Estimation of secondary waste loads generated during environmental restoration activities

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Abstract

The US Department of Energy's (DOE's) Environmental Restoration (ER) program is responsible for remediating thousands of contaminated sites across the DOE complex. A major concern during remediation is the secondary waste generated as a result of ER site remediation. The treatment, storage and disposal (TSD) of secondary waste generated as a result of ER site remediation is the responsibility of the waste management (WM) program. In order to manage TSD operations on ER generated waste, WM needs accurate and consistent estimates of the volume of secondary waste generated. This paper discuss three remedial processes and presents estimates of waste volumes generated during these processes.

1. Introduction

A legacy of the nuclear weapons program is environmental contamination at the sites where research, development, test and production of nuclear weapons took place [1]. The volumes of contaminated media at some locations are quite large. The environmental restoration (ER) program was established to address this problem. It encompasses a wide range of activities such as stabilizing contaminated soil, treating soil and groundwater, decommissioning process buildings, including nuclear reactors and chemical separations plants, and exhuming buried drums of waste. The treatment, storage and disposal (TSD) of secondary waste generated as a result of ER site remediation is the responsibility of

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the waste management (WM) program. In order to manage TSD operations on ER generated waste, WM needs accurate and consistent estimates of the volume of secondary waste generated.

In a preliminary effort to understand the secondary waste generation during ER activities, we estimated waste volumes for three remedial processes:
1. soil washing and carbonate extraction of soils contaminated with uranium and PCBs;
2. sulfide precipitation of mercury from water;
3. capping of contaminated soil sites.

2. Soil washing and carbonate extraction of soils contaminated with uranium and PCBs

Soil washing is a process in which a liquid solution is used to remove contaminants from soil. The solution used in washing depends upon the type of contaminant to be removed [2]. For soil contaminated with PCBs and uranium, soil washing followed by carbonate extraction provides a means for removing a majority of both contaminants from the soil. Soil washing using a washing solution of water and a surfactant has been shown to remove 95% of PCBs from soil [3]. Soil washing with water is not effective in removing uranium from contaminated soils. The water/surfactant wash fluid used to remove PCBs will only remove ~ 5% of the uranium present in the soil [3]. Treatability studies conducted at the Oak Ridge National Laboratory (ORNL) on uranium contaminated soil samples show that a solution of sodium carbonate and sodium bicarbonate used in a carbonate extraction process can remove an average of 84% of the uranium from soil [3]. Soil washing followed by carbonate extraction can remove ~ 95% of the PCBs and ~ 84% of the uranium initially present in the soil.

2.1. Process description

Soil washing and carbonate extraction are processes that require excavation of the soil prior to treatment. This study assumes that the soil has already been excavated, and excavation is not a component of the treatment train. Density separation of the excavated soil often precedes soil washing to segregate the coarse and fine soil particles because the fine soil particles typically have much higher concentrations of contaminants than coarse soil particles [4]. Coarse soil particles are rinsed, dewatered and returned to the site as clean soil without undergoing further treatment. This greatly reduces the volume of material treated, the amount of waste generated, and the cost of treatment.

The first step in soil washing, as shown in Fig. 1, consists of mixing the contaminated soil and the water/surfactant solution in an attrition scrubber. The blending action of an attrition scrubber causes the soil particles to collide. These collisions result in abrasion of the soil particles, and consequently, the removal of weathering products (iron and manganese oxides) on the soil surfaces that would otherwise inhibit contact between the washing solution and the soil particle.

