam is a fairly time-efficient process, but the removal step can be time consuming (Drake, 2011).

The rinse step generates secondary wastewater and poses potential wastewater runoff risks. The USEPA conducted evaluation for Radiation Decontamination Solutions, LLC products QDS-H and QDS-TM. Reported decontamination efficiencies were around 50% or DF of 2 (USEPA, 2011). The decontamination rate was 0.22 to 0.45 m²/h, and the use rate of QDS liquids were 2–3 liters (L) per m².

Chemical reagents can also be applied using low pressure water at varying flow rates, e.g., ranging from a garden hose to a fire hose. In this case, the chemical reagents often take the form of ions which competitively displace radiochemical contaminants from their binding sites. The amount and type of salt additive depends on the chemical form of the radionuclide and the surface being decontaminated. In cases where radiochemical binding is not strong, ionic solutions as weak as most tap waters can be efficacious, but some surfaces may require 0.1 M or more concentrations of salt additive (Kaminski, 2016). Such water-based washing systems require considerable volumes of wash water and also generated considerable volumes of waste water. Therefore, to implement reagent-based washing, an on-site system that combines collection of wash waters with on-site treatment has been reported. This system, referred to as the Integrated Wash Aid Treatment for Emergency Reuse System (IWATERS), utilizes wash waters containing salts and other common additives to enhance the removal of radionuclides from various surfaces (e.g., asphalt, brick, concrete, glass, granite, painted surfaces, metal, wood). The effectiveness of this systems can depend on the salt concentration, type of salt, type of building material, radionuclides present, their physico-chemical properties, and site-specific IWATERS application (Kaminski, 2015).

4.2.2. Gels and Strippable coatings (Appendix A.9 operational information sheet)—This chemical technology applies paint-like polymer coatings onto the contaminated surfaces which contain strong binding agents to extract and bind the contaminants during the coating's curing/drying. The cured coating is peeled off from the surfaces with the contaminants. The coating can be applied using typical paint finishing methods such as by brush, roller, or sprayer. For maximum decontamination,

multiple strippable coating applications are required (USEPA, 2013a 2013c; Heiser, 2009). Strippable coatings can be also used to control loose contaminations that present airborne hazards. Solid waste is generated, and these methods are best suited for relatively small decontamination operations. The major disadvantages of using strippable coatings are that they can remove only loose contamination, and the application and removal processes could be labor-intensive. An alternate use of strippable coatings is to temporarily cover and fix the contaminants on surfaces to prevent further migration of the contaminants while awaiting decontamination operations to begin (USEPA, 2016; Parra, 2009). Published DF values include 11 or higher for removable contaminants on non-porous surfaces and painted carbon steel (Gates-Anderson, 2012), and 1.4–2 and 4–5 for aged and fresh Cs-137 contamination, respectively, on porous concrete, limestone, and asphalt surfaces (USEPA 2013c). Excluding wait and setup times, reported decontamination speeds were 1–1.5 m²/h, with higher rates of 17 m²/h for coating application and 43 m²/h when a sprayer was used for large-area decontamination (Gates-Anderson, 2012). Some of these technologies have been evaluated on aged urban building surfaces in a large scale, and the results showed some limitations on peel off from certain surface types (USEPA, 2016).

4.2.3. Clay film coatings (Appendix A.10 operational information sheet)—

Clays are naturally occurring and ubiquitous minerals, some of which show relatively high sorption affinity for alkali, alkaline earth, and transition metal cations, making clay coatings a choice for decontamination, particularly if appropriate clays are widely available. The clay is applied as a paste to form a film on the surface. Studies have shown that as a sorbent, clay films can enhance desorption of radionuclides from porous surfaces including smooth concrete, brick walls, and roofing slate. This technology was used for cleaning building structural surfaces after the Chernobyl NPP accident. The reported DFs were 2-20 depending on surface types and age of the contamination (Kaminski, 2016). The DF values tend to decrease with increasing contamination aging time. In recent decontamination efforts after the Fukushima NPP accident, clays have been widely used as sorbents for waste water treatment (JAEA, 2015b).

