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Evaluation of Technologies for Enhancing Grout for Immobilizing Hanford Supplemental Low-Activity Waste (SLAW)

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EXECUTIVE SUMMARY

An expert panel consisting of the document authors was assembled by Washington River Protection Solutions, LLC (WRPS) and tasked with (1) identifying technical and regulatory limitations to treating and disposing of grouted low-activity waste (LAW), and (2) with identifying and evaluating a wide range of technical solutions to these limitations. The analysis included technologies to allow inventory management and/or improve retention of iodine, technetium, and nitrate either in the waste packages, or within the Integrated Disposal Facility (IDF), and, technologies to ensure regulated organics are below Resource Conservation and Recovery Act of 1976 (RCRA) Land Disposal Restriction (LDR) levels.

The expert panel completed their analysis in three distinct phases. In Phase I, the team reviewed the chemical and physical characteristics of the most recent projected supplemental LAW (SLAW) feed vector along with disposal site waste acceptance criteria and the fiscal year 2019 results from Pacific Northwest National Laboratory that identified the level of improvement that would be needed to meet onsite disposal requirements in the IDF (Asmussen et al. 2019a). The two regulatory constraints targeted for technology solutions by the expert panel were (1) current estimates based on performance assessment fate-and-transport modeling indicate that if the entire quantity of LAW currently designated for supplemental treatment were solidified as a cementitious material, nitrate, 99-Tc, and 129-I could exceed groundwater standards after DOE's time of compliance (1,000 years) but within 10,000 years after IDF closure and (2) LAW may contain RCRA LDR organics at levels that require removal to meet LDR concentration-based limits. It should also be noted that the first issue is only applicable to on-site disposal, not offsite at WCS and is based on calculations that assume either a low-performing grout is used, or high-performance grout does not maintain its performance (SRNL-RP-2018-00687). Asmussen et al. (2019a) estimated that the inventory of grouted SLAW could meet IDF groundwater requirements if the release rate of nitrate, 99-Tc, and 129-I were reduced by a factor of approximately 3.2, 10, and 32, respectively versus baseline formulation performance.

In Phase II, an extensive list of technologies and engineering approaches was identified and subjected to an initial assessment by the group. Current knowledge associated with technology maturation needs and constraints to implementation at Hanford were recorded in 13 distinct categories. In the final phase of the process (Phase III), the expert panel selected 7 of the 13 categories for qualitative rating and a series of meetings were held between the expert panel to discuss and rate each technology in each category. All ratings were developed by consensus. After the qualitative rating was complete, a numerical scoring criterion for each category was applied and a composite score for the individual technologies was calculated. An uncertainty score was also developed for each technology by counting the number of technology maturation unknowns identified during group discussions. Finally, the expert panel reviewed the list of scored technologies and selected a final list of 51 items that were deemed as the most viable for full-scale application at Hanford and deserving of a more detailed written description. These descriptions are provided in Appendix A. It should be noted that a technology warranting a write-up was deemed as feasible or worthy of investigation to improve a cementitious SLAW venture. The following paragraphs provide a high-level summary of the findings for each target waste component.

Two general approaches were identified by the expert panel that could meet regulatory concertation limits for RCRA LDR organics. The first approach, termed Sample and Send, relies on direct sampling of the retrieved, or in-tank waste, and only sending material to supplemental treatment if it meets the LDR concentration limits in the waste form. The second approach identified for mitigating LDR organics is the

use of a pretreatment process. Three chemical processes and one electrical organic destruction process were identified and evaluated. Two physical removal processes (evaporation and granular activated carbon adsorption) were also evaluated. Of these options, the use of evaporation to volatilize and separate organics from the LAW was identified as the most promising.

Solutions for reducing nitrate, iodine, and technetium release rates to meet groundwater standards was also assessed in this effort. Pretreatment technologies were found for iodine and technetium, but no viable pretreatment options were identified for nitrate. Direct sampling of the retrieved, or in tank waste, and sending only selected material to supplemental treatment (i.e., Sample and Send) was the highest rated option for both technetium and iodine.

Four methods were identified to modify the waste formulation to enhance the retention of iodine and five similar methods were identified for technetium, respectively. For iodine, the use of a strong base anion resin as an adsorption additive and the addition of soluble species to facilitate iodine precipitation were the highest scored options, although both have significant uncertainties. The two highest scoring technical approaches for technetium retention in the waste form are both associated with enhancing the reducing capacity of the waste form. It was also noted that the success of technetium retention methods may be hampered by our lack of understanding of waste form oxidation rate, physical stability of the waste form and the fraction of tank waste technetium that is present in the reduced Tc(I) form (although this has been estimated previously, Serne et al. 2014).

For nitrate only two techniques, the addition of carbon-based getter and the use of layered double hydroxides, were identified as being potential methods to enhance nitrate retention in the waste form. Based on the numeric values of the score and uncertainty, both methods were assessed as having similar technology maturation needs and associated uncertainty.

Contaminant-specific engineered barrier technologies were assessed for iodine, technetium, and nitrate, respectively. The contaminant specific methods are of three general types; those that modify individual waste packages, those that modify the backfill, and those that create a reactive barrier in the vadose or saturated zone below the disposal site. The technical methods that were scored highest by the expert panel were the use of an iron-amended backfill to provide a reduction/sorption zone for nitrate and technetium and injection of dissolved or colloidal AgCl to form a permeable reaction zone for conversion of soluble iodine to insoluble AgI.

Methods to modify bulk transport properties in the waste form or the disposal facility were also assessed by the expert panel. Although there has been extensive characterization work on Cast Stone since its development, work on the development of formulation modifications and alternative formulations has been very limited. There is therefore a substantial opportunity for optimization of this formulation to improve its performance and also for the development of entirely different formulations, such as geopolymers.

Of the four technologies that addressed the disposal facility, two are associated with the benefits obtained by increasing the physical size of the waste package and two are aligned with reducing water infiltration. The use of a high-density polyethylene cap was rated highest by the expert panel followed by improvements provided by increasing the size of the waste packages.

This report is expected to provide guidance for planning technology maturation efforts to ensure that an inventory of SLAW cementitious waste forms, if pursued, would meet, or exceed regulatory requirements for disposal onsite in the IDF or at an offsite facility.

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CFR	Code of Federal Regulations
COC	constituent of concern
DFLAW	direct-feed low-activity waste
DOE	U.S. Department of Energy
DSSF	double-shell slurry feed
DST	double-shell tank
Ecology	Washington State Department of Ecology
EHW	extremely hazardous waste
EPA	U.S. Environmental Protection Agency
FWF	Federal Waste Disposal Facility
FY	fiscal year
GAC	granular activated carbon
GAO	Government Accounting Office
HDPE	high-density polyethylene
HLVIT	high-level vitrification
IDF	Integrated Disposal Facility
LAW	low-activity waste
LDR	Land Disposal Restriction
LLW	low-level waste
MLLW	mixed low-level waste
NEPA	National Environmental Policy Act of 1969
ORP	Office of River Protection
PA	performance assessment
PCB	polychlorinated biphenyls
PNNL	Pacific Northwest National Laboratory
PT	Pretreatment facility
RBDA	risk-based disposal approval
RCRA	Resource Conservation and Recovery Act of 1976
REDOX	reduction and oxidation
RPP	River Protection Project
SLAW	supplemental low-activity waste
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
SST	single-shell tank
TCLP	toxicity characteristic leaching procedure
TOC	total organic carbon
TOPSim	Hanford Tank Waste Operations Simulator
TFPT	tank farm pretreatment system
TPA	Tri-Party Agreement
TSCA	Toxic Substance Control Act
TSCR	tank-side cesium removal
UHC	underlying hazardous constituent
WAC	Washington Administrative Code
WCS	Waste Control Specialists
WRPS	Washington River Protection Solutions, LLC
WTP	Hanford Waste Treatment and Immobilization Plant

1.0 Introduction

1.1 Objective

The objective of this report is to serve as a reference document for planning technology maturation activities for cementitious solidification of low-activity waste (LAW). Cementitious waste forms have been considered as a technology for supplemental LAW (SLAW) immobilization at Hanford (RPP-17675; SRNL-RP-2018-00687; DOE/EIS-0391).

To this end, an expert panel was assembled by Washington River Protection Solutions, LLC (WRPS) and tasked with: (1) identifying technical and regulatory limitations to treating and disposing of grouted LAW, and (2) identifying and evaluating a wide range of technical solutions to said limitations. The analysis included technologies to allow inventory management and/or improve retention of iodine, technetium, and nitrate either in the waste packages or within the Integrated Disposal Facility (IDF) and technologies to ensure regulated organics are below Land Disposal Restriction (LDR) levels. Approaches identified to improve retention and long-term performance of a cementitious LAW form included:

- Inventory management strategies,
- Pretreatment processes to remove key constituents,
- Advanced grout formulations,
- Getters/additives to the grout, and
- Engineered barriers.

This report is expected to provide guidance for planning technology maturation efforts to ensure that an inventory of SLAW cementitious waste forms, if pursued, would meet, or exceed regulatory requirements for disposal onsite in the IDF or at an offsite facility.

1.2 Background on Hanford Tank Waste

Approximately 56 Mgal of radioactive and hazardous waste are stored in 177 underground storage tanks at the U.S. Department of Energy (DOE) Hanford Site in southeastern Washington State. The Hanford Waste Treatment and Immobilization Plant (WTP) is being constructed to treat and vitrify the waste in glass waste forms. The WTP includes a pretreatment facility to separate the wastes into a small volume of high-level waste containing most of the radioactivity and a larger volume of LAW containing most of the nonradioactive chemicals. The high-level waste will be vitrified for ultimate disposal at an offsite federal repository.

The baseline immobilization pathway for LAW is conversion to glass in the LAW vitrification facility and the resulting glass waste form will then be disposed of onsite at the IDF. For high level waste treatment being the driver for mission duration, the LAW vitrification facility is predicted to not have the capacity to immobilize all of the LAW. This scenario leaves a portion of the LAW inventory requiring a supplemental immobilization technology (ORP-11242). SLAW treatment technologies such as a second LAW vitrification facility or a facility to make a cementitious waste form have been considered as candidates to provide additional LAW immobilization capacity (SRNL-RP-2018-00687).

1.3 Background Cementitious Waste Forms

Cementitious waste forms are produced by ambient temperature processes, and contrary to vitrification, do not result in secondary waste streams other than failed processing equipment, maintenance items, and consumables. Cementitious waste forms are widely used for low-level and hazardous aqueous waste stabilization and solidification (GAO-17-306). In setting Resource Conservation and Recovery Act of 1976 (RCRA) LDR treatment standards, cementation is identified by the U.S. Environmental Protection Agency (EPA) as the best demonstrated available technology for many of the "D" code metals and for some "F" code industrial aqueous wastes that contain metals that precipitate as low solubility phases in high pH environments. Cementation is typically not identified as best demonstrated available technology for organic constituents. Because of these benefits, and because of the cost savings associated with low-temperature treatment processes, cementitious immobilization is planned for liquid and solid secondary wastes at Hanford and are a candidate for supplemental immobilization of LAW (GAO-17-306; Cantrell et al. 2016).

The matrix phases in cementitious waste forms based on Portland cement, slag cement (ground granulated blast furnace slag), and supplemental cementitious materials are poorly crystalline low solubility hydrate phases (gels) and crystalline hydrated solids. The individual phases form by reaction between the water in the waste and the unhydrated cementitious materials. The resulting phases, which can depend on the composition of the waste, have a range of solubilities in water with portlandite and gypsum being the most soluble in cement-only waste forms to 11 Å tobermorite being the least soluble for waste forms based on slag and caustic solutions. These intergrown particles have a range of morphologies and particles sizes from submicrons to microns. The phase assemblages depend on the bulk oxide composition of the reactive cement compounds (Langton and Missimer 2014).

Waste loading values for hydrated waste forms can be higher than those of anhydrous, refractory waste forms because water, the carrier for the contaminants, is stabilized in the cementitious material whereas it is not included in the vitrified or other anhydrous waste forms.

The mechanisms for contaminant stabilization in cementitious waste forms are different than for a glass or crystalline ceramic waste form. In a glass waste form, contaminants either become part of, or are, encapsulated in the glass lattice structure. In cementitious materials, some contaminants can occupy lattice positions in the poorly ordered lattice structure of hydrated phases (gel) or crystalline hydrated matrix phases. Other contaminants may be physically or chemically sorbed on the surfaces of the matrix particles, or precipitated as low solubility species (e.g., Cr(III) and Tc(IV) in slag cement waste forms). Many anions will primarily remain in the pore solution in the waste form (e.g., NO_3^-, NO_2^-, I^-, $Tc(VII)O_4^-$) (Cantrell et al. 2016). Some anions such as Cl^-, have a measurable binding capacity in the matrix and SO_4^{2-} and CO_3^{2-} can react with cations to form solid hydrated phases. If reactant cations are not present, then small amounts of SO_4^{2-} and CO_3^{2-} may remain in the pore solution.

Although most of the phases in cement waste forms have low solubilities in water, the matrix structure has a high porosity and the micron and submicron hydrated matrix phases result in a waste form with a large internal surface area available for reaction with infiltrating environmental water, gas, and dissolved ions. The Savannah River Site (SRS) Saltstone has a transmissive porosity of between 40 and 60% (Harbour et al. 2007) depending on the test method, as compared to an in-tact glass waste form that has a few percent porosity that is not transmissive.

The porosity characteristics and the fact that the matrix is already hydrated and susceptible to cation leaching if exposed to water is a consideration with respect to stabilization of very long-lived contaminants whether or not they are incorporated in the lattice structures of hydrated phases. The late stage phase assemblage for Portland cement, slag, and supplemental cementitious materials including waste forms is silica gel and alumina gel which form as the matrix cations, chemical contaminants, and radionuclides are leached from the waste form. The time-frame over which this occurs depends on several factors such as the amount of water that contacts the waste form, exposed surface area, advection, carbonation and others.

1.4 Brief History of Hanford LAW Grout Development

The initial immobilization plan for liquid wastes at Hanford was grouting and subsequent disposal in large vaults located in the Hanford 200 East Area. In 1987 the Grout Treatment Facility was constructed at Hanford and, beginning in 1988, more than 1.4 Mgal of phosphate sulfate waste from decontamination and basin management programs at the Hanford N-reactor were immobilized in a grout vault (Huang et al. 1994). The Grout Treatment Facility was planned to immobilize the phosphate sulfate waste, cladding removal wastes and double-shell tank supernatant (termed double-shell slurry feed [DSSF]) (Serne et al. 1992). For each of these wastes a different formulation was selected (Table 1-1). The phosphate sulfate waste and cladding removal waste formulations were developed at Oak Ridge National Laboratory and were based on that laboratory's hydrofracture grouting program (Sams et al. 1988). The DSSF formulation more closely replicated the Saltstone formulation being developed in parallel at SRS. A 1.5 million gallon demonstration vault was poured using phosphate-sulfate wastes from N-reactor decommissioning activities (Martin and Lokken, 1992). Several vaults to receive the first DSSF grout were constructed but not filled following the revision to the Tri-Party Agreement (TPA) defining glass as the baseline immobilization technology for LAW in 1993. Little grout work at Hanford occurred between 1995 and 2001.

Table 1-1: Selected formulations for use during the Hanford Grout Vault program. Adapted from Table 1 of Serne et al. (1992).

Waste Type	Component	Weight Percent
Phosphate Sulfate (840 g dry mix to 1 L liquid)	Portland type I/II cement	41
	Class F fly ash	40
	Attapulgite clay	11
	Illitic clay	8
Cladding Removal (840 g dry mix to 1 L liquid)	Portland type I/II cement	42
	Class F fly ash	40
	Calcium hydroxide	10
	Illitic clay	8
Double-Shell Slurry Feed (1080 g dry mix to 1 L liquid)	Portland type I/II cement	6
	Class F fly ash	47
	Ground blast furnace slag	47

In 2002, a cleanup mission acceleration plan was proposed and included evaluations of supplemental technologies to support vitrification at the WTP (Brouns et al. 2003). A containerized grout, termed containerized Cast Stone, was evaluated as part of this effort to provide data for a SLAW risk assessment

(Mann 2003). The current baseline formulation for Cast Stone was defined at this time through work by CH2M Hill (Lockrem 2005). The Cast Stone formulation was chosen from a set of four formulations (Table 1-2), which themselves were based on grout vault era programs. More detail on the Cast Stone formulation is given in Section 1.5.

Table 1-2: Formulation considered in the selection of a baseline formulation for SLAW. Adapted from Table 6-1 of Lockrem (2005).

Component (wt%)	DRF1	DRF2 "Cast Stone"	DRF3	DRF4
Portland cement, type I/II	44.90	8.16	41.84	20
Class F fly ash	42.86	44.90	39.78	66
Blast furnace slag, grade 120	0	46.94	0	0
Attapulgite clay	5.10	0	11.22	14
Indian red pottery clay	7.14	0	7.14	0

Following the SLAW risk assessment, little to no development of grout for LAW was performed at Hanford. Instead research was focused on providing qualification data for liquid secondary waste grout where Cast Stone was again identified as the baseline formulation (Pierce et al. 2004; Westsik 2009; Pierce et al. 2010).

Beginning in 2012, WRPS has funded the SLAW development efforts with Pacific Northwest National Laboratory (PNNL) and Savannah River National Laboratory (SRNL) to obtain additional information on cementitious waste forms for Hanford LAW (Westsik et al. 2013a). The Cast Stone formulation served as the baseline for this work. The largest portion of this effort was summarized in fiscal year 2013 (FY13) when PNNL and SRNL teamed to evaluate the performance of the Cast Stone formulation for properties including waste composition, waste concentration as defined by sodium molarity, fly ash, and blast furnace slag sources, water-to-dry-mix ratios and scale up (Westsik et al. 2013b). Other efforts included testing of formulation enhancements, characterizing the Cast Stone product from the engineering-scale demonstration, preparing and characterizing Cast Stone with actual radioactive waste using an adjusted Tank 50 sample from SRNL, and obtaining performance data on a cementitious waste form to support the IDF performance assessment (PA) (Asmussen et al. 2016; Serne et al. 2016; Crawford et al. 2017). These efforts were then followed by an evaluation of historical SLAW data against a performance metric for the IDF (Asmussen et al. 2019a, summarized in Section 3.3 of this report). Concurrent with the SLAW development efforts, a large effort to demonstrate the performance of solidified liquid secondary waste (Um et al. 2016; Saslow et al. 2017a; Saslow et al. 2017b) and solid secondary waste (Nichols et al. 2017; Seitz 2017; Nichols et al. 2018; Asmussen et al. 2019b) were performed. Cementitious solidification of SLAW was also part of the assessment recently completed by a Federally Funded Research and Development Center (FFRDC) team in response to the congressional requirement placed in Section 3134 of the National Defense Authorization Act for Fiscal Year 2017 (SRNL-RP-2018-00687).

2.0 Supplemental Low-Activity Waste Feed Vector

2.1 Sources of LAW in the Feed Vector

The SLAW feed vector reported in the River Protection Project Integrated Flowsheet (RPP-RPT-57991) was used as the basis for this technology assessment. RPP-RPT-57991 reports the result of Hanford Tank Waste Operations Simulator (TOPSim) modeling for the full River Protection Project (RPP) mission and is consistent with the assumptions detailed in RPP System Plan 8 (ORP-11242) and the Integrated Waste Feed Delivery Plan (RPP-40149). Section J.2.3.3 of SRNL-RP-2018-00687 provides a summary of the assumptions in the TOPSim model and their potential impacts to the predicted feed vector for SLAW.

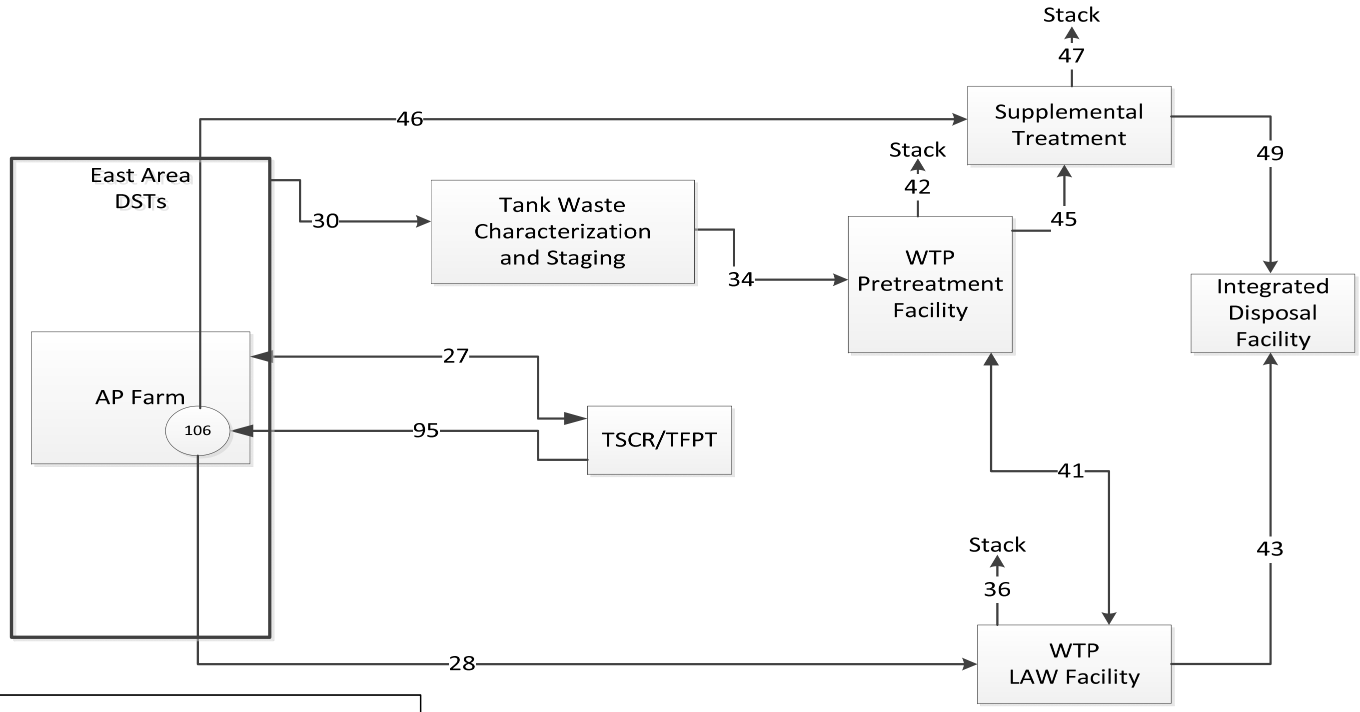
Operations are assessed in RPP-RPT-57991 in three phases and stream properties; including the concentrations of key constituent and mass flow rates passing between, and out of, facilities; are provided in the report for each of three operating phases. The three operating phases are:

- Phase 1 – Near-term and early direct-feed LAW (DFLAW) operations. This phase runs from the start of operations to the end of the 5-year tank-side cesium removal (TSCR) demonstration phase.
- Phase 2 – Remainder of DFLAW operations, which runs from the end of TSCR demonstration phase to the start of WTP Pretreatment facility (PT) operations.
- Phase 3 – Balance of mission, which runs from the startup of WTP PT to the end of the mission.

Only the feed vector for Phase 3 was considered in this analysis since this is the sole phase in which SLAW treatment is operating (RPP-RPT-57991).

Figure 2-1 provides a simplified process flow diagram for processing of SLAW in Phase 3. Solid, slurry, liquid, and gaseous waste streams passing between facilities, and out of facilities to the environment, are depicted in the figure. Stream numbers correspond to those provided in Figure 3-4 of RPP-RPT-57991.

Retrieved tank waste is assumed to be stored in 200 East Area DSTs before routing to either the WTP PT (Stream 34) via the Tank Waste Characterization and Staging facility (Stream 30), or to the TSCR / tank farm pretreatment system (TFPT) (Stream 27). Treated TSCR/TFPT waste is returned to tank AP-106. The LAW fraction treated by either the WTP PT or TSCR/TFPT system can be processed in either the LAW facility or the SLAW facility. Particular waste stream characteristics are not considered in RPP-RPT-57991 when deciding where to send a feed stream since that document assumes that the SLAW facility is also a vitrification system.



Acronyms:
DST – Double Shelled Tanks
LAW – Low Activity Waste
TSCR – Tank Site Cesium Removal System
TFPT – Tank Farm Pretreatment System
WTP – Waste Treatment Plant

Notes:
- Stream numbers correspond to nomenclature of Figure 3.1 in RPP-RPT-57991, Rev. 3.
- Feed to Supplemental Treatment System occurs from both the Pretreatment Facility and TSCR/TFPT for balance of mission

Figure 2-1: Simplified process flow diagram for Phase 3 SLAW treatment.
The figure was adapted from Figure 3-4 in RPP-RPT-57991 and stream numbers correspond to those in the original document.

TOPSim modeling feed vector data was provided as estimated monthly volume and composition data for the 359 month period of SLAW operation (December 2034 to October 2064). A total volume of 20.6 Mgal of feed from tank AP-106 (Stream 46) is predicted to occur during 89 months of the SLAW operating period. The average sodium content of these feed streams is 5.6±0.2 M. The first feed from tank AP-106 occurs in December 2034 and the last in March 2059. Feed from the WTP PT (Stream 45) occurs in 340 months of the operating period and represents a total of 30.0 Mgal. The average sodium content of these feed streams is 6.7±0.5 M. The first feed from the WTP PT is in December 2034 and the last in October 2064. It should be noted these values are slightly lower than that used in the analysis reported in SRNL-RP-2018-00687, which was based on Revision 2 of RPP-RPT-57991. The total volume of SLAW feed reported in SRNL-RP-2018-00687 is 54 Mgal.

As stated above, no particular waste stream characteristics are considered in RPP-RPT-57991 when deciding to send a feed stream to the LAW or SLAW facilities. Rather, the SLAW feed vector is a randomly selected subset of the material flow in the ongoing waste retrieval process that involves simultaneous retrieval of multiple tanks and blending of the retrieved material. The mixed nature of the feed is demonstrated in Figure 2-2, which is a graphical overview of the 10 source tanks in each month that provide the most material to the feed vector. Columns in the figure represent individual tanks (organized by area and farm) and the rows are the 359 months where SLAW is produced. Within each row there are 10 red cells (squares) that indicate the location of the 10 tanks in that month providing most of the monthly feed. It should be noted that for any month there is a minimum of 13 tanks contributing to the blended feed vector sent to the WTP PT (the average is more than 40 tanks). Also, the feed to the SLAW facility is a further blending of this material with recycle streams from the WTP. Hence, it is not possible to relate feed vector concentrations to the concentrations of retrieved waste from individual tanks.