After washing, the slurry passes to a centrifugal separator where the soil and washing solution are separated. The soil is rinsed with clean water and then passes to a second
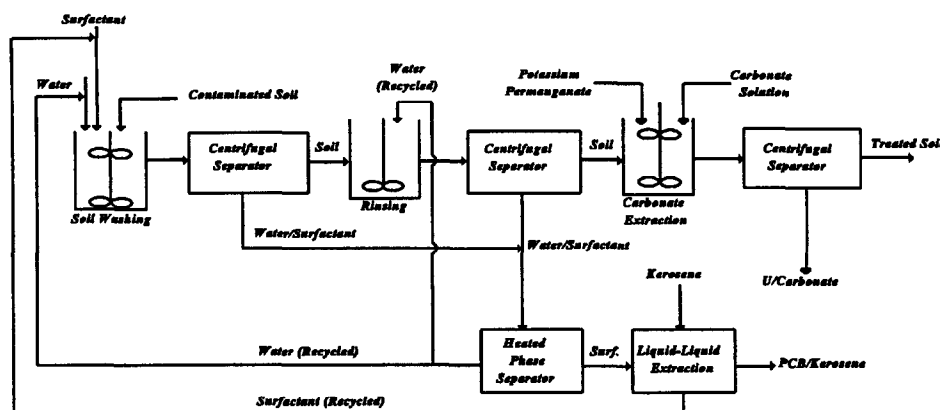
Fig. 1. Flow diagram for soil washing and carbonate extraction of PCB and uranium contaminated soil.

centrifugal separator which separates the rinse water and the soil. The rinsed soil is still contaminated with uranium, but $\sim 95\%$ of the PCB contamination has been removed.

The water/surfactant solutions from both centrifugal separators are piped to a heated phase separator which removes PCB contaminated surfactant from the water. A liquid–liquid extraction process utilizing kerosene as the solvent removes the PCBs from the surfactant. Both the surfactant and the water are recycled and used again in the soil washing process. The PCB/kerosene solution is a generated waste stream requiring further treatment or disposal.

The uranium contaminated soil emerging from the second centrifugal separator is first mixed with an oxidant which converts the tetravalent uranium covering the soil particles into a hexavalent form. Carbonate extraction cannot occur unless the uranium is in hexavalent form. Permanganate salts have been shown to be effective oxidants in treatability studies conducted with soil samples from the Oak Ridge Y-12 Plant [3,5].

A solution of sodium carbonate and sodium bicarbonate is then mixed with the soil in an attrition scrubber. The attrition scrubber greatly decreases the amount of contact time between the extractant solution and the soil that is necessary to remove uranium. Treatability studies show that the use of an attrition scrubber reduces the time from 4 h to approximately 15 min [5]. The hexavalent uranium reacts with the carbonate solution to form the stable complex $UO_2(CO_3)_3$. The entire reaction is shown in the following equation:

$$3UO_2 + 2MnO_4 + 9CO_3 + 4H_2O \rightleftharpoons 3UO_2(CO_3)_3 + 2MnO_2 + 8OH \tag{1}$$
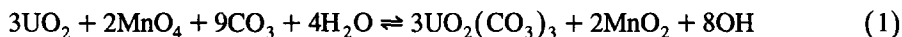
The treated soil slurry then passes to a centrifugal separator which removes the contaminated solution from the soil. The treated soil can usually be returned to the site. If the concentration of contaminants remaining in the treated soil is greater than the allowable limits, additional treatment or disposal of the soil is required.

2.2. Waste volume estimation

The wastes generated from soil washing and carbonate extraction consist of: (1) the personal protective equipment (PPE) worn by the workers during the removal process

like gloves, glasses, and suit; (2) PCB/kerosene solution generated as a result of soil washing; and (3) carbonate solution generated during carbonate extraction. To estimate the volume of waste generated per volume of soil that is treated, it is necessary to make assumptions regarding the amount of soil treated per day and the number of workers involved in the treatment. The following calculations assume that 30 m³ of soil (approximately 50 tons) can be processed daily [1,6], and that four workers are required to operate the system. To estimate the volume of PPE that is generated, it is assumed that each worker fills a 55 gallon ($\sim 0.21\,\mathrm{m}^3$) drum with PPE every 2 days.