4.3. Remediation of roadside soil, roadside vegetation, and nearby land

Roadside remediation is particularly important for restoring infrastructure access. In general, the first step for decontamination is to remove the contaminated vegetation atop the ground, and then depending on the contamination levels, to either strip off highly contaminated topsoil or exchange low-level contaminated topsoil with underlying subsoil. These approaches should be carefully assessed for the future land use prior to implementation because the radioactive contaminants may be revealed via site renovation or reconstruction in the future.

Table 3 summarizes the remediation techniques and technologies for vegetation and soils (JAEA, 2015b) considered to be most relevant to soluble radiological contamination because these technologies address the possibility that the radiochemical has migrated from the surface, penetrating deeper in the soil/land. However, the original data sources did not study whether the contaminant was present in soluble or insoluble forms, although it seems reasonable that because most of the reported contamination was cesium, which is highly

water soluble, that at least part of the content in Table 3 resulted from water contaminated with cesium that entered the vegetation and soil. The original data sources for Table 3 also do not compare the depth of radionuclide penetration into the surface. In the case of the Fukushima NPP accident, the reported soil contamination depths were mostly within the top five cm of the soil layers, but this could vary greatly across locations. In Table 3, the differing techniques and technologies have different capabilities to remove various depths with precision. In general, the larger the equipment, the less precision and control. Reported DFs are summarized in Table 3, and the wide variation may result from site specific conditions, skill of equipment operator, and many other factors. Thus, the DFs tabulated should be viewed as informative but not as being generally predictive.

The names of techniques and technology in Table 3 are self-descriptive as to their function and operation. Some involve manual labor, and some utilize common agricultural or construction equipment. Thus, no general descriptions are provided, as they were for the less familiar technologies discussed above. Technology operational considerations for implementation are provided in Appendix B of the Supplemental Information, such as:

- Speed of implementation for a specified piece of equipment or a team of workers;
- Generated waste, both volume and characteristics are listed. However, subsequent waste treatment may reduce the volume;
- Effectiveness in terms of a DF and/or a % gamma dose rate reduction;
- Cost of equipment and/or labor. As labor costs vary dramatically between localities, the absolute value given is of little relevance, but the relative costs are probably a good indicator;
- Technology gaps, including strengths and weaknesses, as well as desirable features/ functions;
- Environmental safety and health issues;
- References for the listed information. This may include vendors that provided technical data, but their inclusion does not imply endorsement.

5. Remediation technologies for water insoluble radiological contamination of roadways and roadsides

As summarized above, the bulk of the recent wide-area radiological decontamination literature has focused on the removal of mobile water-soluble species. This leaves open the question of the best methods to remediate *insoluble* particulate contamination. Insoluble particulate contamination from a nuclear detonation or RDD can differ from nuclear/radiological accidents (like a nuclear reactor meltdown). Many of the technologies for water soluble contaminants rely on physical removal, so, in principle, may also be applicable to physical removal of insoluble particles. However, the composition and morphology of the particle, as well as characteristics of the contaminated surface, can greatly influence the ability of a technology to remove such particles.

As also noted by Kaminski (2016), much of the particulate cleanup literature to-date has also focused on traditional methods that: a) generate high volumes of secondary waste (e.g., fire hosing); b) can potentially re-disperse the contamination (e.g., street-sweeping); and/or c) apply to mainly “loose” dust-like particulate contamination. Some of the available radiological-site decontamination literature from actual nuclear detonations or accidents/incidents have characterized particulates, but few literature examined *remediation* of particulates and, of those that did, largely only superficial characterizations of the treated particulates were performed. Only 7 of the 41 relevant technologies reviewed in this manuscript reported the characteristics of the radiological particulates. Some reports included multiple particle types. Four examined “loose” particles (one for “large” particles, three for $<2\ \mu\text{m}$ particle sizes), and one described nuclear-blast-derived plutonium particles. Two reports included no particle description, with one of these being a survey method for airborne radioactive particulates; however, it was included here due to the lack of applicable technologies. No mention of the particle solubility, chemical composition, and/or surface roughness of remediated particles was described, except in literature simply reporting particulates found at some of these radiological sites. Also, particle size effects on decontamination efficiencies were not found, except in technology tests (not considered in this paper) using artificial contamination (Jolin, 2019).