2.2 Feed Vector Properties

Figure 2-3 depicts the predicted monthly volume of LAW provided to the SLAW facility. The data in this figure represent the combined feed stream for the two sources. There are four months during the 359-month period where there is no SLAW feed (April, May, June, and July of 2061). Table 2-1 provides the flow rate and physical properties of the combined feed vector for the remaining 355 months. As is evident from both Figure 2-3 and Table 2-1, there is a substantial variation in the volume of material provided to the SLAW facility each month as well as in the concentration of suspended solids. The suspended solids are entirely provided by the WTP PT feed stream and are comprised of materials containing aluminum, sodium, fluorine, phosphate, sulfate, and oxalate. There are no radiological constituents predicted to be associated with the suspended solids.

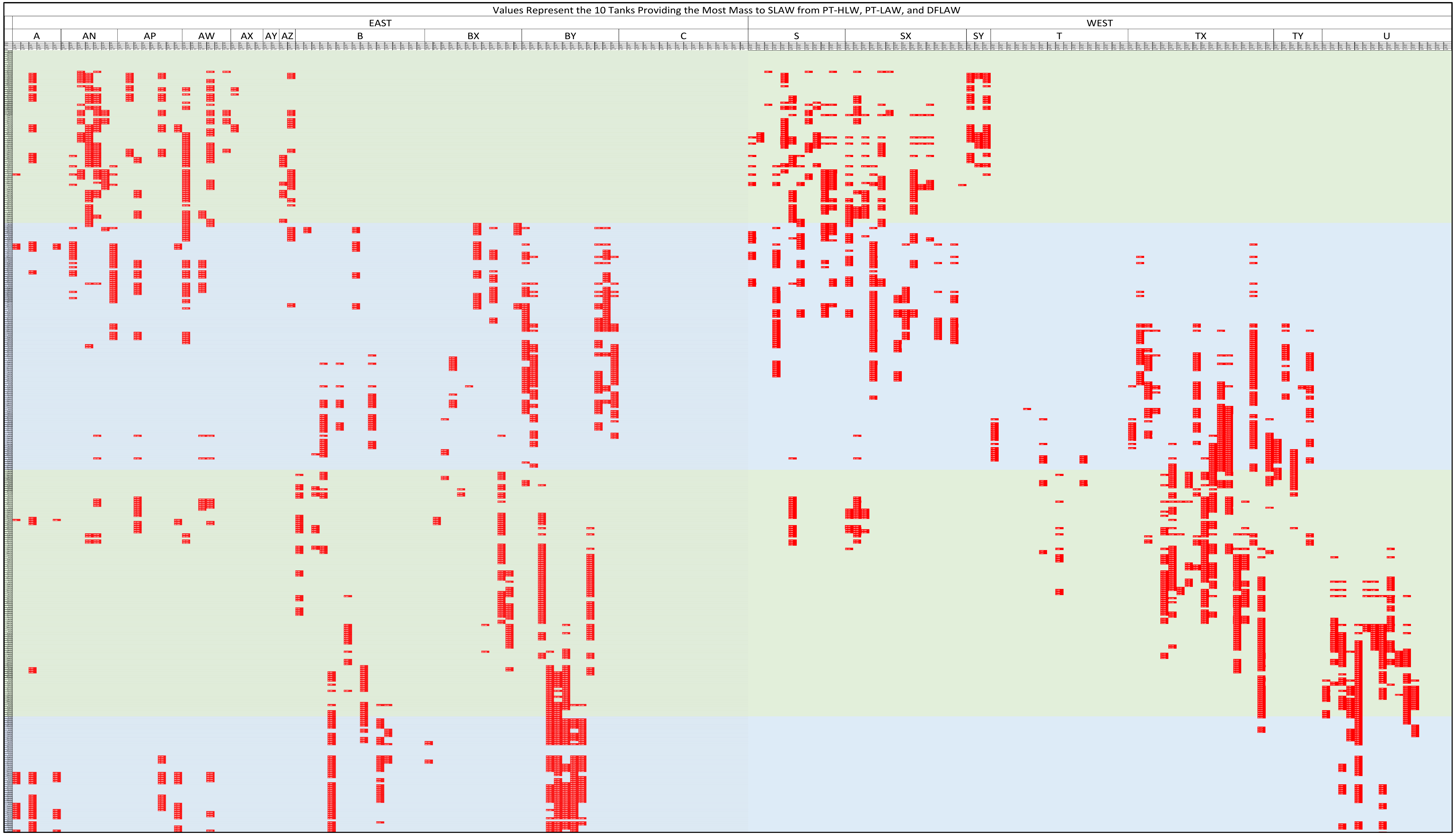


Figure 2-2: Visual representation of sources of feed to the WTP PT or TSCR/TFPT during the period of SLAW operation.
Each row represents one month and columns indicate individual tanks, organized by farm and area. The top two rows indicate tank locations (area and farm). Red entries show when individual tanks contribute to a monthly feed.

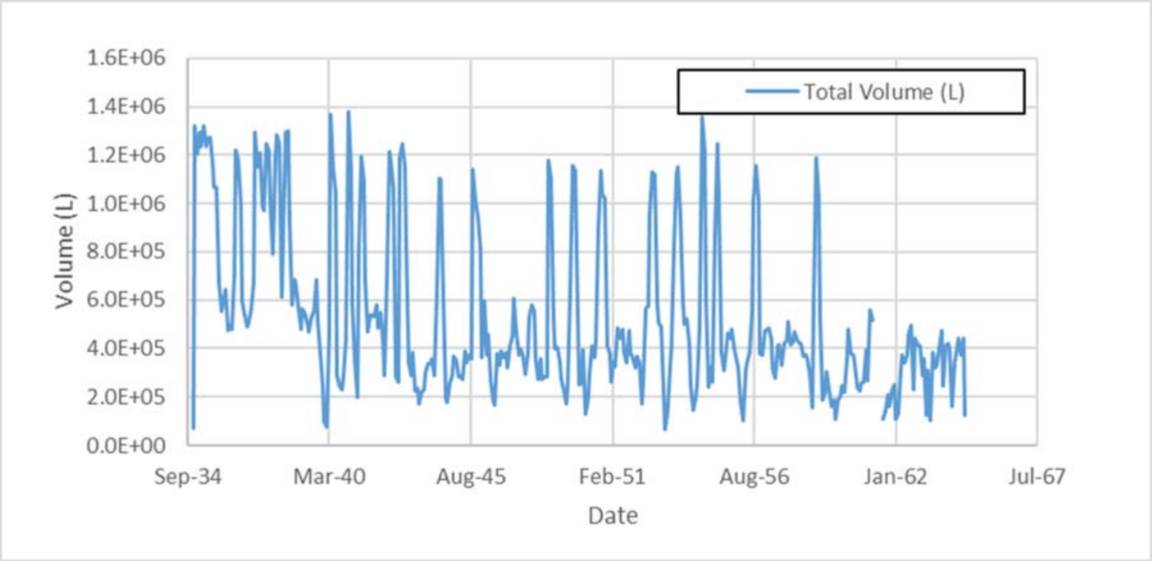


Figure 2-3: Monthly SLAW feed volume.

Table 2-1: Feed vector properties for the combined SLAW feed streams.

Property	Maximum	Average ± Stdev	Minimum
Monthly Volume (m^3)	1,379	533 ± 338	65
Density (g/ml)	1.43	1.34 ± 0.04	1.23
Suspended Solids Concentration (g/l) *	162	48.3 ± 33.6	0.066
Sodium Concentration (M)	7.8	6.6 ± 0.6	5.2

* Data for the 340 months with predicted solids in the feed. No solids are predicted in 15 months of feed.

2.2.1 Non-Radiological Constituents

Table 2-2 lists the most abundant non-radiological dissolved constituents in the combine feed stream for SLAW processing. The items listed in the table comprise 99.2% of the total mass of dissolved non-radiological species processed by the SLAW facility. The first six items in Table 2-2 comprise the majority of the mass of all the monthly SLAW feed vectors (Figure 2-4). Figure 2-4 indicates the frequency (as a percentage of the 355 months) that a non-radiological element or compound is among the top six mass contributors to a monthly feed vector. Excluding the first six items in Table 2-2, the other species that significantly contribute to dissolved mass are Cl^- (2.5%, or 9 monthly feed vectors), total Cr (1.1%, or 4 monthly feed vectors), F^- (0.3%, or 1 monthly feed vector), K^+ (1.1%, or 4 monthly feed vectors), and total organic carbon (TOC) (5.4%, or 19 monthly feed vectors). Plots of the concentration of the Table 2-2 species by month, as well as other non-radiological species in the SLAW feed vector, are provided in Appendix B.

Table 2-2: Compounds and elements comprising 99.2% of the mass of dissolved non-radiological continuants processed in SLAW.

Component	Campaign Total (Kg)	Maximum Conc'n (g/L)	Minimum Conc'n (g/L)	Average Conc'n (g/L)	Standard Deviation (g/L)	Fraction of Total Mass (%)
Na^+	2.73E+07	1.79E+02	1.20E+02	1.51E+02	1.37E+01	43.1
NO_3^-	2.19E+07	1.89E+02	3.78E+01	1.01E+02	3.15E+01	34.5
NO_2^-	5.78E+06	5.55E+01	8.74E+00	2.59E+01	9.45E+00	9.1
CO_3^{-2}	3.82E+06	3.74E+01	6.40E+00	1.71E+01	6.52E+00	6.0
Total Al	1.65E+06	2.66E+01	1.02E+00	1.14E+01	5.32E+00	2.6
SO_4^{-2}	1.12E+06	1.77E+01	2.38E+00	5.08E+00	2.35E+00	1.8
Cl^-	3.28E+05	3.71E+00	6.22E-01	1.56E+00	5.78E-01	0.5
PO_4^{-3}	2.83E+05	2.89E+00	8.01E-01	1.47E+00	2.73E-01	0.4
TOC	3.25E+05	1.91E+01	1.90E-01	1.10E+00	2.15E+00	0.5
F^-	2.10E+05	2.50E+00	1.71E-01	1.09E+00	3.81E-01	0.3
K^+	2.35E+05	6.06E+00	2.62E-01	1.09E+00	8.35E-01	0.4
Total Cr	1.74E+05	3.75E+00	1.83E-01	1.06E+00	5.84E-01	0.3

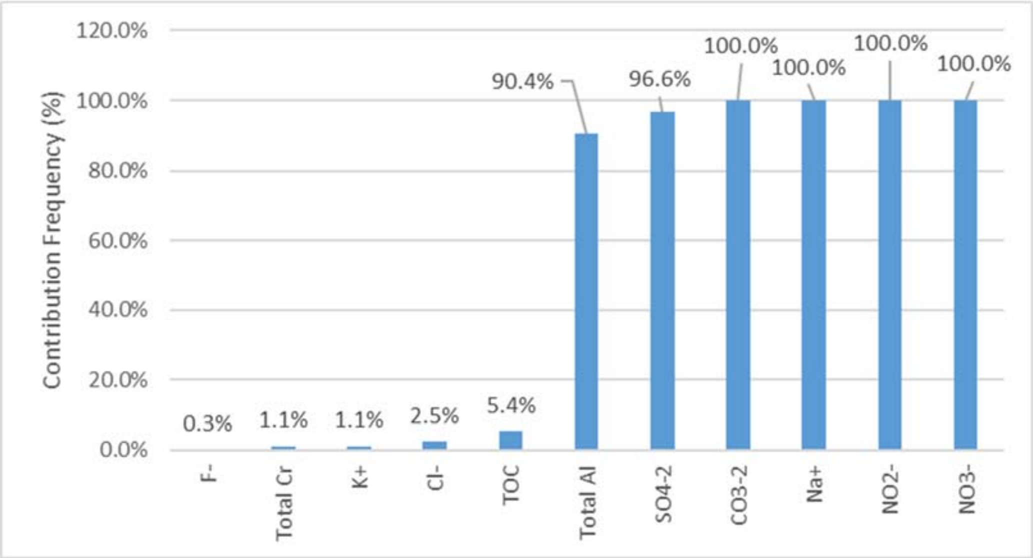


Figure 2-4: Frequency that an individual species is among the top six contributors to the dissolved mass of a monthly feed vector.

2.2.2 Radiological Constituents

Figure 2-5 indicates the relationship between time and the quantity of radiological materials processed in the SLAW facility, expressed as a percent of the total quantity processed over the full SLAW mission (6.04×10^5 Ci). Approximately 78% of the curie content of the SLAW facility feed is processed in the first 10 years of operation.

Table 2-3 lists the most abundant radiological constituents in the combine feed stream. The items listed in the table comprise 99.9% of the total curies processed by the SLAW facility. The isotopes 90-Sr, 90-Y, 99-Tc, 151-Sm, and 63-Ni are among the top six contributors to the curie content of more than 86% of the monthly feed vectors (Figure 2-6). It should be noted that the crystalline silicotitanate (CST) in the

TSCR system has shown evidence of high removal of 90-Sr along with the targeted Cs (Rovira et al. 2018). The TOPSim model does not account for 90-Sr removal by CST. Plots of the monthly concentrations of each of the Table 2-3 isotopes are provided in Appendix C.

Table 2-3: Radiological compounds contributing 99.9% of the Curies processed in SLAW.

Component	Campaign Total (Ci)	Maximum Conc'n (Ci/L)	Minimum Conc'n (Ci/L)	Average Conc'n (Ci/L)	Standard Deviation (Ci/L)	Fraction of Total Ci (%)
90-Sr	2.90E+05	8.78E-03	5.94E-05	1.31E-03	1.46E-03	47.1%
90-Y	2.54E+05	7.27E-03	5.23E-05	1.09E-03	1.32E-03	41.2%
151-Sm	4.62E+04	2.07E-03	4.60E-07	1.71E-04	3.03E-04	7.51%
99-Tc	1.02E+04	2.73E-04	1.78E-05	4.89E-05	2.63E-05	1.65%
63-Ni	7.73E+03	3.93E-04	2.35E-06	3.02E-05	3.95E-05	1.26%
137-Cs	1.86E+03	8.92E-05	9.34E-07	6.69E-06	1.01E-05	0.302%
137m-Ba	1.76E+03	8.42E-05	8.81E-07	6.31E-06	9.53E-06	0.285%
241-Am	1.44E+03	1.04E-04	4.28E-07	6.01E-06	1.26E-05	0.234%
239-Pu	4.58E+02	9.26E-06	4.90E-07	2.46E-06	1.50E-06	0.0744%
93-Zr	4.92E+02	2.83E-05	1.81E-07	1.79E-06	2.90E-06	0.0800%
93m-Nb	4.75E+02	2.61E-05	1.86E-07	1.74E-06	2.77E-06	0.0771%
14-C	3.52E+02	4.88E-06	3.99E-07	1.58E-06	7.61E-07	0.0572%

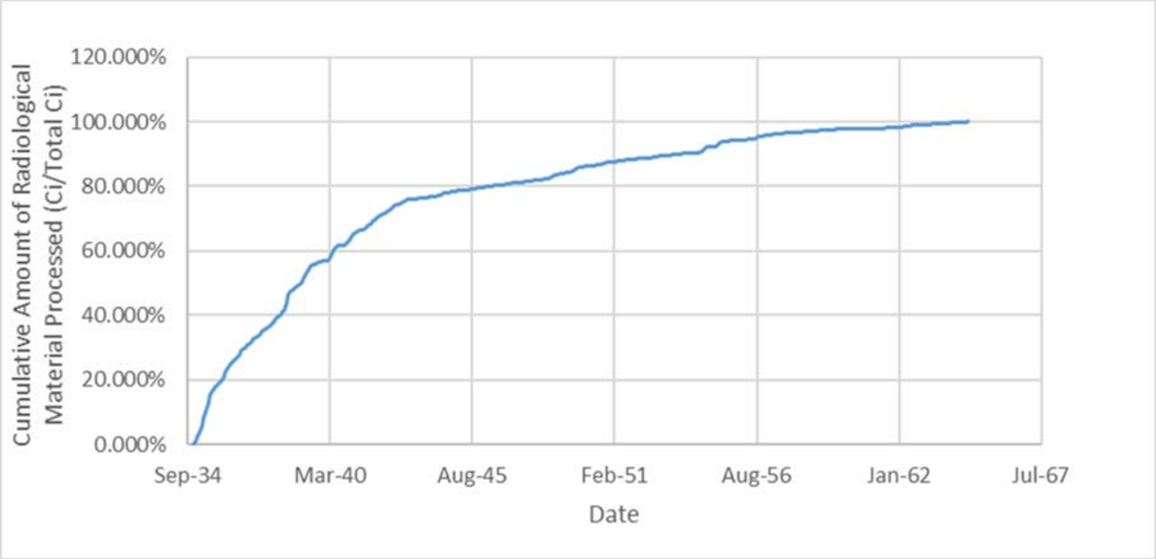


Figure 2-5: Cumulative amount radioactive material processed in the SLAW facility, expressed as a percent of the total amount processed over the life of the facility.

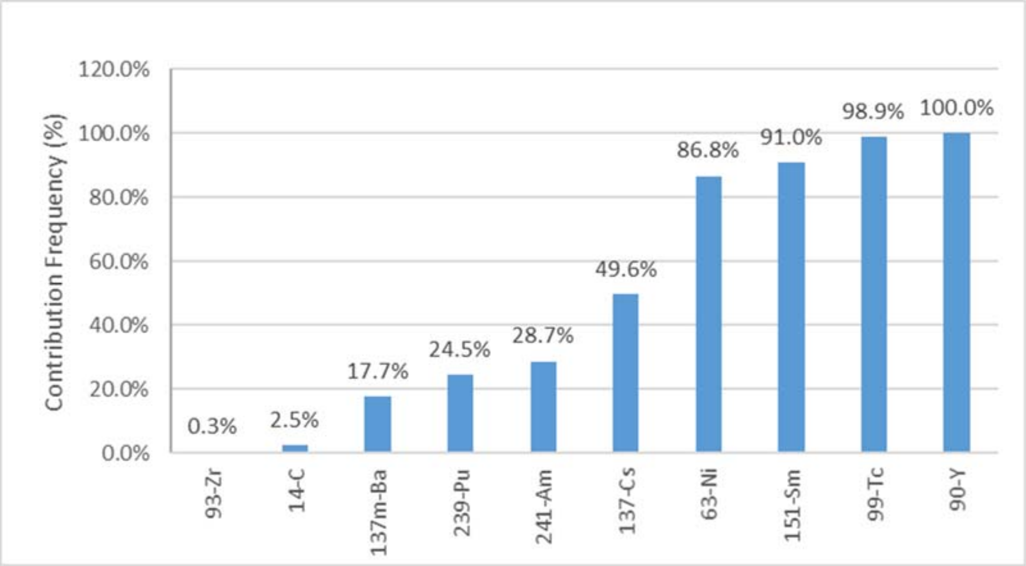


Figure 2-6: Frequency that an individual isotope is among the top six contributors to the curie content of a monthly feed vector.

Out of the major contributing radionuclides, 99-Tc and 129-I are two key constituents of concern for wastes disposed in the IDF (Asmussen et al. 2019a). While not present as a top six contributor in any of the feed vector analyses, 129-I is a concern due to its long half-life and high environmental mobility. Figures 2-7 and 2-8 are a depiction of the concentrations of these two isotopes in the monthly feed vectors. The total quantity of 129-I and 99-Tc contained in the cumulative SLAW feed vector is 10.2 Ci and 1.0×10^4 Ci, respectively. It is interesting to note that there are many monthly feed vectors where both the 129-I and 99-Tc are at relatively low levels. This fact can be seen in Figure 2-9 where the monthly radiological content of these two isotopes are plotted against each other. This correlation might be exploited if it is necessary to minimize both the 99-Tc and 129-I inventory in a LAW stream that is directed to cementation since it is possible to identify feeds low in both isotopes. For example, there are 152 monthly feeds with 99-Tc and 129-I simultaneously at or below their median values of 4.3×10^{-5} Ci/L and 4.2×10^{-8} Ci/L, respectively (Figure 2-10). Combined, these 151 monthly feed vectors represent 31% of the total volume of SLAW to be processed, but only 19% of both the 129-I and 99-Tc. It is recognized, however, that other factors will also impact the LAW feed that must be processed in the SLAW facility, including production requirements and the available lag-storage and vitrification capacity. Hence, additional process flowsheet analysis is necessary to estimate the potential for selective processing to impact the 129-I and 99-Tc inventory contained in SLAW.

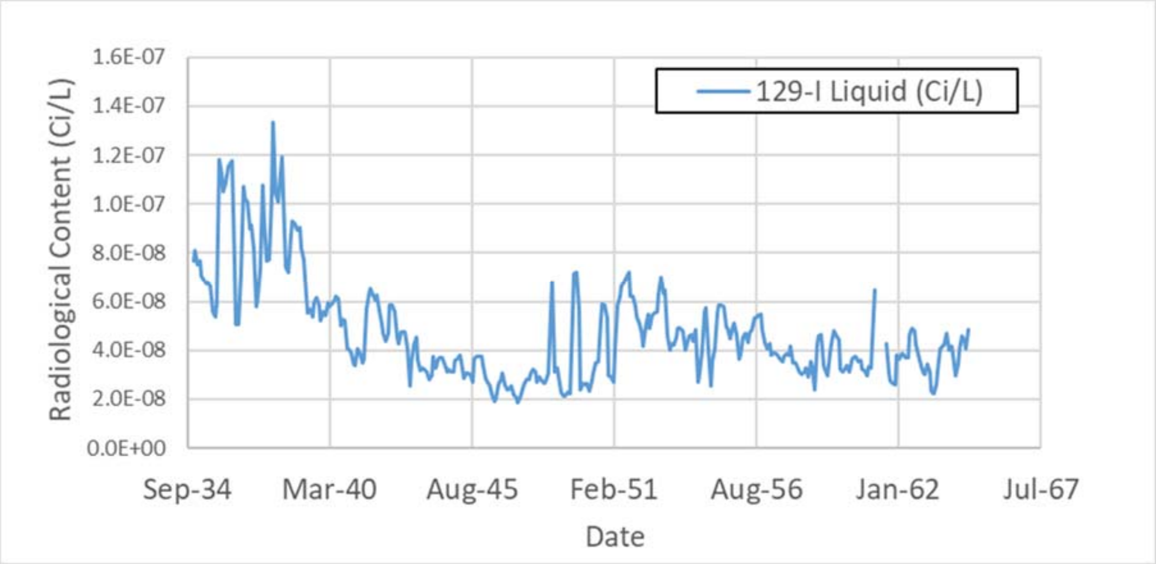


Figure 2-7: Iodine-129 concentration in SLAW monthly feeds.

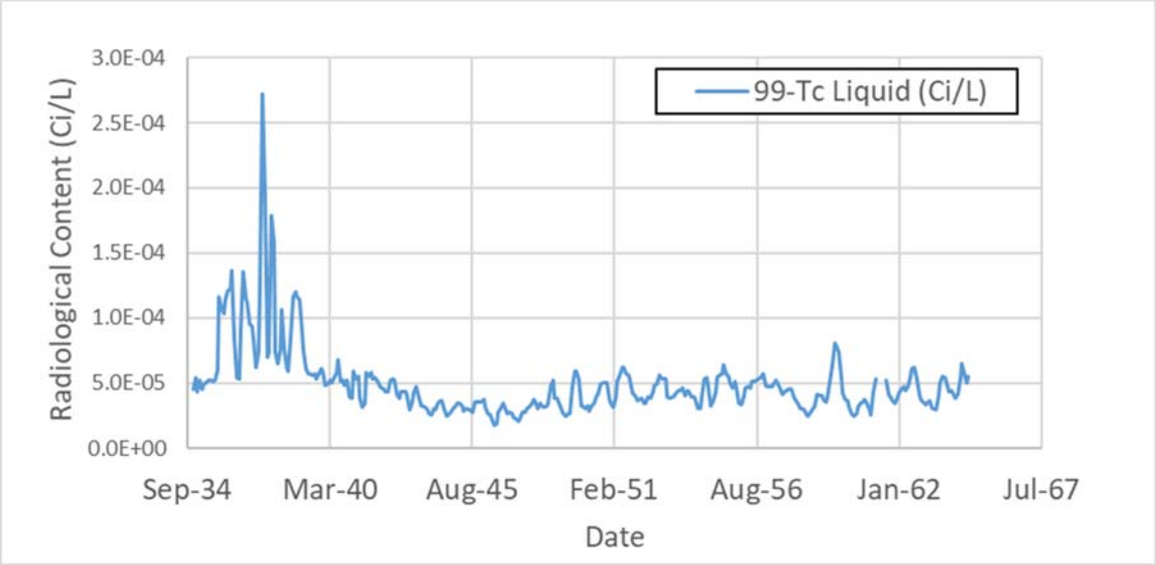


Figure 2-8: Technetium-99 concentration in SLAW monthly feeds.

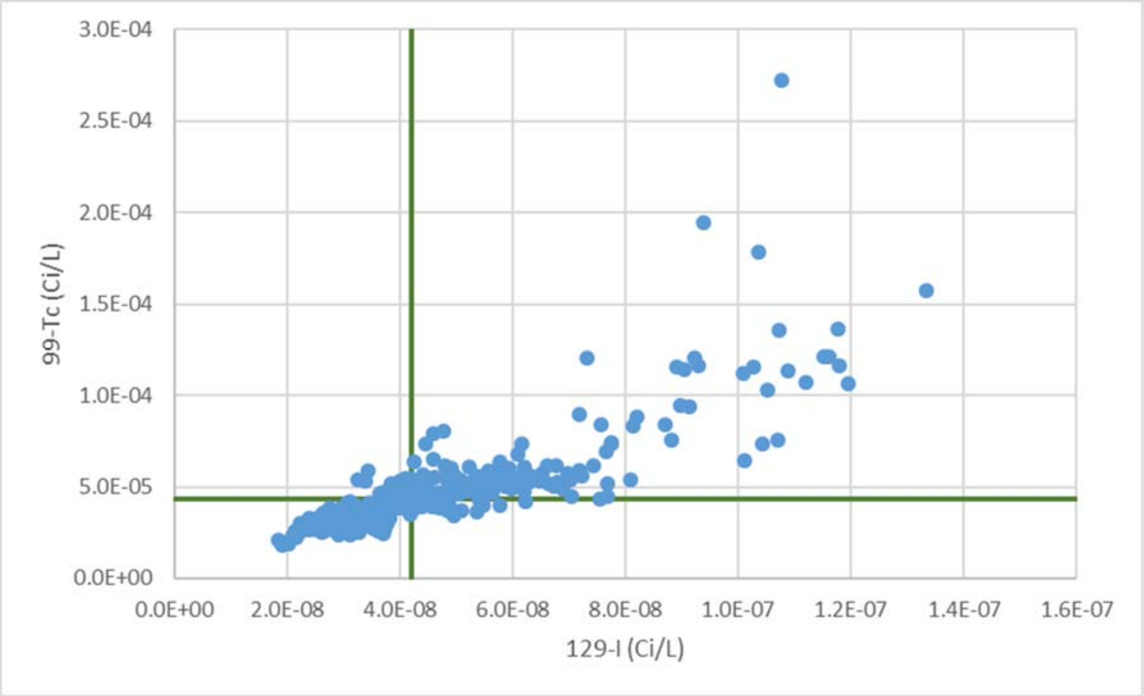


Figure 2-9: Relationship between the monthly concentrations of 129-I and 99-Tc in the SLAW feed. Green lines represent the median values for 99-Tc (4.3E-5 Ci/L) and 129-I (4.2E-8 Ci/L).