The volume of PPE generated is:

$$V_{\mathrm{PPE}} = \frac{4\,\text{workers} \times 55\,\mathrm{gal}_{\mathrm{PPE}}}{2\,\text{days}} \times \frac{1\,\text{day}}{30\,\mathrm{m}^3_{\mathrm{Soil}}} = \frac{0.014\,\mathrm{m}^3_{\mathrm{PPE}}}{\mathrm{m}^3_{\mathrm{Soil}}} \tag{2}$$

Full-scale soil washing systems generate 14 lb of PCB/kerosene solution per ton of treated soil [3]. Assuming that the density of the PCB/kerosene mixture is equal to the density of kerosene, $0.82\,\mathrm{g\,cm}^{-3}$, and assuming a soil density of $1.5\,\mathrm{g\,cm}^{-3}$, the volume of PCB/kerosene solution generated per cubic meter of treated soil is:

$$V_{\mathrm{PCB/K}} = \frac{14\,\mathrm{lb}_{\mathrm{PCB/K}}}{1\,\mathrm{ton}_{\mathrm{Soil}}} \times \frac{1.5\,\mathrm{g}_{\mathrm{Soil}}}{\mathrm{cm}^3_{\mathrm{Soil}}} \times \frac{1\,\mathrm{cm}^3_{\mathrm{PCB/K}}}{0.82\,\mathrm{g}_{\mathrm{PCB/K}}} = \frac{0.013\,\mathrm{m}^3_{\mathrm{PCB/K}}}{\mathrm{m}^3_{\mathrm{Soil}}} \tag{3}$$

The data obtained for carbonate extraction is based on treatability studies and not full-scale operations. In the treatability studies, equal amounts of soil and carbonate solution were mixed to remove uranium from the soil samples. It is estimated that the same would be true for a full-scale carbonate extraction system [5,7]. The calculation which follows assumes that equal amounts of soil and carbonate solutions are mixed, so that for each gram of soil there will be 1 g of carbonate solution. Again assuming a density of $1.5\,\mathrm{g\,cm}^{-3}$ for soil, and assuming that the density of the carbonate solution is the same as the density of sodium carbonate (i.e. $2.5\,\mathrm{g\,cm}^{-3}$), the waste generated as a result of carbonate extraction is:

$$V_{\mathrm{Carb}} = \frac{1\,\mathrm{g}_{\mathrm{Carb}}}{1\,\mathrm{g}_{\mathrm{Soil}}} \times \frac{1.5\,\mathrm{g}_{\mathrm{Soil}}}{\mathrm{cm}^3_{\mathrm{Soil}}} \times \frac{1\,\mathrm{cm}^3_{\mathrm{Carb}}}{2.5\,\mathrm{g}_{\mathrm{Carb}}} = \frac{0.6\,\mathrm{m}^3_{\mathrm{Carb}}}{\mathrm{m}^3_{\mathrm{Soil}}} \tag{4}$$

The total volume of waste generated as a result of soil washing and carbonate extraction of soil contaminated with PCBs and uranium is the sum of the PPE waste generated, the PCB/kerosene solution, and the carbonate solution:

$$V_{\mathrm{Total}} = \frac{0.014\,\mathrm{m}^3_{\mathrm{PPE}}}{\mathrm{m}^3_{\mathrm{Soil}}} + \frac{0.013\,\mathrm{m}^3_{\mathrm{PCB/K}}}{\mathrm{m}^3_{\mathrm{Soil}}} + \frac{0.6\,\mathrm{m}^3_{\mathrm{Carb}}}{\mathrm{m}^3_{\mathrm{Soil}}} = \frac{0.63\,\mathrm{m}^3_{\mathrm{Waste}}}{\mathrm{m}^3_{\mathrm{Soil}}} \tag{5}$$