The following subsection describes the available literature, with operational information in Supplemental Information Appendix C. The content below overlaps with technologies for water-soluble contaminant removal in Section 4. This occurs chiefly because of the similarities in the underlying operation of the technology, even if the underlying chemistry of water-soluble contamination differs from insoluble particulate contamination. The discussion of their operationally relevant characteristic is similar to Appendix A, so is generally not repeated in the discussion below. It is included in both Appendices for the convenience of the reader, but mainly because the Appendix C sheets provide information specific to particulates. Appendix C sheets also include characteristics of the particles remediated and surfaces from which the particles were remediated.

To the extent possible, the dose reduction efficiencies, costs, and other information included in the operational information sheets in Appendix C were taken from remediation of sites resulting from real events while excluding results from models or artificially contaminated site experiments. Also, data from remediating dissimilar indoor surfaces contaminated by other means (e.g., radioactive “hot cells”, accelerators, uranium metal mills, mines) were also avoided to provide as realistic information as possible applicable to decontamination of roadways and roadsides. When field evaluations of individual technologies were available (JAEA, 2015b), those “not recommended” in the JAEA report (e.g., tree trunk washing near Fukushima) were also excluded.

5.1. Decontamination technologies with data directly applicable for particulates

5.1.1. Street/pavement sweeper and washing vehicle (Appendix C.1 operational information sheet)

—Vacuum equipped, municipal street sweepers with a water nozzle can pre-spray the road with a fine mist before using (usually three) rotating brushes to pick up soil, road dust (i.e., large particles), and vegetation. They have shown

varying maximum efficiencies of 29–60% (based on Cs measurements) (MOE, 2014; Roed, 1995; Sutton, 2016). Generally large area coverage is possible (largely, ~1750–3500 m²/day in a single-seat sweeper), but the asphalt/surface should be smooth, undistorted, and undamaged. These types of sweepers are commonly available. Water-spraying is critical to avoid secondary contamination and worker dust exposure, and low waste volume is generated (e.g., 50–200 g/m², 1–1.5 L/m²).

Street-sweeping of model radio-tracer laden sand from asphalt yielded the expected result of higher cleanup effectiveness with large particles (350–700 μm) at low surface mass loadings (3 kg/m²) and lower efficiency at high initial mass loadings (10–65 kg/m²) on asphalt surfaces with small (44–88 and 88–177 μm) particles (Kaminski, 2016). However, opposite results were reported in cold weather decontamination of nuclear detonation fall-out where residual activity for removal of small 20–75 μm particles was 2–9 times higher than for cleanup of large (150–300 μm) particles (Maloney, 1967).

5.1.2. Dry and steam vacuum cleaning (Appendix C.2 and C.3 operational information sheets)—A portable high-performance HEPA filter equipped vacuum system can be very effective (e.g., 99.999% if used in a 3-step process; Gates-Anderson, 2012) in removing loose radioactive particles (e.g., ~0.1–2 μm) from a variety of surfaces, although particle removal efficiency from brick and concrete was 20–45% even with slow sweeping rates (IAEA, 1989). These systems can cover ~12 m²/h and are readily available. Application of steam during vacuuming corresponds to separate operational information (Appendix C.3) when considering radioactive particulates.

5.1.3. Low pressure water washing (e.g., with household or fire hoses) (Appendix C.4 operational information sheet)—A standard fire hose used to rinse a surface contaminated with large nuclear fallout particles has shown potentially high efficiency (~60–70% (Kaminski, 2016), with a low 9% efficiency reported in some cases (Roed, 1995)). Large areas of concrete, single-composition roof shingles, and roads can be sprayed quickly (~100 m²/h) at low operating (e.g., 0.0013 man-day/m²) and capital cost. However, it also can create a large volume of contaminated wastewater which, if not properly contained/managed, will result in the potential secondary contamination of soils and drinking water supplies. In this regard, the IWATERS system, described for water-soluble contamination (Section 4.2.1), may also prove applicable for this waste management challenge. The IWATERS system inherently incorporates a particle removal step, provided the particle size is compatible with the on-site filtration system (Kaminski, 2015).