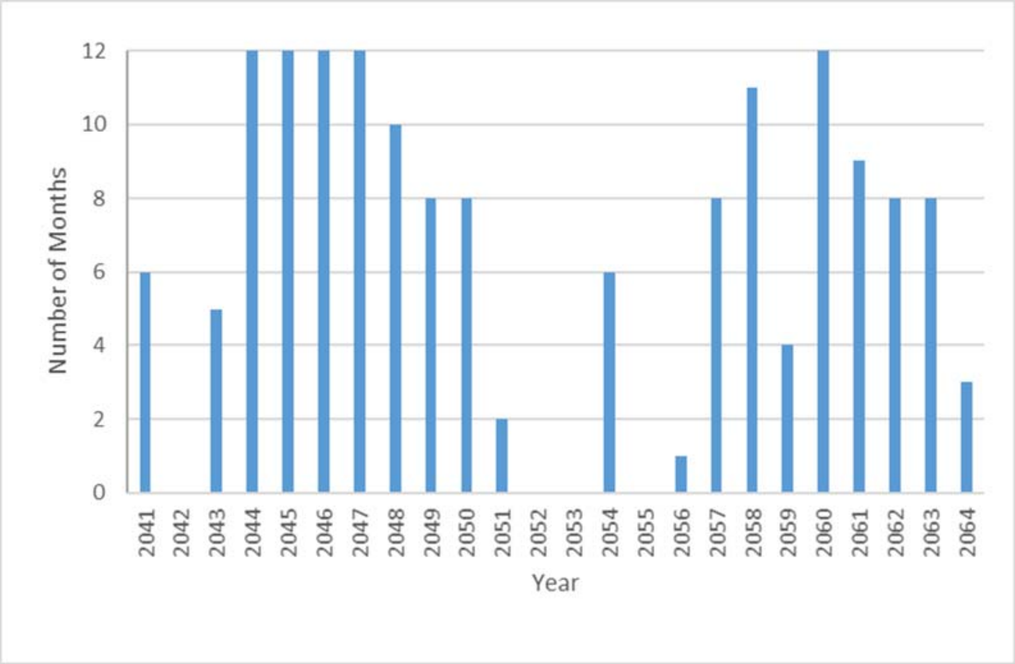


Figure 2-10: Months in each year where the SLAW feed is predicted to contain 5.0×10^{-5} Ci/L or less of 99-Tc and 4.0×10^{-8} Ci/L or less of 129-I.

2.2.3 Organic Constituents

Organic compounds are tracked in the TOPSim feed vector as TOC rather than individual compounds. Figure 2-11 depicts the TOC concentration in the monthly feed vectors on a logarithmic scale. There is a wide variation in TOC concentrations, ranging from a maximum of 19 g/L to a minimum of 0.19 g/L and the data are highly early mission skewed with 78% of the months having TOC values ≤1.0 g/L and 4% of the months having values ≥5 g/L. The skewed nature of the values is also demonstrated by difference between the average (±stdev) and median values, which are 1.1±2.2 g/L and 0.59 g/L, respectively.

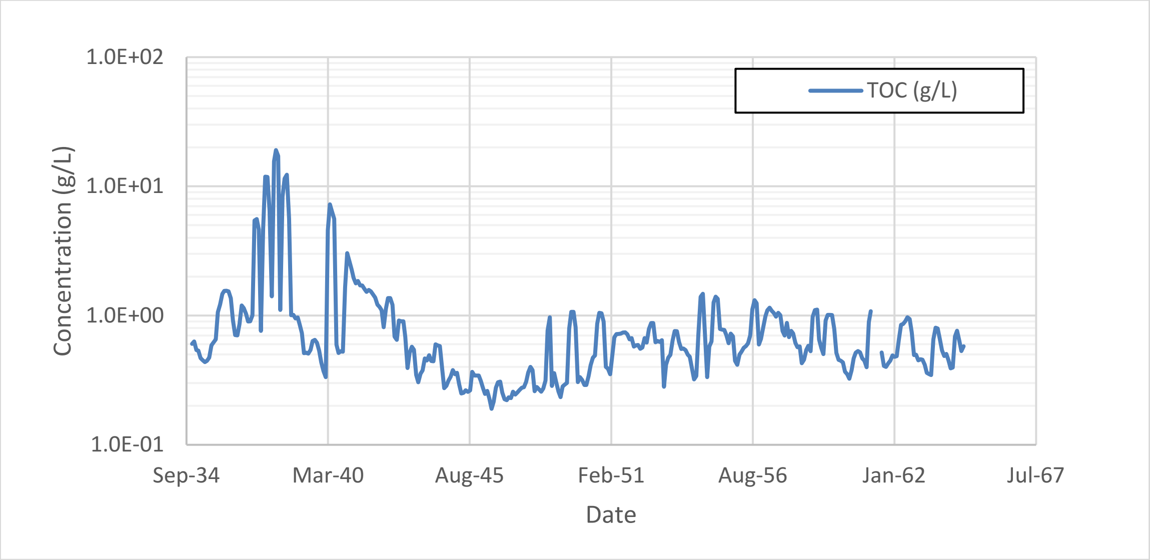


Figure 2-11: TOC concentration in the monthly feed vectors to the SLAW facility.

One note of interest with the feed vectors is that many of the months with low concentrations of both 99-Tc and 129-I (Figure 2-9) also have low levels of TOC. This phenomenon is illustrated in Figure 2-12, which is a reproduction of Figure 2-9 with the 100 months with the lowest TOC concentrations indicated with a "+" symbol. The times when these 100 months occur throughout the SLAW campaign is indicated in Figure 2-13. The 100 monthly data sets represent 18.4% of the total SLAW feed volume.

This correlation is of note since, as discussed in Section 3, RCRA LDR organics, along with 99-Tc and 129-I, could prohibit disposal of cementitious LAW at the IDF. Hence, feed vectors that are low in all these constituents might be the preferential choice for processing in a SLAW cementation facility if the final waste form will be disposed onsite. It is important to note; however, that there is not a definitive relationship between TOC concentration and the concentration of regulated organics and that data on the concentrations of individual LDR organic compounds are needed.

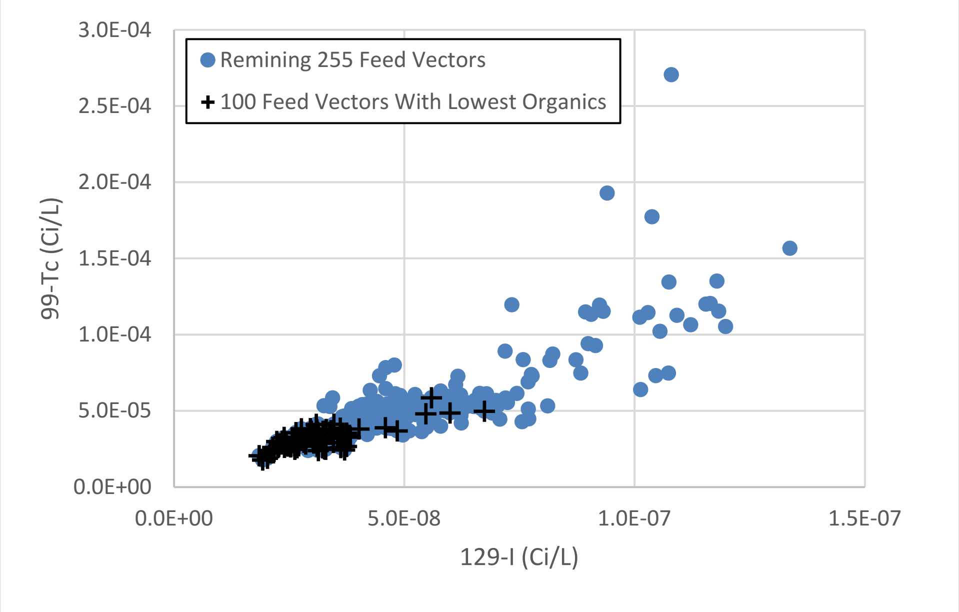


Figure 2-12: Relationship between the monthly concentrations of 129-I and 99-Tc in the SLAW feed with the feed vectors with the lowest 100 TOC concentrations shown with "+" symbols.

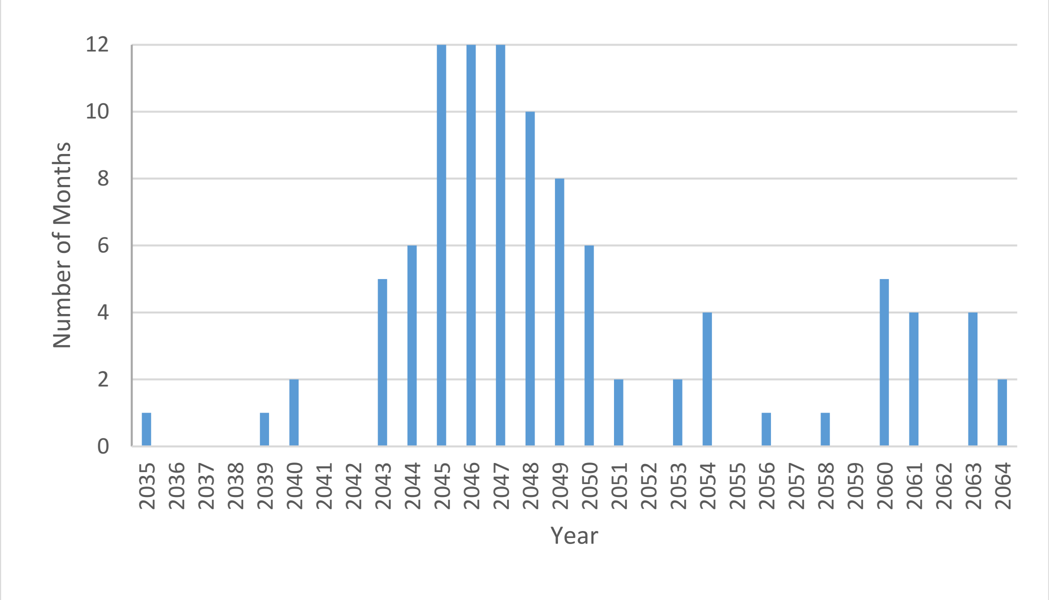


Figure 2-13: Months in each year when the 100 lowest TOC concentrations occur.

3.0 Regulatory Considerations for Onsite Disposal

3.1 National Environmental Policy Act

National Environmental Policy Act of 1969 (NEPA) requirements are contained in the Council on Environmental Quality regulations (40 CFR 1500-1508), DOE regulations (10 CFR 1021), and DOE O 451.1B. NEPA requirements apply to whether the activity takes place on or off the Hanford Site. This report is prepared with the assumption that the DOE will ensure appropriate NEPA coverage is provided for SLAW. DOE is the sole entity responsible to decide whether or not the Final Tank Closure and Waste Management Environmental Impact Statement (DOE/EIS-0391) provides adequate coverage for SLAW activities. To that end, DOE included the following statement in the Summary to DOE/EIS-0391:

> *DOE does not have a preferred alternative regarding supplemental treatment for LAW; DOE believes it beneficial to study further the potential cost, safety, and environmental performance of supplemental treatment technologies. Nevertheless, DOE is committed to meeting its obligations under the TPA regarding supplemental LAW treatment. When DOE is ready to identify its preferred alternative regarding supplemental treatment for LAW, this action will be subject to NEPA review as appropriate. DOE will provide a notice of its preferred alternative in the Federal Register at least 30 days before issuing a ROD.*

3.2 Toxic Substance Control Act

The Toxic Substance Control Act (TSCA) regulates the management and disposal of polychlorinated biphenyls (PCBs). The "*Framework Agreement for Management of PCBs in Hanford Tank Waste*", signed August 31, 2000, by the EPA, Washington State Department of Ecology (Ecology), and DOE set forth how TSCA regulations are applied to Hanford tank waste. The agreement states a preference to use the PCB remediation waste 40 CFR 761.61 regulations for cleanup and disposal. PCB remediation waste remains TSCA regulated even when <50 ppm if the PCBs in the waste are from a regulated source (40 CFR 761.61 and 761.65). However, if <50 ppm, the waste does not require storage under 40 CFR 761.65. PCB wastes will be managed based on source concentration and date of disposal, regardless of dilution or blending from spills or remediation activities.

Analytical data for PCBs in DST waste is more prevalent than single-shell tank (SST) waste. When analytical data is not available for an unsampled layer in a tank, the Tank Farms Best-Basis inventory program uses a PCBs template (RPP-RPT-26878) to assign a PCB concentration to the waste. The Tank Operations Contractor Management Plan TFC-PLN-33 documents the known SSTs containing waste at >50 ppm PCBs: T-105, TY-106, and U-112 (note that a revision is planned for TCF-PNL-33 that will likely add T-102, T-103, and TY-105). When PCBs exist at >50ppm in a waste, those wastes require PCB marking and labeling. No DST waste to date has shown levels of PCBs >50 ppm. The higher feed concentration of PCBs for 242-A Evaporator campaigns has been Aroclor 1016 reported at less than 0.25 ppm. DST effluents will be tested for PCBs prior to TSCR during the DFLAW campaigns.

Hanford tank waste is presumed to be regulated as PCB remediation waste unless proven to contain concentration of PCBs below 0.5 ppb based on 40 CFR 761.79(b)(1)(iii), the decontamination standard for water containing PCBs. PCB Remediation wastes require an EPA approved risk-based disposal approval (RBDA) in order to treat and dispose of SLAW. There is no concentration standard established in TSCA or Hanford RBDAs to provide a target for treatment and disposal of SLAW. An RBDA for SLAW is expected to build on the future RBDA for DFLAW.

3.3 Resource Conservation and Recovery Act

The RCRA established requirements for EPA to develop and promulgate treatment, storage, and disposal requirements for the management of hazardous waste. EPA waste codes and LDRs apply to hazardous wastes whether on or off the Hanford Site. Washington State is an authorized state to implement EPA regulations on behalf of the EPA and has a mature dangerous waste program. Washington State implementation of the RCRA regulations are in some cases broader in scope (e.g., Washington State criteria of toxicity and persistence in WAC 173-303-100) and in other cases more restrictive (e.g., not adopting an exclusion EPA has published) than those imposed by the EPA. . These Washington State-only requirements are discussed in Section 3.4 for the option of onsite disposal.

EPA regulations implementing RCRA are found in 40 CFR 260-280, with the regulations applicable to this document existing in 40 CFR 261 and 40 CFR 268. The Washington State counterpart for identification and listing of hazardous waste is found in WAC 173-303-070 through -100, which in general, operates in lieu of 40 CFR 261. WAC 173-303-140 is the LDR counterpart and 40 CFR 268 is incorporated by reference in WAC 173-303-140.

3.3.1 Identification and Listing of Hazardous Waste

The process of assigning waste codes is called “waste designation” as described in WAC 173-303-070. Since the late 1980s, DOE has identified waste codes applicable to SST and DST waste on the SST Waste System Dangerous Waste Permit Application, Part A Form (SST Part A) and the DST System Dangerous Waste Permit Application (DST Part A), respectively. These Part A forms are periodically updated as required by WAC 173-303-800 and -805 with sampling analytical data when available. Waste codes have typically been revised in both the SST Part A and the DST Part A at the same time and are identical because of a lack of a basis to differentiate between the two. The only waste code difference is that the DST Part A has F039 (landfill multisource leachate) listed where the SST Part A does not. This difference resulted from a 1995 waste planning that assumed the DST system would need to manage F039. This forecast has never been realized and so F039 waste is not discussed further in this report because it does not apply to the waste.

3.3.1.1 Waste Designation

The waste designation process has four steps (WAC 173-303-070(3)(d)(i) through (iv)), of which the first three will be discussed in this section, and the fourth one addressed in the Section 3.4. The regulations state for waste designation:

> *(d) To determine whether or not a solid waste is designated as a dangerous waste a person must:*
> *(i) First, determine if the waste is a listed discarded chemical product, WAC 173-303-081;*
> *(ii) Second, determine if the waste is a listed dangerous waste source, WAC 173-303-082;*
> *(iii) Third, determine if the waste also exhibits one or more dangerous waste characteristics, WAC 173-303-090; and*
> *(iv) Fourth, if the waste is not listed in WAC 173-303-081 or 173-303-082, and does not exhibit a characteristic in WAC 173-303-090, determine if the waste meets one or more dangerous waste criteria, WAC 173-303-100.*

Step (i) and (ii) of the waste designation process address listed waste, discarded chemical products, and dangerous waste sources, and are consistent for the most part, between EPA and Washington State. Differences between EPA and Washington State exist for some of the minor exclusions which does not affect Hanford tank waste. Waste designation of listed waste is not solely based on a concentration, but on how the chemical was used or not used.

Under waste designation step (i), no chemical use records outside of the 1995 policy discussions with Ecology regarding initial start-up of the 200 Area Effluent Treatment Facility exist. Consequently, "U" or "P" listed waste codes are not applicable to the DST Part A or SST Part A (Ecology 1995).

Step (ii) of the waste designation pertaining to dangerous waste sources addresses "K" and "F" listed waste. Listed waste codes starting with a K are industry specific and are not applicable to Hanford. Listed waste codes starting with an F are more general and can apply to historical and present Hanford activities. To support start-up of the 200 Area Effluent Treatment Facility in 1995, the listed waste codes applicable to Hanford treatment, storage, and disposal units were documented in WHC-MR-0517. The "F" coded constituents applicable to Hanford tank waste are indicated in Table 3-1.

Table 3-1: "F" coded listed constituents in SST waste.

Waste Code	Constituent	CAS Number	Basis for Listed Waste
F001	1,1,1-Trichloroethane	71-55-6	Crane degreasing in B Plant
F002	Methylene Chloride	75-9-2	Ingredient in T Plant cleaning product
F003	Acetone	67-64-1	Laboratory solvent
F003	Methyl Isobutyl Ketone (Hexone)	108-10-1	Redox solvent use
F004	Cresols and Cresylic Acid	o-cresol – 95-48-7 m-cresol – 108-39-4 p-cresol – 106-44-5 Mixed – 1319-77-3	Ingredient in T Plant cleaning product
F005	Methyl Ethyl Ketone	78-93-3	Laboratory solvent

It is important to note that subsequent to identification of listed waste identified on the DST Part A and SST Part A, EPA provided guidance that chemicals as ingredients in products are not a listed waste (EPA 1996). No effort has been initiated to remove listed waste codes because the F001, F003, and F005 would still exist.

Without the knowledge on how the chemical waste was originally used, the "F" codes do not apply. For example, finding a chemical in a waste from testing does not make the "F" code applicable. Rather, the "F" code applies if the chemical is in the waste and if it was used in a manner constituent with the listing description in WAC 173-303-9904 and associated guidance on use of solvents.

At this time, the F001-F005 tank farms constituents apply to Hanford tank waste. If information is compiled to show through operational history and waste transfers that the specific listed waste code should not be applied, discussions with DOE may result in establishing process knowledge to limit application of the constituents and/or waste codes. At present, this process knowledge does not exist and it is also not possible to precisely reproduce the impacts of historical transfers. Hence, the F001-F005 waste codes listed in Table 3-2 will be needed to be included as part of the waste designation when managing SLAW.

The third step in the waste designation process in WAC 173-303-070(3)(d)(iii) is assigning characteristic waste codes. Table 3-2 identifies each characteristic waste code from WAC 173-303-090 and indicates if it is listed in the DST Part A and SST Part A.

Table 3-2: Characteristic waste codes from WAC 173-303-090 and relationship to DST & SST Part A.

Property or Constituent	CAS #	Waste Code	Included on Part A	Comment
Property				
Ignitability	N/A	D001	X	Supernate waste does not display characteristic. Position documented in 242-A and TSCR permit application waste analysis plans.
Corrosivity (Liquids)	N/A	D002	X	Always applies to liquids based on tank farms compatibility program maintaining pH >12.5
Corrosive solid/semisolid (State-only)	N/A	WSC2		Not applied to tank waste in storage
Reactivity	N/A	D003	X	Supernate waste does not display characteristic. Position documented in 242-A and TSCR permit application waste analysis plans.
Inorganic Constituent				
Arsenic	7440-38-2	D004	X	Waste may display characteristic
Barium	7440-39-3	D005	X	Waste may display characteristic
Cadmium	7440-43-9	D006	X	Waste may display characteristic
Chromium	7440-47-*3	D007	X	Waste may display characteristic
Lead	7439-92-1	D008	X	Waste may display characteristic
Mercury	7439-97-6	D009	X	Waste may display characteristic
Selenium	7782-49-2	D010	X	Waste may display characteristic
Silver	7440-22-4	D011	X	Waste may display characteristic
Organic Constituent				
Endrin	72-20-8	D012		Insecticide not used at Hanford
Lindane	58-89-9	D013		Insecticide not used at Hanford
Methoxychlor	72-43-5	D014		Pesticide not used at Hanford
Toxaphene	8001-35-2	D015		Insecticide not used at Hanford
2,4-D	94-75-7	D016		Herbicide not used at Hanford
2,4,5 TP Silvex	92-72-1	D017		Herbicide not used at Hanford
Benzene	71-43-2	D018	X	--
Carbon Tetrachloride	56-23-5	D019	X	--
Chlordane	57-74-9	D020		Pesticide not used at Hanford
Chlorobenzene	108-90-7	D021		--
Chloroform	67-66-3	D022	X	--
o-Cresol	95-48-7	D023		--
m-Cresol	108-39-4	D024		--
p-Cresol	106-44-5	D025		--
Cresol [total]	1319-77-3	D026		--

Property or Constituent	CAS #	Waste Code	Included on Part A	Comment
1,4-Dichlorobenzene	106-46-7	D027		--
1,2-Dichloroethane	107-06-2	D028	X	--
1,1- Dichloroethylene	75-35-4	D029	X	--
2,4-Dintrotoluene	121-14-2	D030	X	--
Heptachlor (and its epoxide)	76-44-8	D031		Insecticide not used at Hanford
Hexachlorbenzene	118-74-1	D032		--
Hexachlorobutadiene	87-68-3	D033	X	--
Hexachloroethane	67-72-1	D034	X	--
Methyl Ethyl Keton	78-93-3	D035	X	--
Nitrobenzene	98-95-3	D036	X	--
Pentachlorophenol	87-86-5	D037		--
Pyridine	110-86-1	D038	X	--
Tetrachloroethylene	127-18-4	D039	X	--
Trichloroethylene	79-01-6	D040	X	--
2,4,5-Trichlorophenol	95-95-4	D041	X	--
2,4,6-Trichlorophenol	88-06-2	D042		--
Vinyl Chloride	75-01-4	D043	X	--

The characteristic "D" codes are different than listed codes in that the applicability of each "D" code is re-evaluated at a point of generation to determine if it applies. This evaluation determines if the waste code applies to the waste because the property/constituent still meets the application criteria (e.g., the waste code D008 for lead applies at the point of generation if the concentration is still above the waste designation level in WAC 173-303-090 (8)(c) of 5 mg/L). This evaluation is independent of the LDR program requirements discussed in the next section. Only the "D" codes listed on the Part A are considered when performing a waste designation for Hanford tank waste.

3.3.2 Land Disposal Restrictions

RCRA LDR requirements attach to a waste at the point of generation according to EPA and Washington State regulations and guidance. EPA LDR requirements exist for each "F" code and "D" code, and the requirements are found in 40 CFR 268.40 and are incorporated by reference at WAC 173-303-140. Waste codes can have more than one LDR requirement, called subcategories. Within each subcategory, two LDR requirements exist for treatability groups, one for wastewater and one for non-wastewater. The definitions of wastewater and non-wastewater are found in 40 CFR 268.2:

> *(d) Non-wastewaters are wastes that do not meet the criteria for wastewaters in paragraph (f) of this section.*
> *(f) Wastewaters are wastes that contain less than 1% by weight total organic carbon (TOC) and less than 1% by weight total suspended solids (TSS).*

For constituents with concentration-based treatment standards, the concentration of the constituent has to be below the treatment standard for the waste to be eligible for land disposal. The frequency of testing of the treated waste form is determined on a case-by-case basis on the land disposal facility waste analysis plan contained in the RCRA permit, and the waste acceptance review process.

3.3.2.1 “F” Code Treatment Standards

F001-F005 waste codes in 40 CFR 268.40 have four subcategories. Based on the process knowledge used to assign the tank farms “F” codes from historical Hanford activities, only the first subcategory applies to Hanford tank waste. This subcategory includes (40 CFR 268.40):

> *F001, F002, F003, F004 and/or F005 solvent wastes that contain any combination of one or more of the following spent solvents: acetone, benzene, n-butyl alcohol, carbon disulfide, carbon tetrachloride, chlorinated fluorocarbons, chlorobenzene, o-cresol, m-cresol, p-cresol, cyclohexanone, o-dichlorobenzene, 2-ethoxyethanol, ethyl acetate, ethyl benzene, ethyl ether, isobutyl alcohol, methanol, methylene chloride, methyl ethyl ketone, methyl isobutyl ketone, nitrobenzene, 2-nitropropane, pyridine, tetrachloroethylene, toluene, 1,1,1-trichloroethane, 1,1,2-trichloroethane, 1,1,2-trichloro-1,2,2-trifluoroethane, trichloroethylene, trichlorofluoromethane, and/or xylenes [except as specifically noted in other subcategories]. See further details of these listings in § 261.31.*

The “F” code treatment standards for the tank farms F001-F005 constituents are chosen from this subcategory. The treatment standards assumed to apply to treated SLAW are identified in Table 3-5.

3.3.2.2 “D” Code Treatment Standards

The subcategories for the “D” codes is more complex than the “F” codes. Different “D” codes have different numbers of subcategories. This document assumes DOE will be able to assign the treatment standards for mixed waste to the SLAW activities. How DOE will be able to accomplish this is beyond the scope of this document. When mixed waste treatment standards are applied, the deactivation treatment standard will apply to corrosivity (D002) and the concentration-based treatment standards will apply to the heavy metals (D004-D011). The concern currently unresolved with Ecology is DOE’s ability to use ordinary mixed waste treatment standards when not applying the specified treatment technology of HLVIT to Hanford tank waste. HLVIT is defined in 40 CFR 268.42 as:

> *HLVIT: Vitrification of high level mixed radioactive wastes in units in compliance with all applicable radioactive protection requirements under control of the Nuclear Regulatory Commission.*

The LDR subcategory that calls out this treatment standard for non-wastewaters in 40 CFR 268.40 for corrosivity (D002) and heavy metals (D004-D011) states:

> *Radioactive high level wastes generated during the reprocessing of fuel rods. (Note: This subcategory consists of non-wastewaters only.)*

The treatment standards assumed to be used for SLAW are identified in Table 3-3 for the physical properties. The treatment standards for the organics and heavy metals are identified in Table 3-5.