3. Sulfide precipitation of mercury from water

Sulfide precipitation is used to remove mercury from contaminated wastewater streams [8]. A flow diagram of the process (Fig. 2) depicts the basic components of a
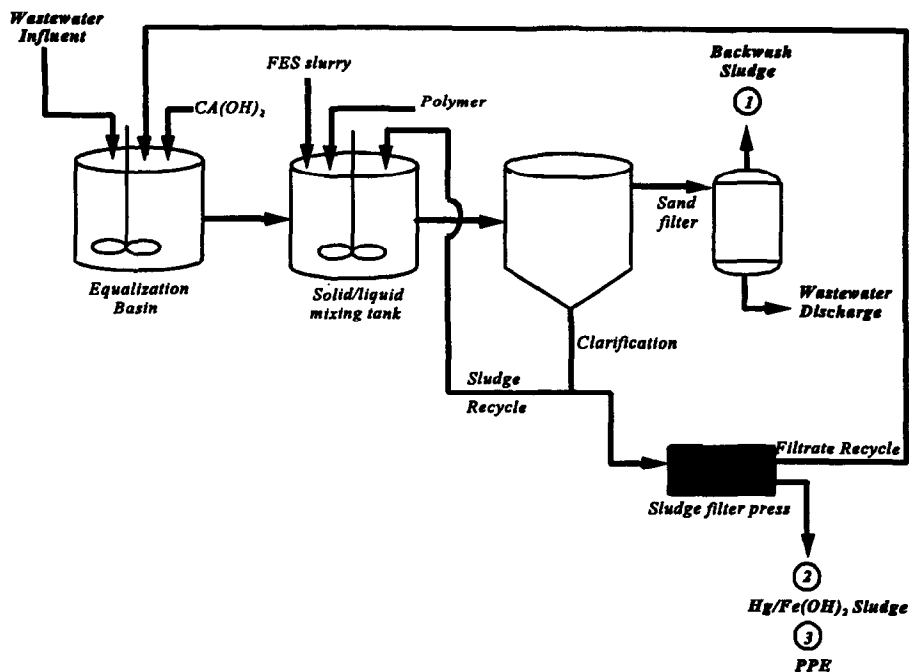
Fig. 2. Insoluble sulfide precipitation flow diagram.

typical insoluble sulfide precipitation (ISP) treatment train. Approximate volumes of RCRA hazardous waste from this process are estimated from the influent mercury concentration and the contaminated wastewater volume.

3.1. Process description

A precipitation reaction occurs when two electrolyte solutions are mixed and an insoluble solid falls out of solution. This concept is often used in industry to remove dissolved metals from a wastewater stream as solid precipitates which are easily removed from the stream by settling and filtration [9]. This process describes the application of a precipitation reaction to a wastewater stream to remove dissolved mercury by precipitating it as mercury sulfide (HgS). The general metal sulfide precipitation reaction is described by:

$$M^{2+} + S^{2-} \rightarrow MS \downarrow \tag{6}$$

For the ISP process, ferrous sulfide (FeS) is used as the source of the sulfide ions. The equations governing the series of reactions for the ISP of mercury are

$$FeS \rightarrow Fe^{2+} + S^{2-} \tag{7}$$

$$Hg^{2+} + S^{2-} \rightarrow HgS \downarrow \tag{8}$$

One general disadvantage of sulfide precipitation is the potential release of excess sulfide in the treatment effluent stream [10]. To eliminate this problem ferrous sulfide is

used as the source of the S^{2-} ions in a process known as insoluble sulfide precipitation. FeS has a low solubility and will maintain a 0.02 ppb sulfide ion concentration in the reactor vessel, eliminating the need for a post-treatment sulfide removal system [10]. This low S^{2-} concentration is adequate to precipitate the mercury ions, and as the sulfide ions are consumed by this process, additional FeS will dissociate to maintain equilibrium.