5.1.4. High-pressure water-jet washing with manual wiping as polishing step (Appendix C.5 operational information sheet)—High pressure washing can remove many particulates but may need supplemental manual wiping as a polishing step for stubborn particles not adequately impacted by the high pressure stream. A high-pressure (e.g., 8 MPa or 1160 psi) water wash has been demonstrated to remove 45–90% of 0.1–2 μm radioactive particles from brick and concrete surfaces at sweep rates of 0.2–10 minutes/m² (IAEA, 1989). Typical treated surfaces include brick, asphalt, concrete, smooth walls (e.g., steel, glass), roofs, and roof gutters, although cesium-based measurements for the last three surfaces have shown lower ~23–60% efficiency. These systems are not costly to acquire,

but slower throughput (1–37 m²/h) can lead to higher cost operations and the need to dispose of high volumes of liquid waste. Care should also be taken to avoid splatter, secondary contamination, and unintended water penetration. Surface pre-testing is required for adequate stripping depth, and pre-treatment (e.g., gutter clearing) is also sometimes necessary.

5.1.5. Shot/media blasting (Appendix C.6 operational information sheet)—

Shot (e.g., steel) or blasting media (e.g., sand, glass beads) is injected into high-pressure air or water streams to remove surface contamination by stripping off a portion of the surface to adequate depth (~2–3 mm is typically necessary). Both wet and dry abrasive jet cleaning has been employed many times in the nuclear industry for diverse applications from decontaminating highly contaminated pipework to lightly contaminated surfaces. Nuclear-blast plutonium particulate contamination was demonstrated to be best removed by sandblasting (Warming, 1984). A 60–95% dose reduction (based on Cs) has been achieved on asphalt pavement, walls, and concrete floors (MOE, 2014; Sutton, 2016), but secondary contamination and worker dust exposure is a high risk without a vacuum dust collection system, which can lead to a higher cost. Operating costs are high (hundreds of dollars per m²), as are waste volumes generated (of order 3–20 L/m²). Practically, the working surface should be fixed and flat (and dry for dry-blasting), away from corners and narrow areas, and pre-testing is necessary to determine blast density for sufficient stripping. Vertical surfaces can also be difficult, but this method is especially good for removing paint and light coatings on concrete surfaces.

5.1.6. Strippable coatings (Appendix C.7 operational information sheet)—

Depending on the mixture, a liquid, foam, gel or paste is sprayed, rolled, or applied onto a hard (porous or non-porous) surface to encapsulate and bind loose smearable radioactive particles (e.g., 0.1–2 μm). It is allowed to dry/cure, and then removed by peeling off the hardened layers. Strippable coatings (~5 to ~50 mils thick) can be used to control airborne contaminants or as a fixative on structures to be demolished. Sample surfaces include painted masonry, concrete, fired clay tiles, wood, metal, plaster, glass, and painted equipment/vehicles. See Appendix C.7 for a more complete list, including use on lawns, flower beds, and other specialized surfaces because they may not be commonly thought of for this type of decontamination technology. A loose particulate removal efficiency of 81–94% was demonstrated (Boing, 2006). This method can involve ~2–48-h curing times per coating, requires worker protective equipment for application and careful removal by hand that can lead to radiation dose, but has the advantage of reduced dust exposure. Equipment application can be costly, coating application fairly slow (of order several m²/h), and waste generation of e.g., ~0.1 kg/m². Raw material shelf-life and application temperature restrictions may apply, and this method may not be suitable for complex surface geometries.

5.1.7. Fixed coatings (Appendix C.8 operational information sheet)—

Depending on the material, a liquid mixture (e.g., water-based acrylic, two-part epoxy resin, concrete) is sprayed, brushed or applied to wet loose particles and bind them to surfaces to prevent resuspension or movement. The applicability of the different types of fixed coatings depends on the surface type. Some require ambient temperature (>~15 °C) application

and/or storage, although some others can be applied as low as freezing temperature. This technology is applicable to most hard surfaces such as metal, concrete, plastic, equipment, debris, and rubble. In some cases, it has reasonably good dose reduction rates when the coating also provides shielding, although shielding potential varies with the coating and type/strength of radiation. For example, 67% Cs gamma radiation reduction was reported when 5 cm concrete was used on a particular surface (IAEA, 1989; Gates-Anderson, 2012). Equipment and material costs can be low, but coatings can require ~8 to 48-hour curing/dry times and can sometimes have stock and application temperature limitations.