Table 3-3: Treatment standards for DST and SST Part A waste codes based on physical properties.

Property	Waste Code	Included in DST and SST Part A	Wastewater Treatment Standard from 40 CFR 268.40	Non-Wastewater Treatment Standard from 40 CFR 268.40
Ignitability	D001	X	DEACT and meet 268.48 standards	DEACT and meet 268.48 standards
Corrosivity	D002	X	DEACT and meet 268.48 standards	DEACT and meet 268.48 standards
Reactivity	D003	X	DEACT and meet 268.48 standards	DEACT and meet 268.48 standards

The treatment standard for DEACT is also defined in 40 CFR 268.42. The definition states:

> *DEACT: Deactivation to remove the hazardous characteristics of a waste due to its ignitability, corrosivity, and/or reactivity.*

Cementitious immobilization as a treatment process for LAW will deactivate the physical property associated with the waste codes.

3.3.2.3 Underlying Hazardous Constituents and Waste Code Hierarchy

In the previous section, the treatment standard for the physical property "D" codes was identified as "DEACT and meet 268.48 standards." Since the tank waste corrosion control program maintains the pH of tank waste using a nitrate/nitrite/hydroxide ratio (usually >12.5), the D002 waste codes always applies to the waste being treated by SLAW, and therefore the "meet 268.48 standards" aspect of the D002 treatment standard triggers the evaluation of underlying hazardous constituents (UHC). According to EPA guidance, a UHC is identified if it can reasonably be expected to be present in the waste above the treatment standard level. A generator has to consider the list of constituents in 40 CFR 268.48. Table 3-5 contains a list of potentially regulated UHC for Hanford tank waste along with their treatment standards. The constituents listed in Table 3-5 correspond to the complete list of UHC listed in 40 CFR 268.48 (less Mercury – Nonwastewater from Retort) and any other compound, associated with the tank waste D Code and F Code listings, that was not contained in the UHC list. This latter requirement added Cresols (m,p,o), 2-Nitropropane, and 2-Ethoxyethanol.

Table 3-5 also provides information about waste code hierarchy. If the constituent is one of the tank farms F001-F005 constituents, then only the "F" code is identified because the constituent will be tracked under the "F" code. For other constituents, if there is an applicable "D" code on the DST Part A or SST Part A, consideration will first be given to whether the constituent will be tracked with a "D" code when the concentration is greater than the waste designation level prior to treatment. The constituent may be below the waste designation level for the "D" code, but then it may exceed the UHC treatment standard, in which the constituent will be tracked as a UHC. It is important to note that treated SLAW waste will need to be tested for the "F" code tank farms constituents, "D" code constituents, and UHCs. If the treated SLAW waste fails testing for any other these constituents, the waste will either need to be retreated, or DOE will need to request a treatment variance from Ecology in accordance with 40 CFR 268.44 to dispose of the treated SLAW waste as-is without further treatment.

It is probably an over-estimate to assume all of the constituents listed in Table 3-5 might apply to Hanford tank waste immobilized in a supplemental treatment system, however the process knowledge is not mature enough to be able to shorten the list at this time. Work is needed to (1) review historical information and refine the understanding of the past use of chemicals at Hanford, (2) determine the stability of a compound in the tank waste environment, (3) define how a chemical may degrade in the tank waste environment, and (4) assess whether any degradation products are regulated compounds. This information can be used in the future to refine the lists of chemicals of LDR constituents. Available knowledge is limited to a subset of tanks, and a subset of the layers (saltcake, sludge, and supernatant) within a tank, as well as the processing strategy for retrieving and blending tank contents. Most of the recent work in RPP-RPT-61301 has been focused on supernatant supporting the Test Bed Initiatives.

Completing a thorough analysis to reduce the list to a set of most probable compounds that would be regulated under RCRA LDR is beyond the scope of this effort. However, the last five columns in Table 3-5 have been included to provide information useful to a future down-selection process. The sixth, seventh, and eighth columns provide information on materials known to have been used at Hanford, or those which have been detected in solid, liquid, or gas tank samples. Note that the detected data is only from the indicated source and does not represent a complete analysis of Hanford tank data. The second to the last column indicates the regulated organics identified in the recent review of SLAW processes conducted by a multi-laboratory team (SRNL-RP-2018-00687). The last column represents reduced list of UHCs generated during a 2003 regulatory data quality objectives process completed in support of LDR/delisting at the WTP (RPP-RPT-54699).

3.4 Regulatory Requirements for Onsite Disposal

Hanford tank waste is a mixed waste (both radiological and hazardous). As a mixed waste SLAW is subject to both the regulatory requirements of DOE O 435.1 and supporting DOE M 435.1-1 for radiological constituents, and to the regulatory requirements of the RCRA, the Washington State specific dangerous waste regulations, and the LDRs for certain non-radiological constituents.

3.4.1 DOE O 435.1 Radiological Waste Management

DOE O 435.1 and its accompanying manual describes the requirements for providing reasonable assurance that disposal of radioactive constituents in low-level waste (LLW) and mixed low-level waste (MLLW) will provide adequate protection of human health and the environment. Assurance is provided through facility siting, design, engineered barriers, and completing a PA of the disposal system to evaluate impacts to human health and the environment. Impacts are expressed as future radiation dose to receptors via multiple exposure pathways. The PA for a given disposal facility typically entails use of sophisticated computer models that simulate the natural and engineered features of the disposal system to predict the fate and transport of contaminants over long periods of time, typically 1,000 to 10,000 years. Key features that influence disposal facility performance include; waste form durability, physical configuration, contaminant inventory, and whether or not a credible groundwater exposure pathway exists. Specific performance objectives for LLW and MLLW disposal facilities managed and regulated by DOE are found in DOE M 435.1, Chapter IV. These objectives include requiring the radiation dose to the public to not exceed 25 mrem/y as well as an assessment of future impacts to water resources.

DOE O 435.1 also addresses criteria for classifying waste radioactive LLW or MLLW. This classification applies to the radioactive content of the waste as the hazardous components of Hanford tank waste are regulated under RCRA. Most tank waste originated from the reprocessing of nuclear fuel and must be incorporated in a solid physical form at a concentration that does not exceed the applicable concentration limits for Class C LLW as set out in 10 CFR 61.55 and meet safety requirements comparable to the performance objectives set out in 10 CFR 61, Subpart C.

3.4.1.1 Implications for DOE O 435.1 for Onsite SLAW Disposal

The most recent PA for the IDF was published in 2018 and included vitrification as the supplemental treatment for LAW (RPP-RPT-59958). If treatment of the SLAW results in cementitious rather than vitrified waste form for disposal in the IDF then the PA would need to be updated to include an analysis of the impacts of the new waste form and this analysis would need to demonstrate that the exposure performance objectives are met.

The radiological waste classification criteria detailed in 10 CFR 61.55 were applied to each of the 355 monthly SLAW feed vectors to determine the waste classification if this material were solidified into a typical cementitious waste form. The final mass and volume of solidified waste for each month was determined by assuming a final waste density of 1.77 g/mL and a value of 1.8 m^3/m^3 for the ratio of the volume of the solidified waste to the volume of treated LAW (SRNL-RP-2018-00687). The result of this evaluation is presented in Table 3-4. None of the monthly feed vectors resulted in greater than Class C waste and only 20 were Class C. The remaining 335 months produced solidified LAW that was below Class C. All waste streams are acceptable for disposal in the IDF since the facility is capable of accepting Class A, B, and C wastes (IDF-00002).

Table 3-4: Estimated waste classification for the SLAW feed vector.

Waste Classification	Number of Monthly Feed Vectors Producing Waste of Given Classification
Class A Waste	1
Class B Waste	334
Class C Waste	20
Greater Than Class C Waste	0
Total →	355

3.4.2 Land Disposal

Section 3.3 describes the LDR requirements associated with the “F” codes and the “D” codes, applicable for onsite disposal. There are no special onsite considerations for the “F” codes and “D” codes in Washington State. Washington State does however impose additional requirements for land disposal in the state. This section explains those requirements and concludes there are no additional requirements imposed for onsite disposal of SLAW.

3.4.2.1 Waste Designation

Four Washington State-only waste codes are also listed on the DST Part A and SST Part A described in WAC 173-303-100. These codes are WT01 (Toxicity for Extremely Hazardous Waste), WT02 (Toxicity for Dangerous Waste), WP01 (Persistence for Extremely Hazardous Waste with halogenated organic compounds), and WP02 (Persistence for Dangerous Waste with halogenated organic compounds). The WT01 waste code was added to the initial Revision 0 of the SST Part A in February 1988; however, no reason was provided as to why the waste code was included (Letter 008344). The other three waste codes were added in Revision 2 of the SST Part A in 1993 (Letter 93-RPB-154) to be consistent with the DST Part A. When the three other waste codes were added, the example of the reason for the addition were WT02-silver, and WP01 and WP02 were included for halogenated hydrocarbons with no specific chemical example given.

When the state-only waste codes were included in the SST Part A revisions, WAC 173-303 was structured differently than it is today for waste designation. Before 1995, the WAC 173-303 waste designation requirements included a section called "special knowledge" which caused a generator to apply the state-only waste codes when federal waste codes were present. Ecology changed the waste designation policy in 1995 to "Additional designation required" in WAC 173-303-070(5) as well as including the waste designation procedure language found today in WAC 173-303-070(3)(d). The fourth step of the waste designation process found in WAC 173-303-070(3)(d) is to evaluate the waste for state-only waste codes of toxicity and persistence in WAC 173-303-100 only when listed waste codes (WAC 173-303-081 and -082) as well as characteristics waste codes (WAC 173-303-090) do not apply to the waste.

WAC 173-303-070(3)(d)(iv) states:

> *To determine whether or not a solid waste is designated as a dangerous waste a person must:…(iv) Fourth, if the waste is not listed in WAC 173-303-081 or 173-303-082, and does not exhibit a characteristic in WAC 173-303-090, determine if the waste meets one or more dangerous waste criteria, WAC 173-303-100.*

Since WAC 173-303-082 requires assignment of the Federal "F" code listed waste designations for tank waste (F001-F005) and WAC 173-303-090 requires assignment of the Federal "D" code designations (e.g., D002 corrosivity), state-only waste codes are not included as part of a proper waste designation for tank waste. The state-only waste codes remain on the Part A forms for historical purposes and do not need to be addressed when planning for treatment and disposal of waste.

3.4.2.2 State-Only Land Disposal Restrictions

Washington State has included four state-only LDR requirements in WAC 173-303-140(4). For completeness, these are discussed here but they do not impose additional requirements on the disposal of treated SLAW onsite. The four requirements are: (1) disposal of extremely hazardous waste (EHW), (2) disposal of liquid waste, (3) disposal of solid acid waste, and (4) disposal of organic/carbonaceous waste.

Disposal of EHW: Section 3.4.2.1 discussed how a waste designation for Hanford tank waste does not include the state-only EHW waste codes of WT01 or WP01 contained on the DST Part A and SST Part A. This LDR is considered waste code dependent in that the waste code needs to apply in order for the LDR to apply.

Even in the event one would assert the LDR would apply independent of the waste code of WT01 and WP01, this LDR does not impose additional requirements on DOE mixed waste based on state law. While the language in WAC 173-303-140(4)(a) appears to generally prohibit disposal of EHW in Washington State. RCW 70.105.050(2) states:

> *(2) Extremely hazardous wastes that contain radioactive components may be disposed at a radioactive waste disposal site that is (a) owned by the United States department of energy or a licensee of the nuclear regulatory commission and (b) permitted by the department and operated in compliance with the provisions of this chapter. However, prior to disposal, or as a part of disposal, all reasonable methods of treatment, detoxification, neutralization, or other waste*

> *management methodologies designed to mitigate hazards associated with these wastes shall be employed, as required by applicable federal and state laws and regulations.*

Cementitious solidification of SLAW would be considered to meet DOE obligation to use reasonable methods "…designed to mitigate hazards…" The IDF meets criteria (2)(a) and (b) from the preceding quote since it is owned by DOE and is permitted by Ecology and operated in compliance "…with this chapter."

***Disposal of liquid waste*:** This LDR is merely a repeat of EPA language in 40 CFR 264.314 regarding disposal of liquids in landfills. No additional requirements are imposed by this LDR.

Disposal of solid acid waste: Under WAC 173-303-090(6), a state-only waste code exists, WSC2 for corrosive semisolids and solids. SLAW immobilized in a cementitious waste form will not be acidic and therefore this LDR does not impose additional requirements.

***Disposal of organic/carbonaceous waste*:** This LDR imposes a disposal prohibition on organic/carbonaceous waste defined in WAC 173-303-140(3)(c) as:

> *(c) "Organic/carbonaceous waste" means a dangerous waste that contains combined concentrations of greater than ten percent organic/carbonaceous constituents in the waste; organic/carbonaceous constituents are those substances that contain carbon-hydrogen, carbon-halogen, or carbon-carbon chemical bonding.*

At first glance, this LDR could be viewed as written very broadly and may impose additional requirements. However, WAC 173-303-140(4)(d)(iii) contains the following provision:

> *This prohibition against the land disposal of organic/carbonaceous waste does not apply to any person who certifies to the department that recycling, treatment and incineration facilities are not available within a radius of one thousand miles from Washington state's borders.*

DOE filed such a certification (DOE 1998) and Ecology approved the request (Ecology 1999), rendering the prohibition of land disposal of organics/carbonaceous waste not applicable to Hanford.

Table 3-5: Compilation of chemical compounds or elements associated with applicable LDR standards for Hanford LAW based on the RCRA Part A waste codes.

Compound or Element Name	CAS Number	DST and SST Part A Associated Waste Code(s)[a]	Wastewater Standard, Concentration (mg/l)	Non-Wastewater Standard, Concentration in (mg/kg) unless noted as "mg/l TCLP"	Used At Hanford (RPP-RPT-61301)[b]	Measured in DST Headspace or Liquid (RPP-RPT-61301)[b]	Reported as Detected in Liquid or Solid RPP-21854[b]	In Table A.1 of SRNL-RP-2018-00687[b]	In Table 3[c] of RPP-RPT-54699[b]
1,1,1-Trichloroethane	71-55-6	F001	0.054	6	X	X	--	--	X
1,1-Dichloroethylene	75-35-4	D029, UHC	0.025	6	X	--	--	--	X
1,2-Dichloroethane	107-06-2	D028, UHC	0.21	6	X	--	--	--	X
2,4,5-Trichlorophenol	95-95-4	D041, UHC	0.18	7.4	--	X	--	--	X
2,4-Dinitrotoluene	121-14-2	D030, UHC	0.32	140	--	--	--	--	--
2-Butanone	78-93-3	F005	0.28	36	X	X	--	X	X
4,6-Dinitro-o-cresol	534-52-1	UHC	0.28	160	--	X	--	--	--
4-Methyl-2-pentanone	108-10-1	F003	0.14	33	X	X	--	X	X
Acetone	67-64-1	F003	0.28	160	X	X	X	X	X
All Aroclors	1336-36-3	UHC, TSCA	0.1	10	X	X	X	--	X
Benzene	71-43-2	D018, UHC	0.14	10	X	X	X	--	X
Butyl benzyl phthalate	85-68-7	UHC	0.017	28	X	X	--	--	X
Carbon tetrachloride	56-23-5	D019, UHC	0.057	6	X	X	--	--	X
Chloroform	67-66-3	D022, UHC	0.046	6	X	X	X	--	X
Cresols (m,p,o)	1319-77-3	F004	0.88	11.2	--	X	--	--	X
Cyclohexanone	108-94-1	UHC	0.36	0.75 mg/l TCLP	--	--	--	--	--
di-n-Butyl phthalate	84-74-2	UHC	0.057	28	--	X	X	--	X
Hexachlorobutadiene	87-68-3	D033, UHC	0.055	5.6	--	--	--	--	X
Hexachloroethane	67-72-1	D034, UHC	0.055	30	--	--	--	--	X
Methanol	67-56-1	UHC	5.6	0.75 mg/l TCLP	X	X	--	X	--
Methylene chloride	75-09-2	F002	0.089	30	X	X	X	X	X
n-Butyl alcohol	71-36-3	UHC	5.6	2.6	X	X	--	X	--
Nitrobenzene	98-95-3	D036, UHC	0.068	14	--	X	X	--	X
Pyridine	110-86-1	D038, UHC	0.014	16	--	X	--	X	X
Tetrachloroethylene	127-18-4	D039, UHC	0.056	6	X	X	X	--	X
Trichloroethylene	79-01-6	D040, UHC	0.054	6	X	X	X	--	X
Vinyl chloride	75-01-4	D043, UHC	0.27	6	--	--	--	--	X
1,1,2,2-Tetrachloroethane	79-34-5	UHC	0.057	6	X	X	X	--	X
1,1,2-Trichloro-1,2,2-trifluroethane	76-13-1	UHC	0.057	30	--	--	--	--	--
1,1,2-Trichloroethane	79-00-5	UHC	0.054	6	X	--	X	--	X
1,1-Dichloroethane	75-34-3	UHC	0.059	6	X	--	--	--	X
1,2,4-Trichlorobenzene	120-82-1	UHC	0.055	19	X	X	--	--	X
1,2-Dibromo-3-chloropropane	96-12-8	UHC	0.11	15	--	--	--	--	--
1,2-Dichlorobenzene	95-50-1	UHC	0.088	6	--	--	--	--	X
1,2-Dichloropropane	78-87-5	UHC	0.85	18	--	--	--	--	X
1,3-Dichlorobenzene	541-73-1	UHC	0.036	6	--	X	--	--	X
1,4-Dichlorobenzene	106-46-7	UHC	0.09	6	--	X	--	--	X
2,4,6-Trichlorophenol	88-06-2	UHC	0.035	7.4	--	X	--	--	X
2,4-Dichlorophenol	120-83-2	UHC	0.044	14	--	--	--	--	X

Compound or Element Name	CAS Number	DST and SST Part A Associated Waste Code(s)[a]	Wastewater Standard, Concentration (mg/l)	Non-Wastewater Standard, Concentration in (mg/kg) unless noted as "mg/l TCLP"	Used At Hanford (RPP-RPT-61301)[b]	Measured in DST Headspace or Liquid (RPP-RPT-61301)[b]	Reported as Detected in Liquid or Solid RPP-21854[b]	In Table A.1 of SRNL-RP-2018-00687[b]	In Table 3[c] of RPP-RPT-54699[b]
2,4-Dimethyl phenol	105-67-9	UHC	0.036	14	--	--	--	--	--
2,4-Dinitrophenol	51-28-5	UHC	0.12	160	--	X	--	--	--
2-Chloronaphthalene	91-58-7	UHC	0.055	5.6	--	--	--	--	X
2-Chlorophenol	95-57-8	UHC	0.044	5.7	--	--	--	--	X
2-Naphthylamine	91-59-8	UHC	0.52	NA	X	--	--	--	--
2-Nitroaniline	88-74-4	UHC	0.27	14	--	--	--	--	--
2-Nitrophenol	88-75-5	UHC	0.028	13	--	X	--	--	X
4-Bromophenyl phenyl ether	101-55-3	UHC	0.055	15	--	--	--	--	X
4-Chloroaniline	106-47-8	UHC	0.46	16	--	--	--	--	--
4-Nitroaniline	100-01-6	UHC	0.028	28	--	--	--	--	--
Acenaphthene	83-32-9	UHC	0.059	3.4	--	--	X	--	X
Acenaphthylene	208-96-8	UHC	0.059	3.4	--	--	--	--	X
Anthracene	120-12-7	UHC	0.059	3.4	--	--	--	--	X
Benz(a)anthracene	56-55-3	UHC	0.059	3.4	--	--	--	--	X
Benzo(a)pyrene	50-32-8	UHC	0.061	3.4	--	--	--	--	X
Benzo(b)fluoranthene	205-99-2	UHC	0.11	6.8	--	--	--	--	X
Benzo(g,h,i)perylene	191-24-2	UHC	0.0055	1.8	--	--	--	--	X
Benzo(k)fluoranthene	207-08-9	UHC	0.11	6.8	--	--	--	--	X
bis(2-Chloroethoxy)methane	111-91-1	UHC	0.036	7.2	--	--	--	--	--
bis(2-Chloroethyl)ether	111-44-4	UHC	0.033	6	--	--	--	--	--
bis(2-ethylhexyl)phthalate	117-81-7	UHC	0.28	28	X	X	X	X	X
Bromodichloromethane	75-27-4	UHC	0.35	15	--	--	--	--	X
Bromoform	75-25-2	UHC	0.63	15	--	--	--	--	--
Bromomethane	74-83-9	UHC	0.11	15	--	--	--	--	X
Carbon disulfide	75-15-0	UHC	3.8	4.8 mg/l TCLP	--	X	--	--	X
Chlorobenzene	108-90-7	UHC	0.057	6	--	X	--	--	X
Chloroethane	75-00-3	UHC	0.27	6	--	--	--	--	X
Chloromethane	74-87-3	UHC	0.19	30	--	--	X	--	X
Chrysene	218-01-9	UHC	0.059	3.4	--	--	--	--	X
cis-1,3-Dichloropropylene	10061-01-5	UHC	0.036	18	--	X	--	--	X
Dibenzo(a,h)anthracene	53-70-3	UHC	0.055	8.2	--	--	--	--	X
Dibromochloromethane	124-48-1	UHC	0.057	15	--	--	--	--	--
Dichlorodifluoromethane	75-71-8	UHC	0.23	7.2	--	--	--	--	X
Diethyl phthalate	84-66-2	UHC	0.2	28	--	X	--	X	X
Dimethyl phthalate	131-11-3	UHC	0.047	28	--	--	--	--	--
Di-n-octyl phthalate	117-84-0	UHC	0.017	28	X	X	--	--	X
Diphenylamine	122-39-4	UHC	0.92	13	--	--	--	--	X
Ethyl acetate	141-78-6	UHC	0.34	33	--	X	X	X	--
Ethyl benzene	100-41-4	UHC	0.057	10	--	X	--	--	X
Ethyl Ether	60-29-7	UHC	0.12	160	X	--	--	--	--

Compound or Element Name	CAS Number	DST and SST Part A Associated Waste Code(s)[a]	Wastewater Standard, Concentration (mg/l)	Non-Wastewater Standard, Concentration in (mg/kg) unless noted as "mg/l TCLP"	Used At Hanford (RPP-RPT-61301)[b]	Measured in DST Headspace or Liquid (RPP-RPT-61301)[b]	Reported as Detected in Liquid or Solid RPP-21854[b]	In Table A.1 of SRNL-RP-2018-00687[b]	In Table 3[c] of RPP-RPT-54699[b]
Fluoranthene	206-44-0	UHC	0.068	3.4	--	X	--	--	X
Fluorene	86-73-7	UHC	0.059	3.4	--	--	--	**X**	X
Hexachlorobenzene	118-74-1	UHC	0.055	10	--	--	--	--	--
Hexachlorocyclopentadiene	77-47-4	UHC	0.057	2.4	--	--	--	--	--
Indeno(1,2,3-c,d) pyrene	193-39-5	UHC	0.0055	3.4	--	--	--	--	X
Isobutyl alcohol	78-83-1	UHC	5.6	170	X	--	--	**X**	X
Naphthalene	91-20-3	UHC	0.059	5.6	--	X	--	--	X
N-Nitroso-di-n-propylamine	621-64-7	UHC	0.4	14	--	X	--	X	X
Pentachlorophenol	87-86-5	UHC	0.089	7.4	--	X	--	--	--
Phenanthrene	85-01-8	UHC	0.059	5.6	--	--	--	--	X
Phenol	108-95-2	UHC	0.039	6.2	X	X	--	X	X
p-Nitrophenol	100-02-7	UHC	0.12	29	--	X	--	--	--
Pyrene	129-00-0	UHC	0.067	8.2	--	X	X	--	X
Toluene	108-88-3	UHC	0.08	10	X	X	X	--	X
trans-1,3-Dichloropropylene	10061-02-6	UHC	0.036	18	--	X	--	--	X
Trichlorofluoromethane	75-69-4	UHC	0.02	30	--	X	--	--	X
Xylene(m,p,o)	1330-20-7	UHC	0.32	30	X	X	X	--	X
1,4-Dioxane	123-91-1	UHC	12	170	--	--	--	--	X
Diphenylnitrosamine	86-30-6	UHC	0.92	13	--	--	--	--	--
1,2-Diphenylhydrazine	122-66-7	UHC	0.087	NA	--	--	--	--	--
Disulfoton	298-04-4	UHC	0.017	6.2	--	--	--	--	--
Endosulfan I	959-98-8	UHC	0.023	0.066	--	--	--	--	--
Endosulfan II	33213-65-9	UHC	0.029	0.13	--	--	--	--	--
Endosulfan sulfate	1031-07-8	UHC	0.029	0.13	--	--	--	--	--
Endrin	72-20-8	UHC	0.0028	0.13	--	--	--	--	--
Endrin aldehyde	7421-93-4	UHC	0.025	0.13	--	--	--	--	--
Ethyl cyanide/Propanenitrile	107-12-0	UHC	0.24	360	--	--	--	**X**	X
Ethyl methacrylate	97-63-2	UHC	0.14	160	--	--	--	--	--
Ethylene oxide	75-21-8	UHC	0.12	NA	--	--	--	--	--
Famphur	52-85-7	UHC	0.017	15	--	--	--	--	--
Heptachlor	76-44-8	UHC	0.0012	0.066	--	--	X	--	--
1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin (1,2,3,4,6,7,8-HpCDD)	35822-46-9	UHC	0.000035	0.0025	--	--	--	--	--
1,2,3,4,6,7,8-Heptachlorodibenzofluran (1,2,3,4,6,7,8-HpCDF)	67562-39-4	UHC	0.000035	0.0025	--	--	--	--	--
1,2,3,4,7,8,9-Heptachlorodibenzofluran (1,2,3,4,7,8,9-HpCDF)	55673-89-7	UHC	0.000035	0.0025	--	--	--	--	--
Heptachlor epoxide	1024-57-3	UHC	0.016	0.066	--	--	X	--	--
HxCDDs (All Hexachlorodibenzo-p-dioxins)	NA	UHC	0.000063	0.001	--	--	--	--	--
HxCDFs (All Hexachlorodibenzofurans)	NA	UHC	0.000063	0.001	--	--	--	--	--
Hexachloropropylene	1888-71-7	UHC	0.035	30	--	--	--	--	--