Another potential problem with sulfide precipitation is the formation of toxic and malodorous hydrogen sulfide (H_2S) gas. The low solubility of FeS reduces this risk by minimizing the concentration of S^{2-} ions available for reaction with hydronium ions (H_3O^+) present in the wastewater. Addition of calcium hydroxide ($Ca(OH)_2$) to maintain a pH between 8.5 and 9.5 further prevents the formation of H_2S gas by reducing the amount of available H_3O^+. The excess hydroxide ions also help to precipitate the ferrous ions from the effluent stream in the form of ferrous hydroxide ($Fe(OH)_2$), thus reducing the Fe^{2+} concentration in the effluent stream.

Although all metal sulfides have very low solubilities the solubility of mercury sulfide is lower than other metal sulfides and mercury will be the first metal to precipitate out in a waste stream containing a mix of dissolved metals [10]. This process can achieve effluent mercury concentrations in the range of $10–20 \mu g l^{-1}$ [11], well below the targeted RCRA limit of $200 \mu g l^{-1}$ [12].

Polyelectrolyte conditioners that flocculate the fine mercury sulfide particles form large, rapid settling particles that facilitate the separation process in mixer/clarifier reactor vessel. Polymer addition is necessary to achieve minimal mercury in the effluent.

The sludge volume is reduced by mechanical dewatering equipment. Recessed plate filter presses are used to increase solids in the sludge to 25–30% [10].

The effluent water stream from the clarifier contains suspended solids not removed by the clarification process. A sand filter will reduce the concentration of suspended solids in the effluent to $10–20 mg l^{-1}$. Wastewater throughput is generally ~ $10 gpm ft^{-2}$. Backwashing is necessary to prevent clogging of the filter media, and is usually carried out daily with a flow rate of $10–40 gpm ft^{-2}$ for 3–10 min [13].

3.2. Waste volume estimations

Based on literature values of the Sulfex insoluble mercury precipitation process [10], the total volume of sludge produced by streams 1 and 2 from Fig. 2 is:

$$V_{\text{Sludge out}} = 1.97 \times 10^{-2} (\text{m}^3 \text{g}^{-1}) \times C_{\text{in}} \times V_{\text{in}} \tag{9}$$

where $V_{\text{Sludge out}}$ = sludge volume (m^3), C_{in} = influent concentration (g m^{-3}), V_{in} = influent waste volume (m^3).

This volume is based on using three times the stoichiometric FeS requirement in the reactor vessel to improve the overall conversion rate and efficiency of the process [10]. An unknown amount of additional waste from PPE will also be generated by this process (stream 3, Fig. 2). The total PPE produced is estimated to be 2% of the total sludge volume produced, or

$$V_{\text{PPE}} = 0.02 \times V_{\text{Sludge out}} \tag{10}$$

making the total volume of waste generated from the process

$$V_{\text{Total}} = V_{\text{PPE}} + V_{\text{Sludge out}} = 2.01 \times 10^{-2} \times C_{\text{in}} \times V_{\text{in}} \tag{11}$$

It is important to note that these calculations are based on a relatively clean influent stream. Like most metals, mercury may form complexes with ethylenediaminetetraacetic acid (EDTA), cyanide, ammonia and other compounds [13]. These complexations can greatly interfere with the precipitation process. These calculations are based on the assumption that the waste stream has been pretreated by alkaline chlorination, air stripping, or other appropriate pre-treatment processes to remove the complexing agents from the waste stream. This process is usually used in tandem with a hydroxide precipitation process as a finishing measure [9,10,12]. The sulfide precipitation process removes any metals with solubilities below ferrous sulfide [9], and therefore the waste volume predictions could be low if there are other dissolved metals in the waste stream. Ideally sulfide precipitation would immediately follow the metal hydroxide precipitation to minimize the possibility of additional metal removal. Other metals should not interfere with the mercury removal, but could cause additional precipitants to form, increasing the output sludge volume. These calculations assume that the sludge produced by this process does not contain the sulfide precipitants of other metals.