5.2. Other likely applicable technologies to radiological particle contamination

As noted above, techniques and technologies for roadside vegetation, soil, and land (Table 3) were not studied as to whether the contamination was in soluble or insoluble form. However, because the mode of action is removal of contaminated substrate, the techniques and technologies in Table 3 should be applicable to contaminated particles. Likewise, technologies for soluble contamination that rely on physical removal (Section 4.1) might also be applicable to particulates, with the primary difference being the depth of penetration of the particles (compared to penetration of water soluble contaminants carried by aqueous flow). Many particles are expected to remain on or near the surface, unless the surface has crevices which the particles can enter. Although many physical removal techniques are discussed already, it is important to keep in mind that the characteristics of the particulates might influence the removal efficiency and potential resuspension concerns; these factors should be considered prior to implementation.

Repaving and resurfacing approaches may also be applicable, although the characteristics of the particles may be important to consider, especially if such particles are resuspended or tracked during the repaving or resurfacing process. Procedural modifications may therefore be required, along with adjustments to typical health and safety plans for workers.

In addition to roadways and roadsides decontamination technologies and repaving/resurfacing, two other techniques are appropriate to discuss for remediating associated infrastructure in addition to roadways. As mentioned above, infrastructure, like toll booths, maintenance garages, etc. can largely be considered the same as other buildings and urban infrastructure. The discussion here is not intended to be exhaustive because it overlaps with the topic of remediation of indoor/outdoor urban areas, which is reviewed elsewhere (Kaminski, 2016). Hence, these two infrastructure-associated techniques are specific to signage, including billboards: motorized brushing and paper (billboard) removal. Signage is often painted, covered in plastic, or otherwise inherently impermeable, such that radioactivity present might be physically associated as particles, or at least not having penetrated the surfaces, unless the surface is unusually weathered or somehow chemically reacts with the contaminant (which is less likely but possible). For example, many roadside signage is constructed of the same materials as other parts of the urban environment (Kaminski, 2016) and might be expected to be remediated similarly. In Appendix C, operational information sheet C.9 describes an example of a motorized brushing system perhaps suitable for signage. Other motorized brushing systems may be available from the local cleaning industry. Sheet C.10 describes removal of paper billboards which are assumed

to be plastic coated. Brushing and paper removal systems will also vary by location, along with specific operational details.

6. Conclusions

6.1. Summary of remediation techniques for roadways and roadsides

For hard surfaces requiring decontamination of soluble radionuclides, established physical cleaning methods identified generally have low (<50%) decontamination efficiencies. However, it is important to keep in mind that even modest (50%) reduction in dose may be significant enough to enable personnel to operate in contaminated areas for extended periods of time. This may be important early in an incident due to a shortage of qualified radiological workers. As with all technologies, temporary high-volume waste storage is likely a key factor in early implementation.

Among the general classes of remediation technologies for physical removal with high throughput, fire hosing may be the most “proven” high-volume particulate removal technology with literature suggesting as high as 60–70% removal (DF of 2.5–3.3), but possibly having a low performance floor of 9%, accompanied by high liquid-waste volumes and secondary contamination risks. The throughput of some methods such as street sweepers can be high (i.e., hundreds of m²/h). Vacuum street sweeper/washers are common, and pre-trained operating personnel might be available to permit early implementation. Some municipalities’ vacuum sweeper fleets are global positioning system (GPS) enabled, contributing to the logistics of their deployment.

Shot blasters also provide higher asphalt road coverage rates (~300–3500 m²/day). However, like street sweepers, particulate-based removal efficiencies are lacking, despite other contaminants suggesting a 60–95% shot blasting efficiency (depending on operations). Although the operating costs for these techniques are low (of order tens of cents to several dollars/m²), the capital cost and delivery times can be significant, if unavailable nearby. Shot blasting equipment with vacuum is somewhat lower in cost than street sweepers to acquire but at least an order of magnitude higher in labor costs to perform (of order several dollars/m²). The sweeping and blasting methods likely will require multiple applications, in addition to being limited to smooth, dry, undistorted/undamaged roads. Again, temporary high-volume waste storage is likely a key factor in early implementation.