Compound or Element Name	CAS Number	DST and SST Part A Associated Waste Code(s)[a]	Wastewater Standard, Concentration (mg/l)	Non-Wastewater Standard, Concentration in (mg/kg) unless noted as "mg/l TCLP"	Used At Hanford (RPP-RPT-61301)[b]	Measured in DST Headspace or Liquid (RPP-RPT-61301) [b]	Reported as Detected in Liquid or Solid RPP-21854 [b]	In Table A.1 of SRNL-RP-2018-00687 [b]	In Table 3 [c] of RPP-RPT-54699[b]
Iodomethane	74-88-4	UHC	0.19	65	--	--	--	--	--
Isodrin	465-73-6	UHC	0.021	0.066	--	--	--	--	X
Isosafrole	120-58-1	UHC	0.081	2.6	--	--	--	--	--
Kepone	143-50-0	UHC	0.0011	0.13	--	--	--	--	--
Methacrylonitrile	126-98-7	UHC	0.24	84	--	--	--	--	X
Methapyrilene	91-80-5	UHC	0.081	1.5	--	--	--	--	--
Methoxychlor	72-43-5	UHC	0.25	0.18	--	--	--	--	--
3-Methylcholanthrene	56-49-5	UHC	0.0055	15	--	--	--	--	X
4,4-Methylene bis(2-chloroaniline)	101-14-4	UHC	0.5	30	--	--	--	--	--
Methyl methacrylate	80-62-6	UHC	0.14	160	--	--	--	--	--
Methyl methanesulfonate	66-27-3	UHC	0.018	NA	--	--	--	--	--
Methyl parathion	298-00-0	UHC	0.014	4.6	--	--	--	--	--
5-Nitro-o-toluidine	99-55-8	UHC	0.32	28	--	--	--	--	--
N-Nitrosodiethylamine	55-18-5	UHC	0.4	28	--	--	X	--	--
N-Nitrosodimethylamine	62-75-9	UHC	0.4	2.3	--	--	X	X	X
N-Nitroso-di-n-butylamine	924-16-3	UHC	0.4	17	--	--	--	--	--
N-Nitrosomethylethylamine	10595-95-6	UHC	0.4	2.3	--	--	--	--	--
N-Nitrosomorpholine	59-89-2	UHC	0.4	2.3	--	--	--	X	X
N-Nitrosopiperidine	100-75-4	UHC	0.013	35	--	--	--	--	--
N-Nitrosopyrrolidine	930-55-2	UHC	0.013	35	--	--	--	--	--
1,2,3,4,6,7,8,9-Octachlorodibenzo-p-dioxin (OCDD)	3268-87-9	UHC	0.000063	0.005	--	--	--	--	--
1,2,3,4,6,7,8,9-Octachlorodibenzofluran (OCDF)	39001-02-0	UHC	0.000063	0.005	--	--	--	--	--
Parathion	56-38-2	UHC	0.014	4.6	--	--	--	--	--
Pentachlorobenzene	608-93-5	UHC	0.055	10	--	--	--	--	--
PeCDDs (All Pentachlorodibenzo-p-dioxins)	NA	UHC	0.000063	0.001	--	--	--	--	--
PeCDFs (All Pentachlorodibenzofurans)	NA	UHC	0.000035	0.001	--	--	--	--	--
Pentachloroethane	76-01-7	UHC	0.055	6	--	--	--	--	--
Pentachloronitrobenzene	82-68-8	UHC	0.055	4.8	--	--	--	--	X
Phenacetin	62-44-2	UHC	0.081	16	--	--	--	--	--
1,3-Phenylenediamine	108-45-2	UHC	0.01	0.66	--	--	--	--	--
Phorate	298-02-2	UHC	0.021	4.6	--	--	--	--	--
Phthalic acid	100-21-0	UHC	0.055	28	--	--	--	--	X
Phthalic anhydride	85-44-9	UHC	0.055	28	--	--	--	--	--
Pronamide	23950-58-5	UHC	0.093	1.5	--	--	--	--	--
Safrole	94-59-7	UHC	0.081	22	--	--	--	--	--
Silvex/2,4,5-TP	93-72-1	UHC	0.72	7.9	--	--	--	--	X
1,2,4,5-Tetrachlorobenzene	95-94-3	UHC	0.055	14	--	--	--	--	--
TCDDs (All Tetrachlorodibenzo-p-dioxins)	NA	UHC	0.000063	0.001	--	--	--	--	--

Compound or Element Name	CAS Number	DST and SST Part A Associated Waste Code(s)[a]	Wastewater Standard, Concentration (mg/l)	Non-Wastewater Standard, Concentration in (mg/kg) unless noted as "mg/l TCLP"	Used At Hanford (RPP-RPT-61301)[b]	Measured in DST Headspace or Liquid (RPP-RPT-61301) [b]	Reported as Detected in Liquid or Solid RPP-21854 [b]	In Table A.1 of SRNL-RP-2018-00687 [b]	In Table 3 [c] of RPP-RPT-54699[b]
TCDFs (All Tetrachlorodibenzofurans)	NA	UHC	0.000063	0.001	--	--	--	--	X
1,1,1,2-Tetrachloroethane	630-20-6	UHC	0.057	6	--	--	--	--	X
2,3,4,6-Tetrachlorophenol	58-90-2	UHC	0.03	7.4	--	--	--	--	X
Toxaphene	8001-35-2	UHC	0.0095	2.6	--	--	--	--	--
2,4,5-Trichlorophenoxyacetic acid/2,4,5-T	93-76-5	UHC	0.72	7.9	--	--	--	--	--
1,2,3-Trichloropropane	96-18-4	UHC	0.85	30	--	--	--	--	--
tris-(2,3-Dibromopropyl) phosphate	126-72-7	UHC	0.11	0.1	--	--	--	--	--
Acetonitrile	75-05-8	UHC	5.6	38	--	--	X	X	X
Acetophenone	98-86-2	UHC	0.01	9.7	--	--	--	X	X
2-Acetylaminofluorene	53-96-3	UHC	0.059	140	--	--	--	--	--
Acrolein	107-02-8	UHC	0.29	NA	--	--	--	X	X
Acrylamide	79-06-1	UHC	19	23	--	--	--	--	--
Acrylonitrile	107-13-1	UHC	0.24	84	--	--	--	--	X
Aldrin	309-00-2	UHC	0.021	0.066	--	--	X	--	--
4-Aminobiphenyl	92-67-1	UHC	0.13	NA	--	--	--	--	--
Aniline	62-53-3	UHC	0.81	14	--	--	--	--	--
o-Anisidine (2-methoxyaniline)	90-04-0	UHC	0.01	0.66	--	--	--	--	--
Aramite	140-57-8	UHC	0.36	NA	--	--	--	--	--
alpha-BHC	319-84-6	UHC	0.00014	0.066	--	--	X	--	X
beta-BHC	319-85-7	UHC	0.00014	0.066	--	--	X	--	X
delta-BHC	319-86-8	UHC	0.023	0.066	--	--	--	--	X
gamma-BHC	58-89-9	UHC	0.0017	0.066	--	--	--	--	X
Benzal chloride	98-87-3	UHC	0.055	6	--	--	--	--	--
2-sec-Butyl-4,6-dinitrophenol/Dinoseb	88-85-7	UHC	0.066	2.5	--	--	--	--	X
Chlordane (alpha and gamma isomers)	57-74-9	UHC	0.0033	0.26	--	--	--	--	--
Chlorobenzilate	510-15-6	UHC	0.1	NA	--	--	--	--	--
2-Chloro-1,3-butadiene	126-99-8	UHC	0.057	0.28	--	--	--	--	--
bis(2-Chloroisopropyl)ether	39638-32-9	UHC	0.055	7.2	--	--	--	--	--
p-Chloro-m-cresol	59-50-7	UHC	0.018	14	--	--	--	--	X
2-Chloroethyl vinyl ether	110-75-8	UHC	0.062	NA	--	--	--	--	--
3-Chloropropylene	107-05-1	UHC	0.036	30	--	--	--	--	X
p-Cresidine	120-71-8	UHC	0.01	0.66	--	--	--	--	--
o-Cresol	95-48-7	UHC	0.11	5.6	--	--	--	X	--
m-Cresol (difficult to distinguish from p-cresol)	108-39-4	UHC	0.77	5.6	--	--	--	--	--
p-Cresol (difficult to distinguish from m-cresol)	106-44-5	UHC	0.77	5.6	--	--	--	--	--
o,p′-DDD	53-19-0	UHC	0.023	0.087	--	--	--	--	--
p,p′-DDD	72-54-8	UHC	0.023	0.087	--	--	--	--	--
o,p′-DDE	3424-82-6	UHC	0.031	0.087	--	--	--	--	--
p,p′-DDE	72-55-9	UHC	0.031	0.087	--	--	--	--	--

Compound or Element Name	CAS Number	DST and SST Part A Associated Waste Code(s)[a]	Wastewater Standard, Concentration (mg/l)	Non-Wastewater Standard, Concentration in (mg/kg) unless noted as "mg/l TCLP"	Used At Hanford (RPP-RPT-61301)[b]	Measured in DST Headspace or Liquid (RPP-RPT-61301) [b]	Reported as Detected in Liquid or Solid RPP-21854 [b]	In Table A.1 of SRNL-RP-2018-00687 [b]	In Table 3 [c] of RPP-RPT-54699[b]
o,p′-DDT	789-02-6	UHC	0.0039	0.087	--	--	--	--	--
p,p′-DDT	50-29-3	UHC	0.0039	0.087	--	--	--	--	--
Dibenz(a,e)pyrene	192-65-4	UHC	0.061	NA	--	--	--	--	X
1,2-Dibromoethane/Ethylene dibromide	106-93-4	UHC	0.028	15	--	--	--	--	X
Dibromomethane	74-95-3	UHC	0.11	15	--	--	--	--	--
trans-1,2-Dichloroethylene	156-60-5	UHC	0.054	30	--	--	--	--	X
2,6-Dichlorophenol	87-65-0	UHC	0.044	14	--	--	--	--	--
2,4-Dichlorophenoxyacetic acid/2,4-D	94-75-7	UHC	0.72	10	--	--	--	--	--
Dieldrin	60-57-1	UHC	0.017	0.13	--	--	--	--	--
p-Dimethylaminoazobenzene	60-11-7	UHC	0.13	NA	--	--	--	--	--
2,4-Dimethylaniline (2,4-xylidine)	95-68-1	UHC	0.01	0.66	--	--	--	--	--
1,4-Dinitrobenzene	100-25-4	UHC	0.32	2.3	--	--	--	--	--
2,6-Dinitrotoluene	606-20-2	UHC	0.55	28	--	--	--	--	--
Antimony	7440-36-0	UHC	1.9	1.15 mg/l TCLP	X	--	--	--	X
Arsenic	7440-38-2	D004 [d], UHC	1.4	5.0 mg/l TCLP	X	--	--	--	X
Barium	7440-39-3	D005 [d], UHC	1.2	21 mg/l TCLP	X	--	--	--	X
Beryllium	7440-41-7	UHC	0.82	1.22 mg/l TCLP	X	--	--	--	X
Cadmium	7440-43-9	D006 [d], UHC	0.69	0.11 mg/l TCLP	X	--	--	--	X
Chromium (Total)	7440-47-3	D007 [d], UHC	2.77	0.60 mg/l TCLP	X	--	--	--	X
Cyanides (Total)	57-12-5	UHC	1.2	590	X	--	--	--	X
Cyanides (Amenable)	57-12-5	UHC	0.86	30	X	--	--	--	X
Lead	7439-92-1	D008 [d], UHC	0.69	0.75 mg/l TCLP	X	--	--	--	X
Mercury - All Others	7439-97-6	D009 [d], UHC	NA	0.20 mg/l TCLP	X	--	--	--	X
Nickel	7440-02-0	UHC	3.98	11 mg/l TCLP	X	--	--	--	X
Selenium	7782-49-2	D010 [d], UHC	0.82	5.7 mg/l TCLP	X	--	--	--	X
Silver	7440-22-4	D011 [d], UHC	0.43	0.14 mg/l TCLP	X	--	--	--	X
Thallium	7440-28-0	UHC	1.4	0.20 mg/l TCLP	X	--	--	--	X

[a] Waste code and UHC identification based on application of waste designation requirements and LDR requirements to the codes listed in DST Part A and SST Part A. When a constituent is assigned one of the F001-F005 waste codes, it is because process knowledge exists to apply the code based on how the constituent was used. If the F001-F005 waste code does not apply, then a characteristic "D" code is listed in the cell if it is also listed on the DST and SST Part A. When a constituents is identified by a "D" code based on the DST and SST Part A, if such a constituent is below the waste designation level for the "D" code when waste is sampled before treatment, it could still be identified as a UHC if the constituent exists above the applicable treatment standard. All other constituents are identified as a UHC, except Aroclors which are potentially regulated under RCRA as a UHC and under TSCA.

[b] An "X" indicates an affirmative response in the indicated column.

[c] Table 3 of Appendix A of RPP-RPT-54699 which is a reduced list of UHCs generated by removing items not applicable to Hanford, or likely not stable in a high radiation and high caustic environment.

[d] Wastewater and non-wastewater concentration-based standards are listed, however, this document assumes these will be the treatment standards used for SLAW and not high-level vitrification (HLVIT).

3.4.3 Additional Groundwater Protection Requirements at IDF

The Hanford Federal Facility RCRA Permit for IDF contains a provision requiring DOE to prepare and maintain a "Risk Budget Tool." This requirement does not exist in WAC 173-303 or 40 CFR 264. The permit conditions under III.11.5.a states:

> *III.11.I.5 Modeling – Risk Budget Tool*
> *III.11.I.5.a The Permittees must create and maintain a modeling - risk budget tool, which models the future impacts of the planned IDF waste forms (including input from analyses performed as specified in Permit Conditions III.11.I.2.a through III.11.I.2.a.ii) and their impact to underlying vadose and ground water. This software tool will be submitted for Ecology review as soon as possible after issuance of Final Tank Closure and Waste Management EIS and receipt of underlying codes and data packages, and at least 180 days prior to the date DOE expects to receive waste at IDF. The risk budget tool shall be updated at least every 5 years. The model will be updated more frequently if needed, to support permit modifications or SEPA Threshold Determinations whenever a new waste stream or significant expansion is being proposed for the IDF. This risk budget tool shall be conducted in manner that is consistent with state and federal requirements, and represents a risk analysis of all waste previously disposed of in the entire IDF (both cell 1 and cell 2) and those wastes expected to be disposed of in the future for the entire IDF to determine cumulative impacts. The groundwater impact should be modeled to evaluate fate and transport in the groundwater aquifer(s) and should be compared against various performance standards including but not limited to drinking water standards (40 CFR 141 and 40 CFR 143). Ecology will review modeling assumptions, input parameters, and results and will provide comments to the Permittees. Ecology comments shall be dispositioned through the RCR process and will be reflected in further modeling to modify the IDF ILAW waste acceptance criteria as appropriate.*
> *III.11.I.5.a.i The modeling-risk budget tool will include a sensitivity analysis reflecting parameters and changes to parameters as requested by Ecology.*
> *III.11.I.5.a.ii If these modeling efforts indicate results within 75% of a performance standard [including but not limited to federal drinking water standards (40 CFR 141 and 40 CFR 143)], Ecology and the Permittees will meet to discuss mitigation measures or modified waste acceptance criteria for specific waste forms.*
> *III.11.I.5.a.iii When considering all the waste forms to be disposed of in IDF, the Permittees shall not dispose of any waste that will result (through forward looking modeling or in real groundwater concentrations data) in a violation of any state or federal regulatory limit, specifically including but not limited to drinking water standards for any constituent as defined in 40 CFR 141 and 40 CFR 143."*

This language requires disposal of SLAW in the IDF must be factored into the Risk Budget Tool. The Risk Budget Tool has been developed and submitted by DOE to Ecology as part of the IDF permitting process for DFLAW (DOE 2020).

3.4.3.1 Implications of RCRA Groundwater Protection for Onsite SLAW Disposal

Currently fate-and-transport modeling that supports the IDF PA assumes that vitrification is used for supplemental treatment of LAW. Developing a cementitious waste for LAW will require careful considerations on the impacts of the new material on the modeled impact of the IDF to groundwater. Factors that will need to be considered include, but are not limited to:

- Increased nitrate and nitrite inventory that will be contained in a cemented LAW form.

- Presence of organic constituents that would have been destroyed in vitrification but remain in cementitious waste form.
- Impacts of the constituent of concern (COC) release mechanisms for the two waste forms (glass dissolution versus diffusion from cementitious materials).
- Changes in the inventory of secondary wastes produced as a result of LAW solidification by a low temperature process.

The last issue is of importance because a vitrification process produces substantially more contaminated solid secondary waste than a cementation facility. Current IDF modeling shows that solid secondary waste contributes a large portion of the predicted 129-I in groundwater. For example, the IDF Risk Budget Tool shows that approximately 42% of the 129-I is associated with solid secondary waste, but the peak groundwater concentration caused by that waste is at least three times higher than that caused by glass (RPP-CALC-63176).

3.5 Characteristics of a SLAW Form Suitable for the IDF

3.5.1 Conceptual Model for Constituent Release from Grout

Radiological waste acceptance criteria for the IDF are set such that the facility meets the radiological performance objectives established in DOE O 435.1 and DOE M 435.1-1. The IDF PA (RPP-RPT-59958) is the tool used to calculate concentration limits for radionuclides for waste forms expected to be disposed of in IDF. In addition, PA fate-and-transport models are used to evaluate non-radiological species against groundwater protection requirements (see condition III.11.I.5 of the IDF Permit WA7-89000-8967 which is reproduced in Section 3.4.3). For a cementitious waste form the PA models simulate the release of constituents as a diffusive process that is affected by physical and chemical processes in the waste form. These properties are commonly generated through physical measurements (e.g., porosity, hydraulic conductivity, water retention); sorption testing (e.g., distribution coefficient, K_d); and leach testing (e.g., observed diffusivities, retardation factors). The following paragraphs are a summary of key characteristics used to model COC release from cementitious materials. A full description is provided elsewhere (Asmussen et al. 2019a).

Non-reacting, soluble, non-sorbing species in a grout waste form are released at the fastest rate possible and this rate is represented as an intrinsic diffusivity, D_i. Other species that sorb to the matrix or are involved in chemical reactions within the waste form (e.g., REDOX, solubility) are released at a slower rate. The rate of release for a sorbing/reacting species is represented as an apparent diffusivity (called D_a, in numerical simulations) or as an observed diffusivity (called D_{obs}, in experimental work). The ratio of the D_i to D_a *or* D_{obs} is termed the retardation factor, R, which represents the processes in the waste form slowing contaminant release. The larger the R-value, the slower a contaminant is released from the waste form.

The R-value itself is not an input parameter in numerical simulations. In order to numerically represent a grout waste form's ability to retain a specific contaminant, as dictated by the magnitude of the R-value, the K_d (contaminant specific solid to liquid equilibrium distribution coefficient) within the waste form is manipulated. A large K_d value for the contaminant within the waste form generates a slowed release. The relationship between R and the simulation K_d value is based on equation 3.1.

$$R = \left[1 + \frac{(1-\varepsilon)\rho_s K_d}{S\varepsilon}\right] \tag{3.1}$$

where:

R = retardation factor, unitless

ε = porosity, volume fraction

ρ_s = solid-phase density, g/cm^3

K_d = contaminant specific solid to liquid equilibrium distribution coefficient, cm^3/g

S = moisture content, volume fraction

The simulation constructs of the IDF are described in detail in the 2017 IDF PA (RPP-RPT-59958). Briefly, water is assumed to infiltrate the IDF, contact the waste and facilitate diffusion of contaminants from the waste form. COC release ignores the presence of an external package surrounding the cementitious waste form. Constituents then migrate through, and interact with, the backfill. Currently, minimal interactions assumed between the backfill and the contaminants. Eventually the soluble contaminants are released from the bottom of the IDF into the unsaturated zone at an assumed failure location in the facility's double liner, commonly represented at the sumps. Once in the unsaturated zone, migration of the COCs with pore water continues until the material reaches the saturated zone where the pore water is incorporated into groundwater flow. The fractional release of the contaminant from the IDF over time is proportional to inventory in the waste form and to the square root of the apparent/observed diffusivity for the waste form (and in-turn in the R value).

Water infiltration is initially reduced from the assumed level of 3.5 to 0.5 mm/y by the presence of the cap that is planned for the IDF. After 500 years, the cap is assumed to fail and the infiltration rate increases to 3.5 mm/yr.

3.5.2 Required Performance for Grouted LAW

In FY19 work was carried out using model simulations to determine a leachability threshold for an inventory of SLAW, similar to that described by the feed vector presented in this report, in the IDF that ensures regulatory limits are met when only the SLAW inventory is considered (Asmussen et al. 2019a). Regulatory limits for the IDF are described in detail in Section 3.2 of this report. The work by Asmussen et al. 2019a focused on assessing historical Hanford grout leachability data (Table 3-6) against this threshold and identifying where improvement is required. In addition, this work established a performance metric for four contaminants (Tc, I, NO_3^-, and Cr) in the SLAW inventory. This performance metric serves as a means to determine if incorporating new laboratory data into the IDF PA model will result in contaminant releases from the SLAW inventory that correspond to predicted groundwater concentrations below the regulatory standards.

Table 3-6: Waste form types included in previous Hanford Site disposal assessments. Adapted from Table 1-1 of Asmussen et al. (2019a).

Year	Assessment Type	LAW Glass	Secondary Waste Grout	LAW Grout
1995	Performance Assessment	- [a]	-	×
1997	Performance Assessment	× [b]	-	-
2001	Performance Assessment	×	-	-
2003	Risk Assessment	×	-	×
2005	Performance Assessment	×	×	-
2012	Environmental Impact Study	×	×	×
2017	Performance Assessment	×	×	-

[a] Symbol indicates the waste form was not included in the assessment

[b] Symbol indicates the waste form was included in the assessment

The performance metric was quantified as the R-value based on the waste form retention capacity relative to a mobile (i.e., non-sorbing/non-reacting) species and a corresponding fractional release rate. The reported R-value represents a minimum needed to maintain contaminant concentrations below the groundwater compliance standard. For comparison purposes, R can be converted to a corresponding laboratory observed diffusivity value (D_{obs}) and overall fractional release rate from the IDF. The R performance metric was determined through an iterative set of simulations varying the grout waste form distribution coefficient (K_d) representing grout waste within the IDF and monitoring the resulting transport to groundwater. A brief summary of the results of this work are presented in Table 3-7 and described in the paragraphs that follow. Note that changes in the inventory of each contaminant, Table 3-7, were not considered in this effort, but a lower inventory of a contaminant present in the IDF would decrease the corresponding performance metric accordingly.

Table 3-7: The distribution coefficient (K_d) used in the simulations, and the corresponding retardation factor (R) performance metric values; the associated fractional release rate from the simulated IDF; and, associated observed diffusivity (D_{obs}). Adapted from Tables 3-2 and S-1 of Asmussen et al. (2019a).

Species	IDF Inventory	Simulation Waste Form K_d, mL/g	Performance Metric, R	Fractional Release Rate	Corresponding Observed Diffusivity (D_{obs})
Technetium	11593 Ci	45	113	2.1×10^{-6} yr^{-1}	2×10^{-11} cm^2/s
Iodine	10.5 Ci	35	87	5.4×10^{-5} yr^{-1}	3×10^{-11} cm^2/s
Nitrate	2.54×10^{7} kg	0.5	2	1.1×10^{-5} yr^{-1}	2×10^{-9} cm^2/s.
Chromium	7.98×10^{5} kg	26	65	3.4×10^{-6} yr^{-1}	4×10^{-11} cm^2/s

Technetium: A contribution from Tc to the fractional release below the IDF that is below 2.1×10^{-6} yr^{-1}, which is equivalent to a waste form that would measure an observed diffusivity of approximately 2×10^{-11} cm^2/s in laboratory leach testing, would ensure Tc concentrations at the point of compliance would stay below the limits. In previous performance and risk calculations the Tc observed diffusivities used in the calculations have been greater than the established performance metric, orange triangles in Figure 3-4a. However, several laboratory tests have measured Tc observed diffusivities below the limit established by the performance metric, including LAW Cast Stone, getter-containing systems, and hydrated-lime based formulations. With the average Cast Stone performance being close to the performance metric, achieving a 5-10 × decrease in overall release (through performance improvement or inventory management) would maintain regulatory compliance. Figure 3-4b shows datasets from the LAW Cast Stone screening tests (Westsik et al. 2013b; SRNL-RP-2018-00687). One of the largest challenges with Tc is understanding and predicting the re-oxidation of immobilized Tc over long time periods in the IDF.

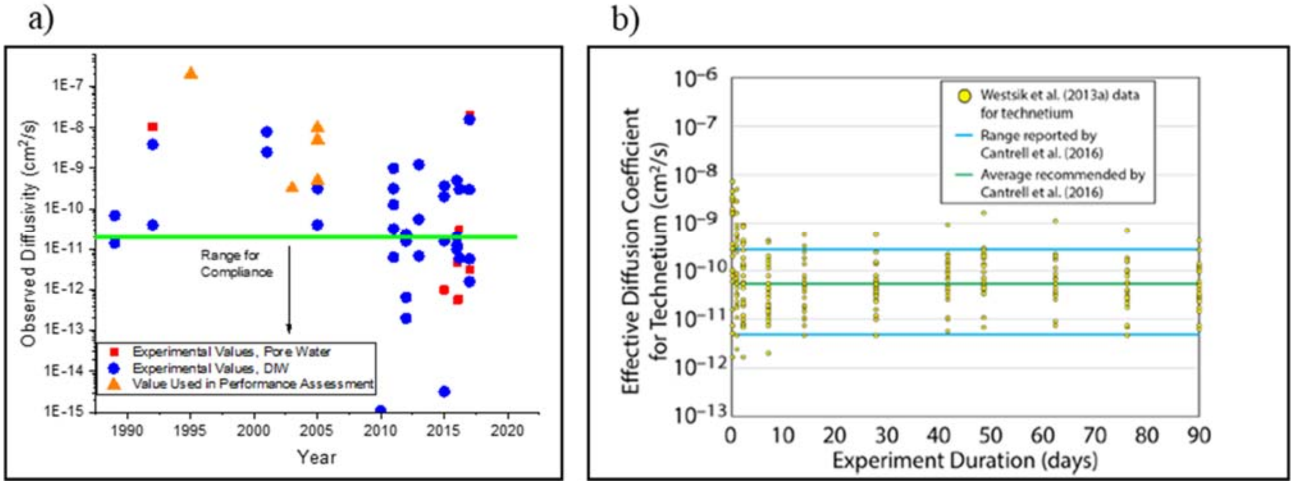


Figure 3-4: (a) Comparison of historical technetium leach test data from experimental results (blue dots and red squares) and those used in prior performance/risk assessments (orange triangles) from PNNL-28992 (Asmussen et al. 2019a) and (b) comparison of Tc leachability in screening tests of LAW Cast Stone (Westsik et al. 2013b; SRNL-RP-2018-00687) and was adapted from SRNL-RP-2018-00687 and the effective diffusion coefficient refers to an experimentally measured observed diffusion coefficient.