4. Capping

Capping is a containment technology designed to minimize contact between infiltrating water and contaminants, thereby reducing migration of contaminants from the site. The reduction in water infiltration is accomplished by covering the site with low permeability material(s). Caps may be single layered or multi-layered. Multi-layered caps are the most common and are required by RCRA land disposal regulations of 40 CFR [14]. The advantages of capping over removing and treating contaminated soil are (1) lower cost, and (2) little (if any) disturbance of contaminants. The fact that contamination is left in place means that there will be a minimal amount of waste generated as a result of capping [15].

4.1. Process description

RCRA caps have three basic components:
- a sloped, upper vegetated soil or gravel layer to minimize erosion and promote drainage off the cap;
- a drainage layer with a minimum hydraulic conductivity of $1 \times 10^{-2} \, \text{cm s}^{-1}$ to minimize water infiltration into the underlying low permeability layer;
- a low permeability layer located completely below the frost zone consisting of a synthetic liner and a layer of compacted soil with a maximum in-place saturated hydraulic conductivity of $1 \times 10^{-7} \, \text{cm s}^{-1}$ to reduce the infiltration of water into the underlying contamination.

Capping consists of four phases: a mobilization phase, an operational phase, a closure phase and a post-closure phase [16–18]. The mobilization phase consists of bringing

equipment onto the site and preparing the site for construction of the cap. Preparation typically includes physically solidifying sludges with rock, compacting or adding soil to form a base capable of supporting the cap, removing free liquids present at the site, or even partially excavating the site. The operational phase consists of the actual construction of the cap. The cap is constructed using typical construction equipment such as backhoes, graders, loaders and compactors. The operation of heavy equipment, the preparation of the site, and the construction of the cap result in resuspension of potentially contaminated particulates (soil) which may accumulate on the equipment and the worker's PPE. The contaminated PPE is collected and contributes to the waste that is generated as a result of capping. Equipment is typically decontaminated by steam cleaning. High pressure water sprays could also be used, but steam cleaning is the preferred method for decontaminating equipment since it requires less water. The wash water is contained and pumped into barrels or a tank, then transported to a facility for temporary storage or treatment [17,18].

The closure phase consists of the installation of monitoring wells and the final decontamination and demobilization of equipment. Monitoring wells are required at capped sites to detect contaminant migration from the site. Contaminated soil from excavation must be packaged and contributes to the waste generated as a result of capping. Drilling equipment is decontaminated after each boring to prevent spreading of contamination. The water collected from cleaning the equipment contributes to the waste that is generated. However, at sites on the Oak Ridge Reservation (ORR), monitoring wells are already in place, and there is usually no need to drill new wells [18,19].

The post-closure phase consists of monitoring and long-term maintenance of the cap. Maintenance consists of mowing, checking for erosion and settling, repairing cracks, etc., and results in no waste generation. The capping flow diagram is shown in Fig. 3.

4.2. Waste volume estimations

The wastes generated as a result of capping include PPE worn by the workers during capping and wash water used to decontaminate heavy equipment. To estimate the volume of waste generated as a result of capping it is necessary to know the area of the
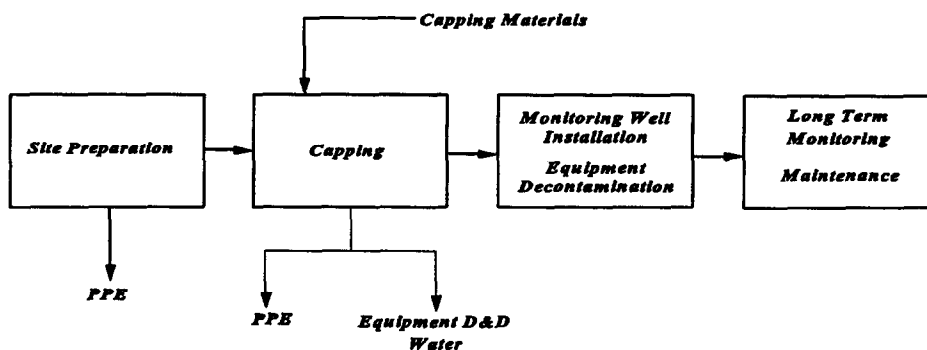