Among lower throughput approaches, typically because they involve more manual labor, physical methods removing a thin surface layer (e.g., blasting, grinding) generally can provide higher (up to ~90% or DF ~10) cleanup efficiencies while providing lower throughput (of-order hundreds of square feet per hour). Washing and sandblasting methods can generate significant secondary waste (e.g., contaminated wastewater and grit material), while others like grinding and shaving produce minimal additional waste. Of the chemical treatment approaches (i.e., coatings, foams) for porous hard surfaces, the removal efficiencies can vary significantly with minimum DFs reportedly as low as ~2, with higher values suggested for non-porous surfaces like painted carbon steel (~11). Available treatment rates were in the m²/h. Strippable coatings have demonstrated as high as 81–94% loose particulates removal and are applicable to a variety of porous and non-porous hard

surfaces. Tens of m²/day application rates are achievable, although drying/curing times may be significant, and multiple applications may be required. Fixed coatings, especially application of a thin layer (~5 cm) of concrete (67% dose reduction of ¹³⁷Cs gamma), could be a relatively high-throughput and low-cost technology, but no literature treatment rates were found. Dry vacuum cleaning of brick and concrete can be as low as ~20–45% of loose <2 μm particles, and one vendor claims a very high efficiency using a 3-step process. HEPA-filtered vacuums can be high cost in both acquisition and operation, but some models allow multiple workers on a single unit. High-pressure washing can suffer from relatively low effectiveness and can require very slow sweep rates to achieve high efficiency (45–90% at 0.2–10 min/m², respectively). In addition, high-pressure washing likely may require significant prepreparation for contaminated water runoff collection.

Similar to road sweepers, resurfacing/repaving of contaminated surfaces is an established approach with known throughput rates and sometimes limited (although potentially beneficial) dose reduction potential (50–75% of dose reduction by applying 5–6 cm of asphalt). However, there are long-term concerns regarding the potential for re-exposure or migration of the original contamination as the applied asphalt ages and/or wears.

Experience from work around the Fukushima NPP indicates that roadside soil and vegetation can be significantly reduced by eliminating a thin layer of contaminated soil after removal of overlaying vegetation. Trimming and mowing of vegetation provided widely varying results (~10–90% dose reduction), but burial or removal of centimeters-deep surface soil (e.g., tillage and stripping, respectively) provided DF greater than 2.8 (~65% reduction).

For insoluble radiological particulate decontamination, very little data was found in the available published literature. The few high-throughput technologies (e.g., fire hosing, vacuum street sweeper/washers, shot blasting) offered similar maximum decontamination efficiencies (e.g., DFs up to 3.3, or 70% removal) but also indicated almost no efficacy in some situations. Fixed coatings, especially application of a thin concrete layer (~5 cm), could be a relatively high-throughput and low-cost technology, although no literature treatment rates were found. Sandblasting of open areas (e.g., concrete floors, roofs) has shown relatively high treatment rates (hundreds of m²/day), but no published radiological particulates-based decontamination efficiencies were identified. Strippable coatings have demonstrated relatively high (81–94%) loose particulates removal and are applicable to a variety of porous and non-porous hard surfaces at lower area throughputs. Technologies that do not cover a lot of surface area quickly for particulate-contaminated surfaces (dry vacuum cleaning, high-pressure washing) generally show higher potential decontamination efficiencies (with wide scatter in reported data) and require waste collection and handling. A number of technologies potentially capable of insoluble particulate remediation were also identified from the available literature, with many of the efficacy data being applicable to soluble contamination, *not* insoluble.