Iodine: A contribution from iodine (as 129-I) to the fraction release below the IDF that is below 5.4×10^{-5} yr^{-1}, which is equivalent to a waste form that would measure an observed diffusivity of approximately 3×10^{-11} cm^2/s in leach testing, would ensure iodine concentrations at the point of compliance would stay below limits. Analysis of historical iodine (as iodide) observed diffusivities showed that the values used in performance and risk assessment do not meet performance requirements to be protective of the groundwater. Most iodine observed diffusivities range between 1×10^{-8} cm^2/s and 1×10^{-9} cm^2/s, Figure 3-5. In order to achieve diffusivities that meet the performance metric a 10× improvement in retention at minimum or a reduction in inventory would be required (note that a 10× decrease in overall release is equivalent to a 100× decrease in D_{obs}). The one exception to this finding was for formulations containing Ag-based getters with getter loadings >5 wt% in the waste form, where reported observed diffusivity values were several orders of magnitude below the observed diffusivity limit established by the performance metric.

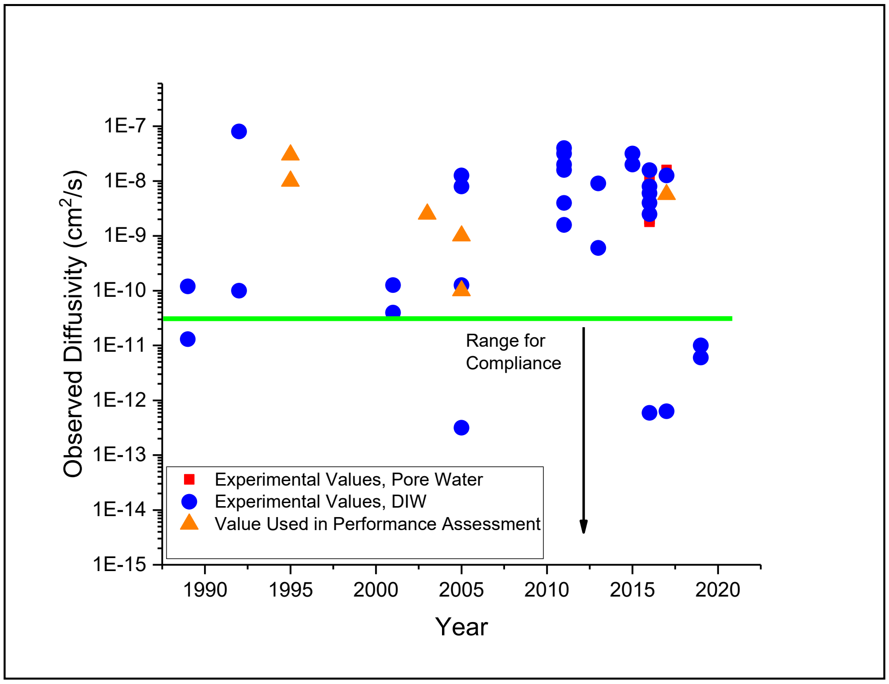


Figure 3-5: Comparison of historical iodine leach test data from experimental results (blue dots and red squares) and those used in prior performance/risk assessments (orange triangles) from PNNL-28992 (Asmussen et al. 2019a).

Nitrate: A contribution from nitrate to the fraction release below the IDF that is below 1.1×10^{-5} yr^{-1}, which is equivalent to a waste form that would measure an observed diffusivity of approximately 2×10^{-9} cm^2/s in leach testing, would ensure nitrate concentrations at the point of compliance would stay below the limits. Of all nitrate leach data assessed, Figure 3-6, only a small number of tests (from immobilized liquid secondary waste simulants) measured an observed nitrate diffusivity below the limit established by the performance metric. However, a magnitude 10× decrease from the average nitrate D_{obs} (which corresponds to a 3.2× reduction in release rate) would likely meet the metric.

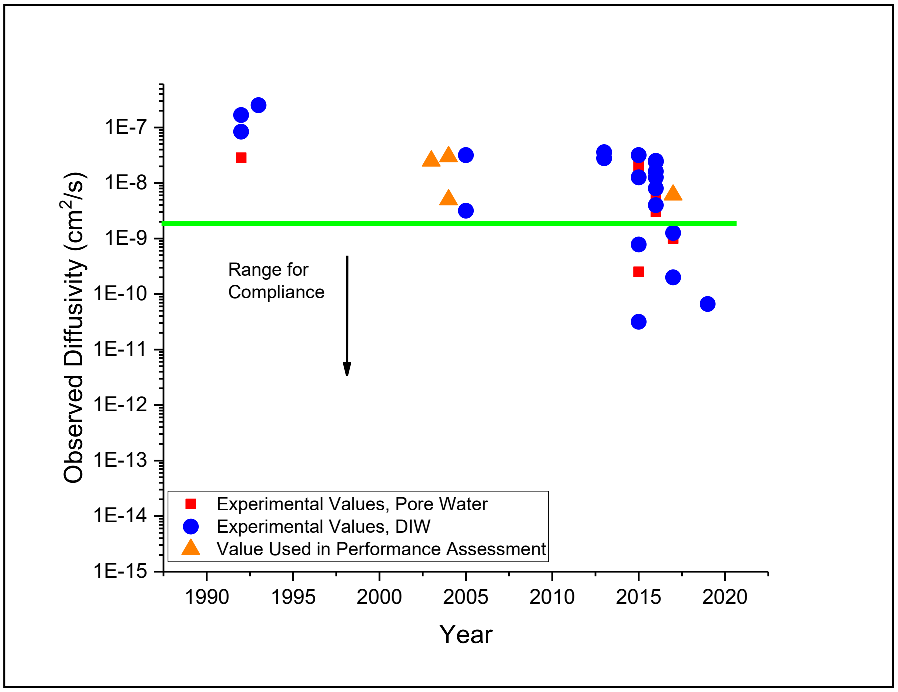


Figure 3-6: Comparison of historical nitrate leach test data from experimental results (blue dots and red squares) and those used in prior performance/risk assessments (orange triangles) from PNNL-28992 (Asmussen et al. 2019a).

Chromium: The evaluation of technologies for Cr was not included in this current work since historical toxicity characteristic leaching procedure (TCLP) testing for Cr indicate adequate retention. According to a corresponding summary report (Westsik et al. 2013b):

> *…System Plan 6 predicts a maximum Cr level in the LAW supplemental feed of 1.0 × 10^{-2} M/M Na. The average Cr concentration is 2.4 × 10^{-3} M/M Na and the 95th percentile for Cr is 4.3 × 10^{-3} M/M Na. The 2003 Cast Stone work used the SST Blend saltcake simulant with a Cr level of 2.0 × 10^{-3} M/M Na and the SST saltcake blend of real waste with a ratio of 3.7 × 10^{-3} M/M Na (Rapko et al. 2003). Simulants used in testing Cast Stone for the secondary waste program used a Cr ratio as high as 4.2 × 10^{-3} M/M Na (Sundaram et al. 2011). Based on the preceding information, a Cr spike level of 4.3 × 10^{-3} M/M Na corresponding to the HTWOS 95th percentile composition was selected for supplemental LAW Cast Stone waste form testing.*

Even with the high Cr content in these tests, the TCLP testing showed no sample with a result >106 µg/L (the regulatory limit is 600 µg/L).

This observation was also confirmed in the 2019 work by Asmussen et al. (2019a) (Figure 3-7). It should be noted that the sufficient performance observed previously in TCLP and leach testing is due to reduction capacity available from slag in the waste form. If any slag-free waste forms are considered, they would need to ensure adequate Cr retention in qualification testing.

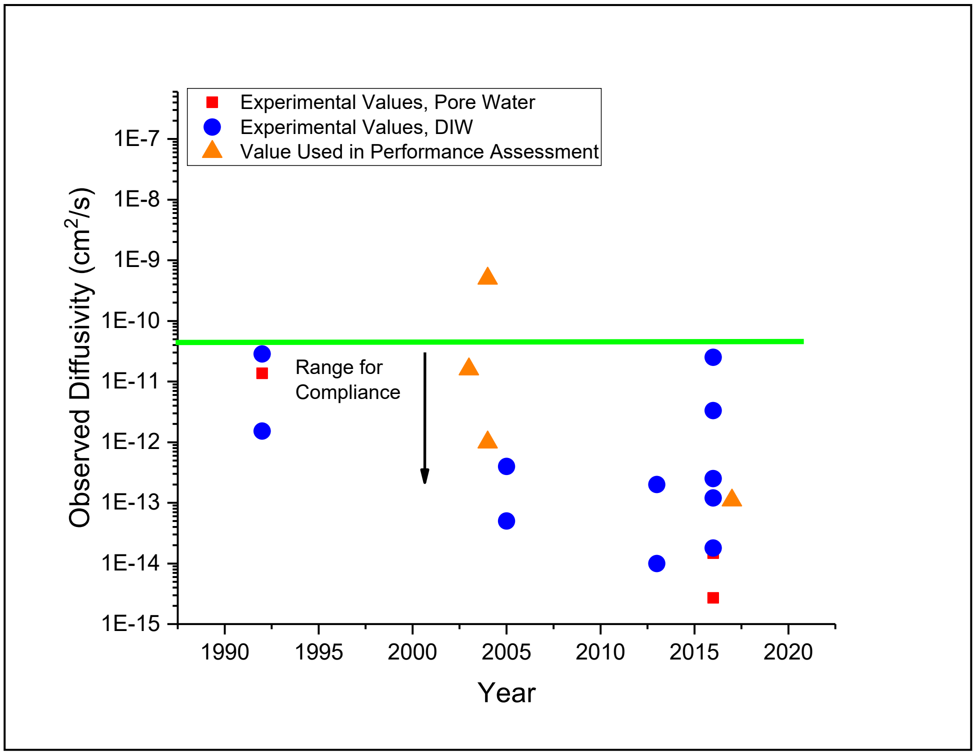


Figure 3-7: Comparison of historical chromium leach test data from experimental results (blue dots and red squares) and those used in prior performance/risk assessments (orange triangles) from PNNL-28992 (Asmussen et al. 2019a).

4.0 Regulatory Considerations for Offsite Disposal

Several other facilities outside the State of Washington may be viable locations for disposal of LAW. These locations include the Chem-Nuclear Site located in Barnwell, South Carolina, Clive Disposal Facility located in Clive, Utah, and Waste Control Specialists, LLC (WCS) located near Andrews, Texas. WCS is the only site immediately ready to receive Hanford SLAW without requiring modifications to their existing permits and so will be the only offsite location discussed in this section.

WSC is a commercially operated radioactive waste disposal facility licensed by the State of Texas to dispose of Class A, B, and C LLW and MLLW. WCS has two Low-Level Radioactive Waste disposal facilities; the Compact Waste Disposal Facility and the Federal Waste Disposal Facility (FWF). The FWF was designed, permitted, and constructed to accept Class A, B, and C LLW and MLLW that is owned by the federal government and managed under the Low-Level Radioactive Waste Policy Act as amended by the Low-Level Radioactive Waste Policy Amendments Act of 1985 (WCS 2015). This includes waste generated by the DOE, waste from U.S. Navy vessel decommissioning, and testing or production waste.

The WCS FWF is licensed to receive up to 26,000,000 ft^3 of Class A, B, or C waste containing no more than a total of 5,600,000 curies (http://www.wcstexas.com/facilities/federal-waste/). There is also an additional limit that only 8,100,000 ft^3 of containerized waste, containing no more than 5,500,000 curies, can be received before September 2024. It is important to note that these restrictions are sufficient to allow disposal of all the predicted cementitious LAW from Hanford. As reported in Table 3-4, cementitious LAW is predicted to be Class C or less. In addition, the quantity of solidified LAW generated from the SLAW feed vector described in Section 2.1 would be approximately 12,000,000 ft^3 (341,000 m^3)[1] and have a total radiological content of 604,300 curies. These values equate to 46% of the total allowable WCS FWF volume and 11% of the total allowable radiological content.

The geologic conditions associated with the WCS FWF eliminate the groundwater concerns described in Sections 3.2.1 and 3.2.3 for onsite disposal in the Hanford IDF. Wastes at the WSC are disposed 25 to 120 ft below the land surface in disposal cells that includes a 7-ft-thick multi-barrier liner. In addition, the WCS facilities are underlain by a 600-ft (185-m)-thick layer of red-bed clays, and there is no drinking water aquifer beneath or adjacent to the WCS facilities. For these reasons, there is no need to consider a drinking water pathway when evaluating the long-term impacts of waste disposal at the FWF.

WCS waste acceptance criteria are well defined (see Tables 4-1, 4-2, and 4-3). The first two tables establish radiological waste classification in a manner similar to 10 CFR 61.55 with the exception that Texas has added Ra-226 to the list of long-lived radionuclides that must be included in the classification process. The addition of this radionuclide does change the waste classifications reported in Table 3-4 for Hanford SLAW. The last column in Table 4-3 (Cask Waste) lists miscellaneous waste acceptance criteria that would apply to solidified SLAW. It is anticipated that solidified Hanford LAW would meet these miscellaneous requirements. Finally, certification that the solidified SLAW meets the LDR requirements described in Section 3.2.2 is also a waste acceptance criterion for the WCS (WSC 2015).

DOE M 435.1-1 contains an additional requirement for DOE to approve the offsite management of mixed waste in non-DOE facilities. This document assumes DOE will provide the approvals necessary for offsite management of treated SLAW. DOE M 435.1-1, Chapter I, Section 2.F(4) states:

> *Approval of Exemptions for Use of Non-DOE Facilities.*
> *(4) DOE waste shall be treated, stored, and in the case of low-level waste, disposed of at the site where the waste is generated, if practical; or at another DOE facility. If DOE capabilities are not practical or cost effective, exemptions may be approved to allow use of non-DOE facilities for the storage, treatment, or disposal of DOE radioactive waste based on the following requirements:*
> *(a) Such non-DOE facilities shall:*
> *1. Comply with applicable Federal, State, and local requirements;*
> *2. Have the necessary permit(s), license(s), and approval(s) for the specific waste(s); and*
> *3. Be determined by the Field Element Manager [MGR-ORP] to be acceptable based on a review conducted annually by DOE.*
> *(b) Exemptions for the use of non-DOE facilities shall be documented to be cost effective and in the best interest of DOE, including consideration of alternatives for on-site disposal, an alternative DOE site, and available non-DOE facilities; consideration of life-cycle cost and potential liability; and protection of public health and the environment.*
> *(c) DOE waste shall be sufficiently characterized and certified to meet the facility's waste acceptance criteria.*
> *(d) Appropriate National Environmental Policy Act (NEPA) review must be completed. For actions taken under the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA), it is DOE's policy to incorporate NEPA values into the CERCLA documentation.*
> *(e) The Office of Health, Safety and Security, the Office of Environmental Management, and the Program Secretarial Officer shall be notified of the basis for using any non-DOE radioactive waste disposal facility prior to the use of such facilities.*

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[1] Based on a density for cementitious Hanford LAW of 1.77 g/mL and a volume expansion factor of 1.80 m^3/m^3.

(f) Host States and State Compacts where non-DOE facilities are located shall be consulted prior to approval of an exemption to use such facilities and notified prior to shipments being made.

Table 4-1: Long-lived isotope waste classification table applicable to the WCS FWF.
From 30 Texas Administrative Code 336.362 Appendix E, Table 1.

Radionuclide	Class A Limit		Class B Limit		Class C Limit	
14-C	0.80	Ci/m3	NL[1]	Ci/m3	8	Ci/m3
14-C in Activated Metal	8	Ci/m3	NL[1]	Ci/m3	80	Ci/m3
59-Ni in Activated Metal	22	Ci/m3	NL[1]	Ci/m3	220	Ci/m3
94-Nb in Activated Metal	0.02	Ci/m3	NL[1]	Ci/m3	0.20	Ci/m3
99-Tc	0.3	Ci/m3	NL[1]	Ci/m3	3	Ci/m3
129-I	0.008	Ci/m3	NL[1]	Ci/m3	0.08	Ci/m3
Alpha-emitting transuranic nuclides with half-life greater than five (5) years	10	nCi/g	NL[1]	nCi/g	100	nCi/g
241-Pu	350	nCi/g	NL[1]	nCi/g	3,500	nCi/g
242-Cm	2,000	nCi/g	NL[1]	nCi/g	20,000	nCi/g
266-Ra[2]	10	nCi/g	NL[1]	nCi/g	100	nCi/g

[1] There are no limits established for these radionuclides in Class B Wastes

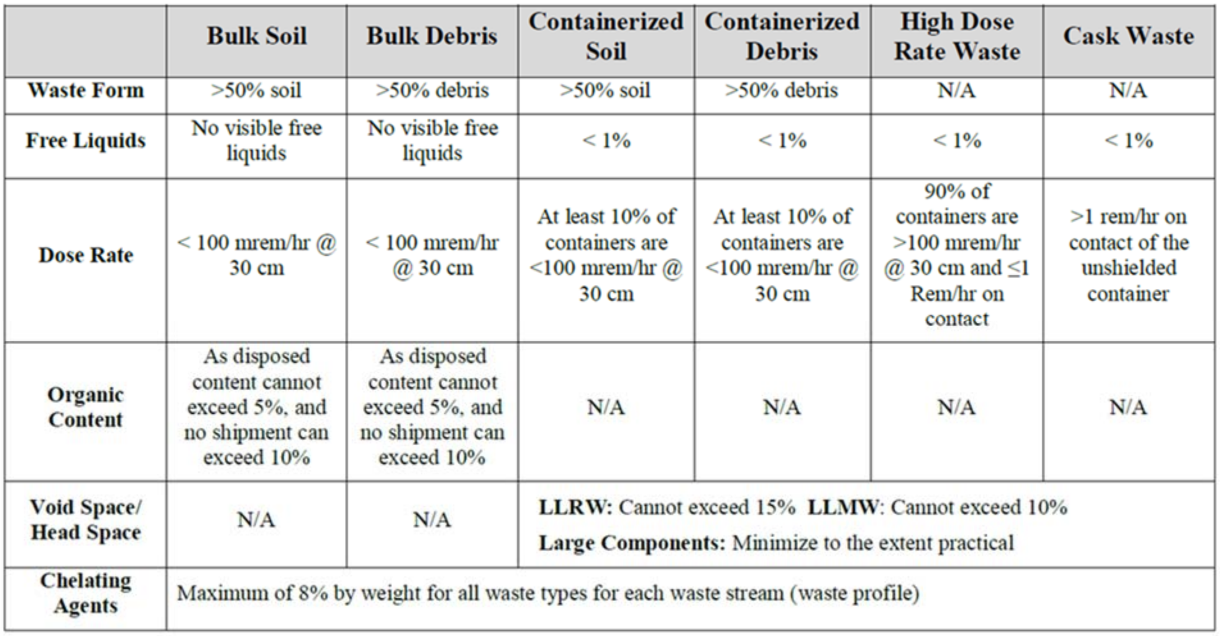
[2] This isotope is not listed in the classification tables in 10 CFR Part 61 but is required by the state of Texas to be included in classification determination

Table 4-2: Short-lived isotope waste classification table applicable to the WCS FWF.
From 30 Texas Administrative Code 336.362 Appendix E, Table 2.

Radionuclide	Class A Limit		Class B Limit		Class C Limit	
Total radionuclides with half-lives less than five (5) years	700	Ci/m^3	NL [1]	Ci/m^3	NL [1]	Ci/m^3
3-H	40	Ci/m^3	NL [1]	Ci/m^3	NL [1]	Ci/m^3
60-Co	700	Ci/m^3	NL [1]	Ci/m^3	NL [1]	Ci/m^3
63-Ni	3.5	Ci/m^3	70	Ci/m^3	700	Ci/m^3
63-Ni in Activated Metal	35	Ci/m^3	700	Ci/m^3	7000	Ci/m^3
90-Sr	0.04	Ci/m^3	150	Ci/m^3	7000	Ci/m^3
137-Cs	1	Ci/m^3	44	Ci/m^3	4600	Ci/m^3

[1] There are no limits established for these radionuclides in Class B or C wastes. Practical considerations such as the effects of external radiation and internal heat generation on transportation, handling, and disposal will limit the concentrations for these wastes. These wastes shall be Class B unless the concentrations of other radionuclides in Table 2 determine the waste Class C independent of these radionuclides.

Table 4-3: Waste stream restrictions from Appendix 2 of the FWF Generator Handbook (WCS 2015).



	Bulk Soil	Bulk Debris	Containerized Soil	Containerized Debris	High Dose Rate Waste	Cask Waste
Waste Form	>50% soil	>50% debris	>50% soil	>50% debris	N/A	N/A
Free Liquids	No visible free liquids	No visible free liquids	< 1%	< 1%	< 1%	< 1%
Dose Rate	< 100 mrem/hr @ 30 cm	< 100 mrem/hr @ 30 cm	At least 10% of containers are <100 mrem/hr @ 30 cm	At least 10% of containers are <100 mrem/hr @ 30 cm	90% of containers are >100 mrem/hr @ 30 cm and ≤1 Rem/hr on contact	>1 rem/hr on contact of the unshielded container
Organic Content	As disposed content cannot exceed 5%, and no shipment can exceed 10%	As disposed content cannot exceed 5%, and no shipment can exceed 10%	N/A	N/A	N/A	N/A
Void Space/ Head Space	N/A	N/A	**LLRW:** Cannot exceed 15% **LLMW**: Cannot exceed 10% **Large Components:** Minimize to the extent practical			
Chelating Agents	Maximum of 8% by weight for all waste types for each waste stream (waste profile)					

5.0 Method for Screening Technologies

A team of subject matter experts from PNNL, SRNL, Vitreous State Laboratory, and WRPS was assembled by the WRPS Chief Technology Office to identify technological and regulatory constraints to treatment and disposal of Hanford SLAW in a cementitious (grouted) waste form, and then to evaluate a wide range of technical solutions to mitigate the constraints. This process was completed in three distinct phases. In Phase I, the team reviewed the chemical and physical characteristics of the most recent projected SLAW feed vector along with FY19 results from PNNL that identified the level of improvement that would be needed to meet onsite disposal requirements in the IDF (Asmussen et al. 2019a). The expert panel relied on this information in group discussions to develop an extensive list of candidate technologies to mitigate the identified limitations.

In Phase II, the expert panel organized the technologies by contaminant (129-I, 99-Tc, NO_3^-, organics, or non-specific) and by implementation methods (pretreatment, modification to waste form chemistry, or engineered barrier) and collected relevant information on each. This information was subsequently discussed and recorded in 13 distinct categories that were chosen to elucidate facts associated with technology maturation needs and impediments to implementation at Hanford. These categories are:

- Technology Description
- Literature References
- Development Level
- Project Cost Impacts
- Project Schedule Impacts
- Likely Technical Limitations
- Impacts to LAW Waste Loading (As Compared To Cast Stone)
- Impacts on Non-Target COCs
- Availability of the Technology
- Long-Term Effectiveness
- Impact to Site Hazard
- Known Regulatory Issues
- Known Stakeholder Issues.

Technology maturation and data needs were documented during the discussions as specific unknowns for each technology. During Phase II, the original list of 86 technologies was reduced to 63 since 23 technologies were deemed to not be applicable to the high salt, high caustic content of LAW.

In the final phase of the process (Phase III) the expert panel selected 7 of the 13 categories for qualitative rating, and a series of meetings were held to discuss and rate each technology in each category. All ratings were developed by consensus. The rated categories along with their qualitative rankings are:

- Development Level (H/M/L)
- Likelihood of NOT INCREASING Cost compared to Cast Stone (H/M/L)
- Ability to be Ready to Process LAW by 2034 (Yes/No)
- Likelihood of NO Technical Limitation that Would Stop Deployment (H/M/L)
- Likelihood of Increasing or Decreasing Waste Loading (Cast Stone Baseline) (+/0/-)
- Likelihood of Improving or Diminishing Retention of Non-targeted COCs (+/0/-)
- Availability of technology or components (H/L).

After the qualitative rating was complete a numerical scoring criterion for each category was applied (Tables 5-1 to 5-4) and a composite score for the individual technologies was calculated. An uncertainty score was also developed for each technology by counting the number of technology maturation unknowns identified during group discussions. Finally, the expert panel reviewed the list of scored technologies and selected a final list of 51 items that were deemed as the most viable for full-scale application at Hanford and deserving of a more detailed written description. These descriptions are provided in Appendix A. It should be noted that a technology warranting a write-up was deemed as feasible or worthy of investigation to improve a cementitious SLAW venture.

Table 5-1: Rankings and associated scoring levels used for the Development Level, Cost Impacts, and Technical Limitations categories.

Ranking	Score for Ranking
Low (L)	0
Medium Low (M/L)	0.5
Medium (M)	1
Medium High (H/M)	1.5
High (H)	2
N/A	0
Unknown	0

Table 5-2: Rankings and associated scoring levels used for the Availability category.

Ranking	Score for Ranking
Low (L)	0
Medium (H/L)	0.5
High (H)	1
N/A	0
Unknown	0

Table 5-3: Rankings and associated scoring levels used for the Schedule Impacts category.

Ranking	Score for Ranking
No	1
Yes	-2
N/A	0
Unknown	0

Table 5-4: Rankings and associated scoring levels used for the waste loading and effects on other COCs categories.

Ranking	Score for Ranking
Increase Waste Loading or Improve PA Performance (+)	1
Reduce Waste Loading or Degrade PA Performance (+)	-1
No impacts (0)	0
Mixed Result on Waste loading or PA Performance (+/-)	0
N/A	0
Unknown	0

6.0 Results

Tables 6-1 through 6-10 provide a summary of the results of the expert panel review of technologies to enable LAW to be solidified into a cemented waste form, disposed in the IDF, and meet the accompanying regulatory requirements. COCs; LDR organics (Table 6-1), 129-I (Tables 6-2, 6-5, and 6-7), 99-Tc (Tables 6-3, 6-6, and 6-8), and nitrate (Table 6-4); which are known to currently challenge IDF regulatory limits were considered separately. In addition, technologies that modify the bulk waste form to simultaneously reduce the release and transport of all COCs were reviewed and evaluated (Tables 6-9 and 6-10). Technologies considered for individual COC were grouped by their method of application:

- a pretreatment process before solidification,
- an additive or waste form modifier to the solidification formulation, or
- an engineered barrier technology that would be applied to the waste package or at the IDF.

This grouping allowed the expert panel to simultaneously compare, contrast, and rank individual treatment approaches within the context of similar treatment options and helped to ensure consistent thought processes were applied. Note that the dark grey rows in each table represent technologies that were originally listed, but were viewed as unviable after initial discussions by the expert panel. The light grey rows represent technologies that were further evaluated and scored, but were deemed of a lower priority when compared to other technologies in a grouping and so did not receive a full written description.

The last two columns in each table provide an overall score and uncertainty level for each technical approach. Score values were generated from the individual ranking criteria using the methods described in Section 5. The "Uncertainty" values represent a summation of the number of major unknown issues identified by the expert panel for a given technical approach. For example, an Uncertainty of 3 would be assigned to an item if; (1) the impact of competing reactions was unknown, (2) the effectiveness of the method in LAW has not been tested, and (3) the long-term effectiveness in buried waste was unknown. It is important to note that these values should only be compared within a particular application grouping (e.g., pretreatment for 129-I or engineered barriers for nitrate). Also, it was the consensus of the expert panel that the scores and uncertainty values should not be the sole consideration as a ranking for possible investments in technology maturation. Rather, they summarize the expert panel's general opinion on how successful technology maturation efforts will be at producing a full-scale technology that meets the intended objective and all listed criteria should be considered in target improvement efforts.