Fig. 3. Capping flow diagram.

site that will be capped. Capping is different from the other technologies in this respect. Treatment technologies "treat" volumes of waste; capping consists of covering the area over the waste. A RCRA cap is constructed at an average rate of $0.3 \, m^2 \, h^{-1}$ [15]. This rate is based on the combined activities of the preparation phase, the operational phase, and decontaminating equipment in the closure phase. The amount of PPE generated as a result of capping, if any, is minimal [16,20]. During the capping of a 50 acre site at Y-12 plant only two 55 gallon drums with PPE were collected for disposal [20]. To estimate the volume of PPE that is generated, we assume that every 5 days, all of the five workers fill a 55 gallon drum with PPE, and that each worker works 8 h per day. The volume of PPE generated is:

$$V_{PPE} = \frac{55 \, gal_{PPE}}{5 \, d} \times \frac{1 \, d}{40 \, h} \times \frac{1 \, h}{0.3 \, m^2_{Cap}} = \frac{4.6 \, gal_{PPE}}{m^2_{Cap}} = \frac{0.02 m^3_{PPE}}{m^2_{Cap}} \qquad (12)$$

The volume of water used to decontaminate equipment depends upon the frequency of the decontamination, the number of items to be decontaminated, and the type of contamination being removed. Equipment must be cleaned at least twice; at the beginning of remediation and at the end of remediation. The number of intermediate cleanings is site specific. On small projects, the equipment may only be cleaned twice, while monthly cleanings are common for large sites [17]. Monthly decontaminations are assumed for these calculations. The compliment of equipment assumed for calculations consists of one backhoe/loader, one bulldozer, and one compactor. Approximately 75 gallons of water per piece of equipment is required for steam cleaning. Based on these assumptions, the total amount of waste water generated per square meter of area being capped is:

$$V_{H_2O} = \frac{3 \times 75 \, gal_{H_2O}}{1 \, month} \times \frac{1 \, month}{732 \, h} \times \frac{1 \, h}{0.3 \, m^2_{Cap}} = \frac{0.0039 \, m^3_{H_2O}}{m^2_{Cap}} \qquad (13)$$

The total waste generated as a result of capping is the sum of the PPE waste volume and the decontamination wash water:

$$V_{Tot} = \frac{0.02 \, m^3_{PPE}}{m^2_{Cap}} + \frac{0.0034 \, m^3_{H_2O}}{m^2_{Cap}} = \frac{0.0234 \, m^3_{Waste}}{m^2_{Cap}} \qquad (14)$$

For the above calculations, it is assumed that none of the pretreatment activities (i.e. sludges may need to be physically solidified with rock, uneven ground surfaces may need to be graded) results in the generation of waste (other than PPE). In addition, it is assumed that capping does not take place in an enclosure and that any resuspended contamination is not collected or treated.

5. Discussion

From Eq. (5), Eq. (11) and Eq. (14), the volume of waste generated during cleanup could be quite large. The amount of waste generated during cleanup will greatly influence the cost of remediation.

Accurate and consistent estimates benefit WM by:

- improving the quality of waste forecasting data provided to WM by providing educated and consistent estimates of the volumes of wastes generated during remediation of waste sites;
- allowing project managers to compare a variety of feasible technologies by assessing the type and volume of secondary waste generated by each;
- allowing project managers to examine technology efficiencies and restrictions to determine whether cleanup goals for an individual site can be achieved with currently available technologies; and
- improving the waste management planning process by providing the type of secondary waste generated so that like waste streams can be consolidated for TSD of ER generated waste.

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