6.2. Future work

Opportunities for future work include significant research in expanding the knowledge of the remediation of insoluble particulates from nuclear accidents and incidents such as nuclear reactor meltdowns (e.g., Chernobyl, Fukushima), nuclear detonations, and other nuclear

accidents. This would help fill the technical gaps summarized in the Operational Information Sheets in the Appendices, which may also be a helpful resource for inferring needs in future technology development and/or improvements, as well as for identifying which missing data (e.g., contaminant, throughput, cost) to focus on deriving in future field studies.

Possible research opportunities include: a) increased study of remediation of insoluble radiological particulates isolated at contaminated sites (especially at Fukushima and indoor facilities); b) more detailed analysis of identified particulates to better understand particle physical and chemical characteristics (e.g., particle size distribution, aqueous solubility/insolubility, chemical composition as they change with distance from release point, environment/weather, age, etc.); and c) more studies involving insoluble particulates. Some of this may be accomplished by re-analyzing both earlier samples for particulates and earlier test data. Similar work at former nuclear weapons test sites may help expand knowledge in relation to RDD/IND derived contamination and remediation.

There appears to be potential work to understand how to best combine multiple technologies and techniques to enhance the results of the overall remediation effort. Such combinations may also help achieve the goal for starting these activities as soon as possible. For example, dedicated road sweepers can be combined with dedicated vacuum cleaning equipment to overcome their individual limitations relative to radionuclide decontamination. The required equipment are fairly common, and pre-trained operating personnel are likely available to permit early implementation.

Finally, work is warranted to apply the information above for radioactive contaminants to challenges with other types of contaminants (e.g., toxic chemicals), particularly when the underlying chemistry and physics of the radioactive contaminants are similar to those of non-radioactive ones. While some techniques for radioactive materials may be unnecessary for contaminants with lesser toxicity concerns, some may be applicable to non-radioactive contaminants with high persistence and human toxicity, or that present other environmental toxicity or bioaccumulation concerns. To do so may significantly help with the “dearth” of scientific information on the topic (Amato, 2010), and perhaps enhance the application of the technologies using the implementation strategies and novel equipment application suggested for radiological decontamination (Kaminski, 2018).

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Table 1
Qualitative analysis of remediation technologies (Reproduced from USEPA, 2013b).

Criterion	Scarification	Vacuuming	High-Pressure Washing	Street Sweeping
Safety, health & environment	H/A	M/N	M/N	M/N
Time to implement	M/N	M/N	H/A	M/N
Technical performance	H/A	M/N	M/N	M/N
Availability	M/N	H/A	H/A	H/A
Costs	M/N	M/N	M/N	M/N
Process waste	H/A	M/N	L/NA	M/N
Throughput	M/N	M/N	M/N	H/A

Green (H/A) = High/advantageous; yellow (M/N) = medium/neutral; red (L/NA) = low/not advantageous

Table 2

Conversion of decontamination factor to percent removal of contamination,

DF	Removal (%)
1.11	10
1.33	25
2.00	50
4.00	75
10	90
20	95
100	99
1000	99.9

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Table 3

Techniques and technologies applicable to remediation of roadside soil, roadside vegetation, and nearby land.

Chief applicability	Technique or technology	DF ranges (depends on depth of removal)	Appendix operational information sheet
Roadside vegetation	Manual litter and humus removal	1.1-10	B.1
	Fallen Leaf and thin layer topsoil removal	1.3-5	B.2
	Lower tree Branch Trimming	1.2-2.1	B.3
	Tree Trimming & Underlying Soil Removal	2.5-5	B.4
	Tree felling	50	B.5
Roadside soil and land	Soil Tilling/Plowing	1.4-10	B.6
	Topsoil Exchange	4-15	B.7
	Topsoil Stripping by Hammer Knife	1-17	B.8
	Topsoil Stripping by Digger/Front Loader	1-100	B.9
	Topsoil Stripping by Road Surface Stripping Equipment	1.2-3	B.10
	Topsoil Stripping by Motorized Land Grader	5-10	B.11
	Manual Topsoil Stripping	1.2-1000	B.12
	Topsoil Stripping with Mechanical Excavator/Digger	1.1-20	B.13
	Gravel Stripping	1-13	B.14
	Turf Stripping/Manual Sod Cutting	2-20	B.15
	Topsoil Stripping with Surface Hardening (Solidification)	1.7-5	B.16