6.1 Pretreatment for Organics

The regulatory requirements for LDR organics are concentration based and must be met in the waste form when it is received at the disposal location, whether onsite or offsite. The expert panel identified only two general approaches that could meet this requirement. The first approach (Table 6-1, entry labeled Line Number 4) relies on direct sampling of the retrieved or in tank waste to identify material that, once solidified, would meet the LDR concentration limits (hereafter termed the Sample and Send approach). It should be noted that WTP assumes volatile organic compounds and a portion of semi-volatile organic compounds volatilize from the cold cap with little or no destruction in the plenum and move downstream to the ETF via the EMF. The resulting influence of a sample and send approach to an integrated flowsheet must be considered. The second approach would rely on a pretreatment process to remove or destroy the LDR organics in the retrieved LAW stream. Three chemical destruction pretreatment process were evaluated (Table 6-1, Line Numbers 5, 6, and 7) along with one electrical pretreatment process (Table 6-

1, Line Number 12). Two removal technologies, use of granular activated carbon (GAC) (Table 6-1, Line Number 13) and stripping by evaporation (Table 6-1, Line Number 14), were also evaluated. The two options which scored the highest by the expert panel (Sample and Send and evaporation to remove organics) also were deemed to have a low uncertainty associated with the technology maturation process.

6.2 Pretreatment for Iodine, Technetium, and Nitrate

Tables 6-2, 6-3, and 6-4 list the results of the evaluation of pretreatment technologies to remove iodine, technetium, and nitrate from the SLAW feed vector, respectively. For iodine, three removal methods (Table 6-2, Line Numbers 4, 5, and 6) and a Sample and Send approach (Table 6-2, Line Number 13) were the technical approaches that received full evaluations. Similarly, for technetium there were three removal methods (Table 6-3, Line Numbers 4, 5, and 6) and a Sample and Send approach (Table 6-3, Line Number 9) that were identified as viable concepts. The disposition pathway for separated technetium and iodine, and the resulting influence on the integrated flowsheet, would need to be considered. The Sample and Send approaches direct the LAW stream to either the vitrification or supplemental treatment systems based on the COC concentration to minimize contaminant release from solidified LAW in the IDF. Sample and Send was the highest scoring pretreatment approach for both iodine and technetium. Again, it should be noted that the impact of a sample and send approach on the integrated flowsheet must be acknowledged. There were no viable pretreatment technologies identified by the expert panel for nitrate in LAW due to the aggressive waste environment that would render commercial nitrate removal technologies (designed for drinking water) ineffective

6.3 Waste Form Modifications for Iodine, Technetium, and Nitrate

Four approaches to enhancing iodine retention by adding modifiers to the cementitious waste formulation were evaluated (Table 6-5). It should be noted that methods that rely on chemical properties of iodine to remove or retain 129-I must be scaled appropriately to address both 127-I and 129-I present in the waste since the chemical properties of the two isotopes are identical. Three of these approaches (Line Numbers 15, 16, and 18) use a solid material that has iodine adsorptive properties. The fourth method suggests the addition of soluble materials that will form precipitates with iodine. The use of a strong base anion resin as an adsorption additive and the addition of soluble species to facilitate iodine precipitation were the highest scored option, although both also have significant uncertainties.

Of the five waste form modifications identified for technetium, three rely on reduction of Tc(VII) to Tc(IV) and the formation of insoluble Tc(IV) species in the solidified waste (Table 6-6, Line Numbers 11, 12, and 13). The remaining two methods rely on an additive (iron oxyhydroxides) to precipitate 99-Tc in either oxidation state (Line Number 14), or use an ion exchange resin to bind 99-Tc (Line Number 15). The two highest scoring technical approaches are both associated with enhancing the reducing capacity of the waste form.

Line 29 of Table 6-6 does not represent a technology that directly addresses technetium removal or retention within the waste form. Rather, Line 29 introduces a fundamental issue for many of the technologies that address technetium; the fact that not all technetium in the tank waste is present as pertechnetate (TcO_4^-) or Tc(IV), but some is thought to be present as Tc(I) primarily in the AN and SY Tank Farms (Serne et al. 2014). If a significant fraction is in this reduced Tc(I) state then technologies designed to reduce Tc(VII) to Tc(IV) and capture the Tc(IV) in the cementitious matrix may be ineffective, at least for the fraction of 99-Tc that is present as Tc(I). It is unknown if the REDOX technologies would impart any resistance to oxidation of the Tc(I) to Tc(VII). The technology description in Appendix A for Line 29 provides a roadmap to ascertain the impact this issue will have on 99-Tc retention efforts.

Waste form modifications identified to aid retention of nitrate are listed in the middle portion of Table 6-4. The two techniques identified are the addition of a carbon-based getter which would see large competition from other anions (Line Number 11) and the use of layered double hydroxides to incorporate nitrate (Line Number 14). Based on the numeric values for the scores and uncertainties, both methods were assessed as have similar technology maturation needs and associated uncertainty.

6.4 Engineered Barriers for Iodine, Technetium, and Nitrate

Tables 6-7, 6-8, and the lower section of Table 6-4 provide details on the contaminant specific engineered barrier technologies assessed for iodine, technetium, and nitrate, respectively. Barrier technologies that affect bulk transport properties of the disposal facility are discussed in Section 6.6. The contaminant specific methods can be grouped into the following three general categories:

- Those that modify individual waste packages (Table 6-7 Line Number 29 and Table 6-8 Line Number 19),
- Those that modify the backfill (Table 6-7 Line Numbers 21, 22, 24, and 30; Table 6-8 Line Numbers 17, 18; and Table 6-4 Line Number 16), and
- Those that create a reactive barrier in the vadose or saturated zone below the IDF (Table 6-7 Line Numbers 25, 26, and 31; Table 6-8 Line Number 26; and Table 6-4 Line Number 18).

The in-package technologies describe placing a barrier grout in the waste package that would surround the waste grout. The barrier grout could contain an amendment that reacts with the contaminants migrating out of the waste grout and slow transport. Specific amendments for each contaminant of concern could be selected from those suggested as adsorptive/reactive treatments in the pretreatment and waste form modification sections.

The suggested COC specific amendments to the backfill material around buried waste packages rely on physical/chemical interactions to remove soluble forms of contaminates from the water percolating through the disposal facility. GAC is suggested for both iodine (Table 6-7 Line Number 24) and technetium (Table 6-8 Line Number 18). It should be noted that GAC may also have some impact on nitrate (Table 6-4 Line Number 11). Various forms of iron may also retard the migration of soluble iodine (Table 6-7 Line Number 30), technetium (Table 6-8 Line Number 17), and nitrate (Table 6-4 Line Number 16). Finally, it is suggested that a naturally occurring or synthetic clay can be functionalized to enhance stability and iodine capture (Table 6-7, Line Number 22).

Five permeable barrier technologies are included in the analysis for iodine (three methods; Table 6-7 Line Numbers 25, 26, and 31), technetium (one method, Table 6-8 Line Number 26), and nitrate (one method Table 6-4 Line Number 18). Two of the iodine techniques rely on injection of AgCl into a permeable subsurface formation to create a zone were iodine can react to form insoluble AgI. It should be noted Ag brings an associated RCRA limitation. The third iodine method requires the injection of gaseous CO_2 to co-precipitate iodate with carbonates. The technetium and nitrate reactive barriers also rely on introducing chemicals into the subsurface to create a region where Tc(VII) is reduced to Tc(IV), or where nitrate reacts biologically to produce N_2 (denitrification).

The contaminant specific engineered barriers that were scored highest by the expert panel were the use of an iron-amended backfill to provide a reduction/sorption zone for nitrate and technetium and injection of dissolved or colloidal AgCl to form a permeable reaction zone for conversion of soluble iodine to insoluble AgI.

6.5 Formulation Modifications to Alter Bulk Properties

Table 6-9 summarizes the modification approaches proposed to the waste formulation to alter bulk transport properties. Three of the technical approaches attempt to reduce the porosity of the material by either reducing the water-to-cement ratio (Line Numbers 4 and 17) or enhancing particle packing (Line Number 5). The remaining four technical approaches propose different formulation development efforts to identify mixtures that enhance contaminant retention (Line Numbers 6, 8, 9, and 15). The highest scoring items were both formulation development approaches (Line Numbers 6 and 8). As described in Section 1.4 and page 79, the test effort defining the Cast Stone formulation was both limited in extent and heavily influenced by the nominal SRS Saltstone formulation. From a current perspective, the heavy influence of the Saltstone formulation on Cast Stone is less necessary in view of the fact that Saltstone was developed to be pumpable over large distances into vaults, whereas the Cast Stone is intended for an in-container grouting approach. While the properties of LAW Cast Stone measured to date largely meet expected WAC or LDR requirements, the formulation constraints with respect to properties such as rheology, mixing, and workability are quite different. This difference leaves considerable room for further optimization. There is therefore a substantial opportunity for optimization of this formulation to improve its performance and also for the application of new matrices, such as geopolymers. These concepts are captured in Line Numbers 6 and 8.

6.6 Engineered Barriers to Alter Bulk Properties

Four barrier technologies were assessed and scored. Two are associated with the benefits obtained by increasing the physical size of the waste package (Table 6-10 Line Numbers 19 and 20) and two are aligned with reducing water infiltration into the disposal facility (Table 6-10 Line Numbers 21 and 22). The benefits targeted by increasing waste package size are a reduction in surface area to volume ratio, which is linearly proportional to diffusive release rate (Line Number 19), and enhanced technetium retention from reduced diffusion of oxygen into the waste form [also proportional to surface area] (Line Number 20). The water infiltration methods assessed were the use of a low-density cementitious backfill around the emplaced waste packages (Line Number 21) and the addition of a high-density polyethylene (HDPE) cap (Line Number 22). Use of a low-density cementitious backfill (Line Number 21) scored far below the other three and has much higher uncertainty. Use of an HDPE cap was the highest rated technology in this group and was also assigned a low uncertainty value.

Table 6-1: Pretreatment technologies considered for organics.

Note: Dark grey rows indicate technologies the expert panel did not rank and the light grey rows indicate technologies that were ranked but not considered viable enough to warrant a detailed written description.

Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
4	Low LDR organic inventory tanks selected specifically for SLAW grout	Use direct tank sampling and/or process knowledge to select tanks that do not require organic removal or treatment.	H Tank Sampling approaches available and ability to measure organics in tank samples	M Tank sampling is a lower cost activity	**NO** Could be implemented by 2034; 3 yrs	M Would require sampling strategy and diversion of certain tanks to SLAW. Relies on TSCR type unit and a separate feed tank (part of baseline process); Number of applicable tanks is unknown.	(0) Does not increase or decrease waste loading.	N/A	H Protocols already defined for tank sampling and analysis	6	1
5	Chemical Destruction	Use an alkaline permanganate strike for the organics then quench the residual permanganate with sulfite. The adjusted LAW would be slurried and then disposed as a grout	M Commercial treatment; Effectiveness in LAW is unknown; Would need demonstration with LAW; Full Scale for non-radiological use	M/L Dependent on flowsheet and unit operations; Could be in mixing vessel to minimize requirements	**NO** Could be implemented by 2034; 14 yrs	M Impacts of precipitated Mn-oxides unknown. Some organics not destroyed e.g. biphenyl; Slight increase in grout vol compared to current Cast Stone; Nonregulatory organics and NO2 will compete and increase oxidation requirements.	(-) Slight increase in grout volume (i.e. decreased waste loading) compared to current Cast Stone.	(0) Needs to demonstrate with LAW for effect on other constituents; Does not oxidize NO2 May increase Tc mobility by converting Tc+4 to Tc+7	H Commercially Available	3.5	2
6		Use a hypochlorite strike for the organics then then quench the residual hypochlorite with sulfite. The adjusted LAW would be slurried and then disposed as a grout	H Commercial treatment; Effectiveness in LAW is unknown; Would need demonstration with LAW	M/L Dependent on flowsheet and unit operations; Impacts of high nitrite on amount of reagent needed is unknown Could be in mixing vessel to minimize requirements	**NO** Could be implemented by 2034; 14 yrs	M Unknown impacts on organics; Successful at converting non-VII Tc to Tc-VII; Nonregulatory organics and NO2 will compete and increase oxidation requirements; May make chlorinated hydrocarbons.	(-) Slight increase in grout volume (i.e. decreased waste loading) compared to current Cast Stone.	(0) Needs to demonstrate with LAW for effect on other constituents; May increase Tc mobility by converting Tc+4 to Tc+7	H Commercially Available	4.5	3
7		Use of ozone to oxidize organics then grout the resulting waste stream.	H Has been done on Hanford tank waste. Effectiveness for all LDR organics is unknown.	M Require additional flowsheet step	**YES**; May take longer than 2034; >14 yrs	M Nonregulatory organics and NO2 will compete and increase oxidation requirements.	(0) Does not increase or decrease waste loading.	(+) May drive off iodine, but can leverage older test data	H Commercially Available	4	1
8	Thermal Destruction	Wet air oxidation followed by grouting the residual solids. Treatment of off-gas and resulting secondary waste would also be necessary.	---	---	---	---	---	---	---	---	---
9		Fluidized Bed Steam Reforming (FBSR) followed by grouting the residual solids. Treatment of off-gas and resulting secondary waste would also be necessary.	TRL 8 to 9 Being implemented at INL	---	---	---	---	---	---	---	---
10		Calcining followed by grouting the residual solids. Treatment of off-gas and resulting secondary waste would also be necessary.	Demonstrated on acidic rad waste. Improvements required	---	---	---	---	---	---	---	---
11	Irradiative Destruction	UV Exposure / treatment	---	---	---	---	---	---	---	---	---
12		Electron Beam Exposure	L Unproven approach in LAW at this time.	M Would need high power inputs and large irradiation contact	**NO** Could be implemented by 2034; 6 yrs	M/L Unknown in salt wastes	(0) Does not increase or decrease waste loading.	Unknown	H Commercially Available	3.5	2
13	GAC	Granular activated carbon for sequestration of nonpolar organics in LAW. Then grout the resulting waste stream	L Unproven approach in LAW at this time (i.e. Unknown); There are questions on the ability of GAC to treat polar LRD organics. The ability to directly dispose of the GAC may be limited and an additional treatment method such as organic destruction might be required.	M Require additional flowsheet step	**NO** Could be implemented by 2034; 6 yrs	L Only works for non-polar organics; Overall effectiveness to meet required removal levels is unknown	(0) Does not increase or decrease waste loading.	(+); May absorb other COCs such as Tc, Iodine, and RCRA metals, but this is unknown.	H Commercially Available	4	3
14	Evaporation to Steam Strip Organics	Steam strip organic components of regulatory concern using an evaporator. Condensate goes to ETF. Then grout the resulting bottoms waste stream.	H Proven technology with tank waste supernatant.	H Will probably do it anyway	**NO** Could be implemented by 2034; 3 yrs	H Of the identified LDR organics, the relative volitilitites indicate adquate sparation; Overall effectiveness to meet required removal levels is unknown	(+) Concentrates the waste stream and so will reduce overall immobilized waste volume.	(0)	H Commercially Available; Could rely on 242A or a new, skid mounted unit.	9	1

Table 6-2: Pretreatment technologies considered for iodine.
Note: Dark grey rows indicate technologies the expert panel did not rank and the light grey rows indicate technologies that were ranked but not considered viable enough to warrant a detailed written description.

Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
4	Ag based material	Iodine would be captured on a Ag-based material from filtered and ion exchanged LAW. Media would be disposed as secondary waste. The system would require ion exchange columns. Example candidate: Ag-mordenite The resulting iodine loaded waste form could be a directly densified material as demonstrated for material used for iodine vapor capture. However, Ag is a RCRA metal	M/L Demonstrated iodide capture in lab. Commercially available. Iodine Stabilization with Ag known, substrate and process needs to be identified; Materials degraded quickly; Minimal testing with highly alkaline waste.	M Would require ion exchange columns; Silver is expensive reagent	**NO** Schedule Impacts; Could be implemented by 2034; 14 yrs	M Long term performance in alkaline system unknown; Stability of solid substrate unknown; Removal efficiency of iodate unknown;	(0) Does not increase or decrease waste loading.	(0) Lowers potential iodine release; Potential to allow reduction chemistry; Adds Ag which is a RCRA metal	H Commercially available; Sorbent unknown	4.5	2
5	Superlig 639 Elutable Resin	Iodine would be captured on a SuperLig 639 from filtered and ion exchanged LAW. The system would require ion exchange columns. The resulting iodine column could be immobilized in grout or solidified with polymer encapsulation for disposal on/off-site.	M/L Components commercially available; testing indicated low DF for I; Resin needs improvement.	M Would require ion exchange columns; Resin costly but reused multiple times	**NO** Schedule Impacts; Could be implemented by 2034; 14 yrs	L Unlikely to be viable for I in current form; tested w/SRS tank waste and exhibited poor DF; unknown if it removes both Iodine and iodate. New Liquid waste stream with no defined pathway for disposal.	(0) Does not increase or decrease waste loading.	(0) Lowers potential iodine release; Potential to allow reduction chemistry; Removes Tc from LAW	H Commercially available, Media unknown; Resin is produced by sole source vendor	3.5	2
6	Strong base anion exchange resins	Iodine would be captured on a strong base anion exchange resin (e.g. Purolite A530E) from filtered and ion exchanged LAW. The system would require ion exchange columns. The resulting iodine column could be immobilized in grout or solidified with polymer encapsulation for disposal on/off-site.	M Components commercially available; Resin unknown, some may not be stable in high pH (Duncan, et al., LAB-RPT-12-00002, rev. 1); Resin is marginally effective for Iodine.	M Would require ion exchange columns; Resin costly but reused multiple times	**NO** Schedule Impacts; Could be implemented by 2034; 14 yrs	M Evidence of iodine removal of purolite A530E exists; A530E is not stable in high pH environments; Other resins may be stable at high pH and may be elutable.	(0) Does not increase or decrease waste loading.	(+) Lowers potential iodine release; Potential to allow reduction chemistry; Removes Tc from LAW; Effect on some COCs unknown	H Commercially available, Resin unknown	6	3
7	Oxidation of Aqueous Iodine species to I2 and Separation of I2 by Absorption	Chemically oxidize the aqueous iodine species to I2 and then adsorb the evolved I2 on a solid media.	L Need to workout oxidation chemistry in LAW; Oxidizer unknown	H/L Many unknowns Add two new unit operations, but adsorption process is known.	Unknown schedule Impacts;	M Will alter the waste stream; Materials of construction needs to be considered; Conversion in LAW is unknown; Completing reactions need to be consider.	(0) Does not increase or decrease waste loading.	+/- May destroy organics; Convert nonpertechnetate to pertechnetate;	L Likely commercial available; Materials of construction need to be determined.	3	4
8	Mercurex	Acidic treatment that uses Hg(NO3)2 in a column to capture iodine.	---	---	---	---	---	---	---	---	---
9	Alkaline solution with Strike (Ag/Bi/Ba)	Involves the addition of a cation species (dissolved rather than on a solid substrate) to filtered, ion exchanged LAW to specifically precipitate out low solubility phases. Could use Ag/Bi/Ba/Pb and then either remove the precipitate, or directly included into the grout.	L Unknown effectiveness; Need experimental demonstration with LAW	M Adds two new unit operations, but adsorption process is known.	**NO** Schedule Impacts; Could be implemented by 2034; 14 yrs	M Limitations are mainly capacity and interference from other halides/anions; Removal efficiency of iodate unknown; Some reagents are RCRA regulated.	(0) Does not increase or decrease waste loading; TCPL impacts are unknown	Unknown; May add RCRA Metals.	L Most compounds are commercially available in large quantities; Some reagents are unidentified	3	4
10	Electrolytic Scrub	Acidic treatment with Co(3+) oxidant to target iodine removal.	---	---	---	---	---	---	---	---	---
11	IODOX	Acidic treatment with an HNO3 scrubber to sequester iodine species.	---	---	---	---	---	---	---	---	---
12	Oxidation of Aqueous Iodine species to I2 and Separation of I2 by Evaporation	Use chemical treatment **and evaporation** to generate I2 which will volatilize the I2 for gas phase capture.	L Unknown effectiveness; Need to workout oxidation chemistry in LAW.	H/L Many unknowns Add two new unit operations, but adsorption process is known.	Unknown schedule Impacts;	M Will alter the waste stream; Materials of construction needs to be considered; Conversion in LAW unknown; Completing reactions need to be consider	(-) Would require the addition of large quantities of acid which will increase waste volume and thus decrease waste loading.	(+/-) May destroy organics; Convert nonpertechnetate to pertechnetate;	L Likely commercial available; Materials of construction need to be determined. Some reagents are unidentified.	2	4
13	Low iodine inventory tanks selected specifically for SLAW grout	Many tanks at Hanford have low iodine inventories. These would be ideal candidates for SLAW grout (more so if also low Tc and organics). Can use direct knowledge of tank inventories from sampling and historical knowledge to identify low iodine containing tanks/farms as candidates for SLAW grout. High iodine tanks would be sent to WTP.	H Tank Sampling events are regular and efforts are on-going for iodine speciation in tanks.	H No additional costs to identify candidate tanks; TSCR Like units are part of the full mission baseline cost.	**NO** Schedule Impacts; Could be implemented by 2034: 3yrs	M Would require sampling strategy and identification of tanks as candidates for grout; Would strengthen justification for low-risk tank to go to grout; Number of applicable tanks is unknown.	(0) Does not increase or decrease waste loading.	N/A	H Protocols already defined for tank sampling and analysis	7	1

Table 6-3: Pretreatment technologies considered for technetium.

Note: Dark grey rows indicate technologies the expert panel did not rank and the light grey rows indicate technologies that were ranked but not considered viable enough to warrant a detailed written description.

Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
4	Elutable Ion Exchange/Sorbent Molecular Recognition Technology (MRT)	Use of an elutable sorbent/ion exchange resin (i.e. an MRT) containing column to strip Tc out of an LAW stream. Was considered as a previous Tc-removal unit operation in the Hanford flowsheet. Commercially available materials can strip pertechnetate. The eluted Tc (warm water) can be disposed of separately or sent to LAW or HLW melter for vitrification in borosilicate glass final waste form.	H/M Can use commercially available materials and leverage prior Tc removal configurations from WTP or TSCR-style system. Lacking manufacturing scale up.	M Would require treatment system to be added to flowsheet. Resin is reusable.	**NO** Could be implemented by 2034; 3 yrs	M Capacity/breakthrough of sorbents/resins, would require demonstration in TSCR-style system. May be viable for non-organic tanks/farms. Unknown disposal pathway for resin	(0) Does not impact waste loading in grout but would lower Tc inventory. Requires disposal of spent resin after elution	(0) Specific to Tc but no adverse effects identified in previous testing campaigns.	H Commercially available resins but would need some scale up production. Sole source supplier.	5.5	1
5	Strong Base Anion Exchange Resin (SBAR)	Use of a SBAR containing column to strip Tc out of the LAW stream. Was considered as a previous Tc-removal unit operation in the Hanford flowsheet. Commercially available materials can strip pertechnetate. The eluted Tc (nitric acid) can be disposed of separately or sent to LAW or HLW melter for vitrification in borosilicate glass final waste form. Or loaded resin could be directly disposed of.	L Limited demonstration in LAW as a full system approach (i.e. is an unknown). Need economic analysis of single use resin vs. elution.	M Resin relatively inexpensive but complex system	**NO** Could be implemented by 2034; 6 yrs	M Only removes pertechnetate, Kds for SBARs are low (~270 mL/g). Other work suggest quaternary amine resins are degraded by salt solutions	(0) Does not impact waste loading in grout but would lower Tc inventory. Requires disposal of spent resin	(+) May also remove iodide.	H Commercially available resins but would need some scale up production. Sole source supplier.	5	1
6	Redox Strike/Co-precipitation/Filtration/Grout or otherwise dispose as secondary waste	Precipitation of Tc through the addition of a redox active species (e.g. Sn(II) or sulfide). The precipitated Tc could then be filtered out of the feed, or kept in to blend into final grout mix.	M Engineering scale demonstration could be performed based on lab data Sn is ineffective in alkaline, sulfides are active.	M Filtration and collection of product would be an added cost	**NO** Could be implemented by 2034; 14 yrs	M Competition by other constituents is unknown (e.g. Cr) Collection of precipitated Tc Only removes Tc(VII)	(0) Does not impact waste loading in grout but would lower Tc inventory	(+) Unknown if strike would change base properties of grout Process would also precipitate Cr and Hg. Waste form would also need to stabilize these phases	H Use simple, commercially available reagents	6	2
7	Precipitation of pertechnetate with tetraphenylphosphonium (TPP) ion	Precipitate Tc(VII) from LAW with tetraphenyl phosphonium ion. Collect precipitate and move off-site or dispose of separately.	L Limited lab demonstration in LAW (i.e. effectiveness is unknown)	M Would require additional unit operations and TPP destruction	**NO** Could be implemented by 2034; 14 yrs	L Moderate decontamination factor (DF); expected residual TPP in LAW to be problematic; need additional TPP destruction and disposal process; Stability of TPP is unknown.	(-) Does not impact waste loading in grout but requires additional waste to be disposed of	(-) Require additional organic treatment/disposal	L Chemicals are commercially available, supporting infrastructure unknown	0	3
8	Electrolytic Reduction	Using electrochemical processes to remove Tc species through reduction to a final product of Tc metal/TcO2 on the electrode surface. The Tc metal could then be used or converted to a metallic waste form.	L Would need to scale up process based on prior lab data. More studies for process efficiency are needed (i.e. is an unknown)	L Would require a entirely new system .	**Yes** Likely not available until past 2034.	M DF of 19 reported in actual tank waste, may need to be higher and cost/reward not sufficient Competition by other constituents is unknown (e.g. Cr) Electrode materials not optimized Can look to electrochemical recovery process in DOE:NE program at INL Improved efficiency requires organic removal	(-) Increases waste volume due to caustic addition needed to destroy organics.	(0) Unknown if process would change other contaminants, likely to reduce Cr as well.	H Flow cells are used in industry along with simple electrodes (Pt/Ti)	-1	3
9	Low Tc inventory tanks or low-organic complexant tanks selected specifically for SLAW grout	Many tanks at Hanford have low Tc inventories. These would be ideal candidates for SLAW grout (more so if low in both iodine and organics). Can use direct knowledge of tank inventories from sampling and historical knowledge to identify low Tc containing tanks/farms as candidates for SLAW grout. High Tc/complexant tanks would be sent to WTP.	H Tank sampling events are regular and previous efforts have furthered the understanding of Tc in tanks.	H Would require sampling strategy and identification of tanks as candidates for grout. TSCR Like units are part of the full mission baseline cost. Would strengthen justification for low-risk tank to go to grout.	**NO** Could be implemented by 2034; 6 yrs	H Protocols already defined for tank sampling and analysis; Number of applicable tanks is unknown.	(0) Does not increase or decrease waste loading.	N/A	H No additional costs to identify candidate tanks	8	1

Table 6-4: Pretreatment, waste form chemistry modifications, and engineered barrier technologies considered for nitrate.

Note: Dark grey rows indicate technologies the expert panel did not rank and the light grey rows indicate technologies that were ranked but not considered viable enough to warrant a detailed written description.

Mode	Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
Pretreatment	*4*	Nitrogen to Ammonia and Ceramic (NAC) Process	Chemical reduction of nitrate to ammonia through reaction with Al metal. Ammonia tolerant grout could then be used to immobilize resulting waste stream. Need high amounts of Al added to LAW.	M Used industrially but limited demonstration in high salt wastes. Works in alkaline environments. 1990's development for Hanford	M Costs would be higher to develop flowsheet unit operations for process.	**Yes** Would impact 2034 schedule start; 14 yrs	M/L Unknown if the process would work in high salt wastes. Unknown impact of Al additions and resulting ammonia management.	(0) Does not decrease grout volume but decreases NO3 levels	Unknown but pretreatment will alter waste stream and potentially impact known waste form properties. Heat of process may drive off some organics	H Industrially used	1.5	2
	5	Biological Denitrification	Addition of microbial species to LAW to destroy nitrates. Used at Y12 for nitrate wastes in fluidized bed reactor.	---	---	---	---	---	---	---	---	---
	6	Chemical reduction	Use of organics (formic acid, formaldehyde) to convert nitrate to nitrogen, Used by CEA and Tokai-Mura	M Used internationally; Applicability in alkaline waste is unknown;	M Requires additional treatment step.	**Yes** Would impact 2034 schedule start; 14 yrs	L Would it work in high salt wastes and handling of the newly added organics	(0) Does not decrease grout volume but decreases NO3 levels	(+/-) Effects unknown May complex with other contaminants	H Chemicals commercially available	1	2
	7	Electrochemical Destruction	Electrochemical destruction of nitrate/nitrite in flow cell or loop.	M Demonstrated in actual waste	L Much higher costs	**Yes** Would impact 2034 schedule start; 14 yrs	M Ability to meet removal requirements is unknown; Selective removal of nitrate vs impacting other contaminants Cathode fouling by Cr observed previously.	(-) Increases grout volume (i.e. reduces waste loading).	(+) Effects unknown May alter oxidation states of other components at anode (Tc, Hg, Cr) and destroy organics	H Parts would be commercially available	1	2
	8	Calcining/Hydrothermal	Use calcining to convert liquid waste to a granular product that will evolve the nitrate as Nox.	---	---	---	---	---	---	---	---	---
	9	Sorbent columns	Use of a sorbent column to extract nitrate from LAW	---	---	---	---	---	---	---	---	---
Waste Form Chemistry	*11*	Carbon based getters	Sorption of NO3 is observed to carbon based materials. Only need small "Kd" to meet nitrate acceptance levels. Unknown if sorbent would work in grout if sorbent was present in large quantities.	L Sorption of nitrate only shown in water. Effectiveness in LAW is unknown.	M Although low cost, carbon materials likely required in high amounts and could increase costs.	**NO** Could be implemented by 2034; 6 yrs	L Competition in LAW and grout pore water with other anions; SRNL has observed carbon addition can cause ammonia generation	Unknown A large quantity of carbon may be needed and this would increase waste loading.	(+) If sorbent works for anions may also slow release of I/Tc etc.	L Some carbons commercially available but ones used here likely synthesized	3	2
	12	Natural Sorbents (Clays and Zeolites)	Sorption of NO3 observed to natural materials Only need small "Kd" to meet nitrate acceptance levels Unknown in grout systems	---	---	---	---	---	---	---	---	---
	13	Use of high additions of fly/coal ash to the waste form in hopes of increasing Kd of nitrate to the grout matrix.	Sorption of NO3 is observed to industrial materials (e.g. ashes). Only need small "Kd" to meet nitrate acceptance levels. Unknown in grout systems.	---	---	---	---	---	---	---	---	---
	14	Layered Double Hydroxides (LDH)	Immobilize NO3 in LDHs. LDH can be added to grout or generated in grout matrix.	L Only shown in water; Effectiveness with LAW is unknown	H/M M - if synthesized material added H - if minerals generated in grout	**NO** Could be implemented by 2034; 6 yrs	L Effectiveness is unknown; There may be competition in LAW and grout pore water with other anions	Unknown If large amount of LDH is needed to be added to the grout then waste loading may decrease, or would have no effect on waste loading if small amount of LDH added	(+) If LDH works for anions may also slow release of I/Tc etc.	L Likely development required	3.5	3
Engineered Barriers	*16*	Fe amended Backfill in IDF	The use of zero valent iron provides a reduction/sorption zone for nitrate in the near field around waste package. The technology is already being developed by the DOE-RL DVZ program	M Could move lab results to field scale test	H Fe source can be low cost	**NO** Could be implemented by 2034; 6 yrs	M Effectiveness is unknown; Retention mechanism is not understood; Permeability of barrier/water velocity/reactivity needs to be understood for proper design	N/A	(+) Could retard the movement of other contaminants (iodine/iodate/Tc/Cr)	H Fe sources available	7	1
	17	Pump and treat	Use of pump and treat facility at IDF to manage NO3	---	---	---	---	---	---	---	---	---
	18	Gas phase bioremediation using pentane and butyrate	Create a reactive barrier for denitrification. The use of gas-phase carbon amendments has been successfully demonstrated for degradation of perchlorate and nitrate in the vadose zone as part of the ESTCP program (DOD, 2010). Based on the ESTCP demonstration results and preliminary laboratory testing (PNNL-28055), gas-phase bioremediation may be an effective treatment to reduce the mass of nitrate.	L, Effectiveness will be measured during a treatability study for Hanford vadose zone	M	**NO** Could be implemented by 2034; 14 years Technology will be tested as part of the DV-1 OU Laboratory Treatability Study in the FY21-22 timeframe (depending on funding)	L Length of activity is unknown Effectiveness of the technology will be tested as part of the DV-1 OU Laboratory Treatability Study where the technology is considered for saturated conditions at perched water or a water table PRB application beneath the BY Cribs.	N/A	(0) Unknown Treatability testing will include other COCs and their effects	H Used before	3	2

Table 6-5: Modifications to waste form chemistry for iodine.
Note: Dark grey rows indicate technologies the expert panel did not rank.

Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
15	Ag on Inorganic Substrate added to solidification reagents (getters)	This getter treatment would lead to precipitation of iodine as AgI or AgIO3. When the Ag is present on solid sorbents capture/retention appears to be improved. Demonstrated removal success in LAW. Requires higher amounts of getter for success (~5 wt%), but performance threshold for getter amount is unknown. Getter may be blended into dry mix or added as a first treatment step before incorporation into grout.	M Demonstrated iodide capture in lab using simulated LAW and adjusted tank 50 sample. Demonstrated improved grout performance well below required level at 5wt% getter addition Commercially available; PNNL-28545 and 26443 are only secondary waste.	M/L Ag-based materials increase cost of dry mix at current demonstrated level of 5 wt%. Although lower limit of successful iodine retention is unknown	**NO** Schedule Impacts; Could be implemented by 2034; 6 yrs	M Long term stability of AgI upon disposal is unknown; Removal efficiency of iodate not as well studied as iodide; Ag is RCRA regulated;. Substrate stability not as important in this case; May reduce the retention of redox sensitive RCRA Metals but could be balanced with increased slag addition	(0) Does not increase or decrease waste loading.	(-) Can impact reduction capacity Slag interference on AgI at low Ag concentration in waste form	H Commercially available	3.5	2
16	Non-RCRA Reactants on particulate (Getters)	As Ag is a RCRA metal, its release would also need to be managed. Other cations can lead to precipitation of low solubility iodine phases. Pb/Ba previously proposed but are also RCRA. Low soluble iodides/iodates exist for non-RCRA species, e.g. Bi, Ce Can be impregnated into solid structure similar to Ag.	L Demonstrated iodide capture in lab in other solutions (e.g. groundwater) Commercially available. Effectiveness in LAW Unknown and needs demonstration.	M/L Materials may have associated scale up cost but would be lower than Ag	**NO** Schedule Impacts; Could be implemented by 2034; 6 yrs	M/L Long term stability of precipitated product upon disposal; Solubility of Pb and Ba in waste	(0) Does not increase or decrease waste loading.	Unknown Likely less than Ag	L Reagents are undefined so unknown.	2	3
17	Graphene	Sorbent based approach in waste form with graphene or other carbon source added as graphene with affinity for iodine in vapor phase.	---	---	---	---	---	---	---	---	---
18	Ion exchange getter using strong base anion resins	Commercial resins have shown ability to adsorb iodine species (e.g. purolite). Unknown if the iodine-loaded **resin** would be added to waste stream first or included into dry mix. Resin stability in grout is unknown.	M/L Demonstrated iodide capture in lab in other solutions (e.g. ground water) Commercially available; Some resins were unstable in other testing. Need demonstration in LAW, effectiveness unknown.	H Cost of resin would be marginally higher compared to overall mission cost for SLAW grout; Unknown waste loading so quantity of resin not known	**NO** Could be implemented by 2034; 6 yrs	M Long term stability of resin loaded product in grout is unknown; Insufficient selectivity	(0) Does not increase or decrease waste loading.	(+) Positive impact on Tc would be likely if resin is stable in grout;	H Commercially available	6.5	3
19	Addition of chemicals to the waste/dry mix to provide cations that can precipitate low solubility iodine phases. Incorporated directly to the grout	Many cations can lead to precipitation of low solubility iodine phases. Ag/Pb/Ba previously proposed but are also RCRA metals. Low soluble iodides/iodates exist for non-RCRA species. The cation would be added in a soluble form to the waste prior to grout fabrication	L No testing. No demonstration. Stability unknown; Need to confirm RCRA metal stability	H/M Lower cost than modified sorbents	**NO** schedule impacts Could be implemented by 2034; 6 yrs	M Limitations are mainly capacity and interference from other halides/anions; Removal efficiency of iodate unknown; Some reagents are RCRA regulated. Ag may be unstable in slag containing systems	(0) Does not increase or decrease waste loading; TCPL impacts are unknown	Unknown	H Commercially available	5.5	4

Table 6-6: Modifications to the waste form chemistry considered for technetium.

Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
11	Chemical reduction of Tc(VII) by slag glass containing S in caustic solution	Reduction of Tc(VII) to Tc(IV)-O or Tc(IV)S by S containing Slag glass	H	H	**NO**	M Long term performance of reducing conditions in IDF unknown	(0)	(0)	H	7	1
12	Redox Getters	Getters would be added directly to the waste stream or into the dry ingredients to target reduction of Tc(VII) to Tc(IV)-O or Tc(IV)S. Candidate materials include redox active cations (e.g. Fe, Sn) or non-metals (e.g. sulfide, amino)	M Could do a scaled demonstration if required, but optimal getter loadings unknown	M/L Depends on materials: Some getters are simple materials, others have demonstrated scale up production, but some are lab only	**NO** Schedule Impacts; Could be implemented by 2034; 6 yrs	M Unknown long-term stability, Unknown optimal loading and addition sequence Some Sn-getters lose efficiency in LAW, but others are effective.	(0) Does not increase or decrease waste loading. Could decrease waste loading if the amount of getter is a significant volume fraction.	(0) Can interfere with getters targeting iodine (Asmussen Wmsym et al. 2016) Can also improve Cr	Unknown Some getters are cheap/commercially available (e.g. Fe, or sulfur impregnated GAC), other require synthesis	3.5	3
13	Reduction Capacity Control in Waste Form	Control Tc release (and Cr) through bulk redox control in the waste form by increasing overall reduction capacity. Would also slow the oxidation of the waste form. The additional reduction capacity would be provided to grout through increased slag, Fe amendment, or another suitable materials.	M Can do scaled demonstration to test reduction capacity.	H No additional costs as cheap/dry mix ingredients used.	**NO** Schedule Impacts; Could be implemented by 2034; 6 yrs	M Modeling exercises and scoping experiments have been done on reoxidation rate. Lysimeter test will assist with this. Need to resolve scrutiny on reduction capacity measurements (i.e. are an unknown). Excess reduction capacity already exists	(0) Does not increase or decrease waste loading.	+/- May destabilize an iodine getter system, benefits Hg and Cr	H Simple materials or alternate formulations	6	1
14	Iron Oxyhydroxide Treatment	Stable Tc phases are generated in precipitation or synthesis processes at various temperatures. Simple oxides would be added to the waste feed to grow solid phases which capture Tc and other contaminants. The phases stay in the LAW and are incorporated into the grout. Evidence in LAW-like conditions show Tc incorporates into Fe-oxide structure.	L Laboratory studies performed for simulant (pH 13 and high Cr) treatment with Fe(OH)2 to produce spinels, need to grout treated simulant and perform testing in simulated/real LAW. Effectiveness in LAW is unknown	M Add Fe(OH)2 to waste as pre-treatment (requires anoxic storage conditions to maximize reducing capacity)	**NO** Schedule Impacts; Could be implemented by 2034; 6 yrs	M Fe(OH)2 requires storage in anoxic conditions. Competition in LAW is unknown. Optimal loading amounts unknown.	(0) Does not increase or decrease waste loading. Could decrease waste loading if the amount of additive is a significant volume fraction.	(+) co-mingled redox species (e.g. Cr(VI) compete with Tc, reduced/immobilized provided enough Fe(OH)2 addition	L synthesized in the laboratory for testing	4	2
15	Use of ion exchange resins (SBAR/MRT) as getters for incorporation into grout waste form.	Use commercially available ion exchange resins for Tc capture then solidification directly in grout	L Can use commercially available materials. Select resins shown to collect Tc before stability of material is lost. Where to add the resin (liquid vs dry mix) is unknown.	M Resin likely to increase the cost of grout mix.	**NO** Schedule Impacts; Could be implemented by 2034; 6 yrs	M Stability of resin when in the grout, dehydration/expansion processes and resulting impact on COC release are unknown, Kinetics of Tc capture prior/during mixing. Should be compared with slag-based grout as a base case.	(0) Does not increase or decrease waste loading.	(+) Some resins also observed to sequester iodine.	H Commercially available resins	5	2
29	Conversion of non pertechnetate to pertechnetate	Focused mainly on AN/SY farms Non-pertechnetate is believed to be Tc(I) and redox reactions are expected to be ineffective for stabilizing Tc(I). Need one of two approaches first to address the non-pertechnetate: 1) a PA calculation with relevant data on non-pertechnetate leachability and 2) a scaled inventory of pertechnetate vs non-pertechnetate with improved analytical approaches. These would determine if a conversion of the non-pertechnetate is required.	L Limited lab testing done in LAW for conversion processes and efficiency. Tank inventories require better sampling and analytical approaches	Unknown as process not defined.	**NO** Could be implemented by 2034; 14 yrs	L -Process chemistry unknown, -Interferences in LAW, -How application looks, -Single test effort showed Dobs for Tc(I) is an order of magnitude higher than Tc(VII). -Need enabling technology to detect and quantify non pertechnetate in tank waste and deployed in 222-S to g et an inventory of non pertechnetate in tanks -Must first oxidize the organics, and probably nitrite; very energy intensive. Need to add caustic for organic destruction so increases volume.	(0) Unknown	(0) Unknown, but oxidation process may drive off other COC's/organics.	H Chemicals likely available.	2	4

Table 6-7: Engineered barrier technologies for iodine.

Note: Dark grey rows indicate technologies the expert panel did not rank and the light grey rows indicate technologies that were ranked but not considered viable enough to warrant a detailed written description.

Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
21	Sorbent addition to backfill around waste packages.	Amend backfill around waste packages to give increased Kd to iodine species. Can use commercially available materials. Low cost (e.g. Fe) amendments would be best. Can draw on previous experience in getter placement in Yucca Mountain development.	M/L Demonstrated in laboratory tests related to subsurface development; Effectiveness in natural environment is unknown.	M/L Dependent on the material used for the amendment;	**NO** Could be implemented by 2034	M Long term stability of the sorbent in the IDF is unknown; Not known if process would extend past the 500 year cap failure scenario	N/A	(+) Other COCs already have low Kd assigned in the subsurface but could also slow their migration	H Some commercially available	5	2
22	Functionalized clays placed around waste packages; could be mixed in with IDF backfill	Altered clays added to the backfill can sorb iodine in subsurface	M/L Demonstrated in laboratory tests related to subsurface development; Effectiveness in natural environment is unknown.	M/L Dependent on the material used for the amendment;	**NO** Could be implemented by 2034	M Long term stability of the functionalized clay in the IDF is unknown; Not known if process would extend past the 500 year cap failure scenario Need to characterize modified backfill properties for PA	N/A	(+) Other COCs already have low Kd assigned in the subsurface but could also slow their migration	L Require fabrication	4	2
23	Modify hydraulic properties of backfill	Modify the backfill around the waste packages to have lower hydraulic conductivity, thus slowing the migration of contaminants.	---	---	---	---	---	---	---	---	---
24	Granular Activated Carbon (GAC)	Granular Activated Carbon (GAC) layer beneath grout to capture leached Iodine as is used for groundwater applications	M GAC readily available. GAC documented to adsorb Iodine from groundwater. Affinity and competition with other anions unknown Iodine removal will depend on speciation	M Would add cost to IDF to amend backfill with GAC; Quantity of GAC needed is unknown	**NO** Could be implemented by 2034	M Long-term stability of the GAC is unknown Competition with other anions may occur	N/A	(+) Likely beneficial by also sorbing TcO4-	H Commercially available	6	3
25	Permeable Reactive Barrier (Type I) - Colloidal Ag	Injection of **colloidal silver chloride** into subsurface media to form a permeable treatment zone. Relies on low solubility of AgI to remove iodine from water (i.e., forms a stable solid). Highly effective for iodide but less effective for iodate and organoiodine species.	H/M Used at the Savannah River Site,	M Unknown; Dependent on single injections after IDF closure or during install	**NO** Would be implemented after IDF closure	M/L Injection requires permeable formation and access for deployment; Long-term stability of injected silver system is unknown, need to ensure no Ag migration;	N/A	(+) Ag may interact with other species as well	H Commercially available	6	2
26	Permeable Reactive Barrier (TYPE II) - Injection to form AgC1	Injection of dissolved silver **with in-situ reaction** to form AgCl in permeable treatment zone. Relies on low solubility of AgI to remove I from water. Similar to colloidal AgC1 treatment (above) but has potential for better subsurface distribution.	H/M Approved by South Carolina regulators and EPA and currently being used at SRS (F-Area).	M Unknown; Dependent on single injections after IDF closure or during install	**NO** Would be implemented after IDF closure	M/L Injection requires permeable formation and access for deployment; Long-term stability of injected silver system is unknown, need to ensure no Ag migration;	N/A	(+) Ag may interact with other species as well	H Commercially available	6	2
27	Pump and treat	Placement of a pump and treat system at the IDF to remove Tc/I leaching out over time. Can leverage from development under the deep vadose zone iodine remediation/control program.	---	---	---	---	---	---	---	---	---
29	Amended grout barrier in package	Prepare a "barrier" in waste packages with an amended grout first, then pour the waste grout slurry into the waste package. The amended grout layer would serve as a barrier to iodine migration. The amendment can be cation species, Fe, or clays.	M Process to make package commercially available or could be made	M Would increase cost of waste packages	**NO** Could be implemented by 2034	M Long term effectiveness of the barrier layer, stability of the amended grout to prevent fast transport pathways (e.g. cracks) is unknown	(-) Container material will take up space.	(+) Amendment layer may interact with other species	L Amendment species not defined Waste package preparation is commercially available.	4	1
30	Iodine reactions with corroding Fe and naturally present Fe minerals.	Rely on natural and waste package generated iron oxides to retard iodine mobility. Recent observations show iodine species interacting with Fe-oxides, May show subsurface models to be excessively conservative, Interactions with steel package possible to slow iodine migration.	M/L Field scale effectiveness unknown; Lab demonstration in subsurface conditions	H Fe materials are low cost and if corroding package the material is already present	**NO** Could be implemented by 2034	M Long term stability of I-sorbed onto Fe in IDF conditions is unknown	N/A	(+) Fe-oxides known to interact with other contaminants.	N/A	5.5	2
31	Sequestration below IDF (works only for iodate)	Injection of gaseous CO2 for co-precipitation with carbonates	M Field scale effectiveness unknown; Recent laboratory evaluations have shown that iodate can be co-precipitated with carbonates (PNNL-24709). Iodine association with carbonate precipitates has also been observed in recent vadose zone characterization studies (PNNL-26208; PNNL-26266), although those chemical extractions could also indicate iodine associated with iron oxides. Need to know rate of iodide to iodate conversion in subsurface	M Unknown Dependent on single injections after IDF closure or during install	**NO** Could be implemented by 2034 Technology will be tested as part of the DV-1 OU Laboratory Treatability Study in the FY21-22 timeframe (depending on funding)	L Effectiveness of the technology is unknown and will be tested as part of the DV-1 OU Laboratory Treatability Study where the technology is considered for saturated conditions at perched water or a water table PRB application beneath the BY Cribs. Interactions with other species leaching from IDF. How long will process work for	N/A	(0) Unknown; Treatability testing will include other COCs and their effects	H Injection processes available	4	4

Table 6-8: Engineered barrier technologies considered for technetium.

Note: Dark grey rows indicate technologies the expert panel did not rank and the light grey rows indicate technologies that were ranked but not considered viable enough to warrant a detailed written description.

Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
4	Enhance microencapsulation by reducing porosity through **lowering water to cement ratio** of base case Cast Stone	Optimize the water-to-cement ratio to minimize the final porosity.	H/M Dependent on available data relevant to Hanford LAW as existing data only goes to 0.4 w/cm	M Lowering water to cementitious reagents will increase waste form volume which will add cost.	**NO** Could be implemented by 2034	M Existing data on Hanford LAW is from 0.4 - 0.6 w:dm ratio. Material properties are unknown outside this w:dm ratio. Potential Limits on mixability container filling. Processing admixtures for high ionic strength, high Na, and v. high pH solutions not highly effective and introduce organic chemicals.	(-) Less than current Cast Stone	(+) Unknown, but if successful could reduce release of multiple contaminants	H	5.5	2
5	Enhance microencapsulation reduce porosity by **increasing particle packing** of Base Case Cast Stone.	Optimize particle packing plus reduce water to solids ratio (cementitious materials or inert fillers) to densify the matrix and reduce porosity. Could use modeling software to design.	L It is possible to use the theories of interparticle spacing (IPS) but has not been demonstrated for salt waste grouts	H Conceptually simple and uses low cast additives. Would need R&D efforts and longer if modeling incorporated	**NO** Could be implemented by 2034	M Unknown if successful in high salt waste grouts and if model projections would lead to improved properties. Cast Stone reagents or novel reagents must be customized w/respect to particle size which will add cost.	(-) Unknown impact on waste loading, but likely less waste loading than base case	(+) Unknown, but if successful could reduce release of multiple contaminants	H Materials used would be the same as those currently used in cement industry	5	3
6	Optimize Cast Stone Formulation	Cast stone development to date has been limited to a relatively narrow range of compositions. This option would expand that envelope to potentially identify better performing formulations.	M Optimization of grout properties via formulation changes is well established. However, the extent of the possible gains is unknown and needs to be determined via testing.	H Likely little or no increase in cost from base case.	**NO** Could be implemented by 2034; 14 yrs	M Limits unknown, which is why testing is needed.	(0) Likely no impact on waste loading.	(+) Unknown, but if successful could reduce release of multiple contaminants	H Would employ commonly available materials	7	3
8	Geopolymer matrix	Cement like materials made by alkali activated polymerization of aluminosilicates. Basic process is simple and involves mixing of dry powders with an aqueous activator solution, typically at ambient temperature.	M Tested at up to engineering scale. Tested with various waste streams and with Tc. Waste form qualification work done for secondary wastes. Passes TCLP for secondary wastes	H Likely little or no increase in cost from base case.	**NO** Could be implemented by 2034; 14 yrs	M Limits unknown due to limited development for LAW since ~2012. Exiting work indicates comparable to cast stone. Need to show property improvement vs cost compared with Cast Stone Passing TCLP for high Cr LAW.	(0) Likely no impact on waste loading.	(+) Unknown, but if successful could reduce release of multiple contaminants	H Employs commonly available materials	7	2
9	Mineral incorporation or sorption of contaminants to matrix phases.	Calcium sulfoaluminate hydrate-based matrices made from calcium sulfoaluminocement or mixture of ingredients resulting in same matrix mineralogy. The phases generated could incorporate specific contaminants or increase interactions with the matrix.	L Tested at lab scale to prove incorporation mechanism. Effect in LAW is unknown. Need optimization studies to prove desired effect in LAW and upscaling	H Likely no increase in cost from base case.	**NO** Could be implemented by 2034; 14 yrs	M Some waste form cracking/expansion could occur without formulation optimization for controlled mineral growth.	(0) Likely no impact on waste loading.	+/- Could work for multiple contaminants but can also cause interference e.g. iodate, pertechnetate, chromate and other oxyanions expected to compete for mineral incorporation.	H Likely use common materials	5	1
10	Acid - Base Cement Matrix	Phosphate cements (e.g. Mg-PO4) could be used to immobilize the liquid wastes. Mg-phosphate materials were considered in previous secondary waste formulation down selections with documented technical challenges.	---	---	---	---	---	---	---	---	---
11	Hydroceramics	Generate a hydroceramic as a waste form designed to immobilize LAW that is made from metakaolin plus NaOH and/or NaOH rich LAW supernatant. Requires a calcining step and control of Na/NO2/NO3 ratios to generate ceramic form	---	---	---	---	---	---	---	---	---
12	Bitumen	Original candidate in Hanford/SRS down-selection Used in the European Union. Is a thermal process that operates at ~ 140 C.	---	---	---	---	---	---	---	---	---
13	Sulfur polymer cement	Sulfur polymer cements use a sulfur source and commonly an aggregate to form a hardened material using a thermal process at ~140 C .	---	---	---	---	---	---	---	---	---
14	Steam reformed mineralized waste form or grouted mineralized waste form	Mineralized product or powder ingredient incorporated into cement or other ambient temperature monolith.	---	---	---	---	---	---	---	---	---
15	Review Hanford Grout original formulations	Original down selection process was focused on a pumpable mix and this is no longer a primary requirement. Assess if other mixes evaluated in the 1980's meet current needs for waste form qualification? The data can be reviewed to assess waste form qualification and assist with identifying promising alternate formulations.	Unknown Dependent on formulations and available data	Unknown Dependent on formulations	**NO** Identified formulations could be implemented by 2034	Unknown Dependent on formulations, waste form qualification data may be limited	Unknown	Unknown	Unknown Dependent on formulations	1	6
17	Evaporation of waste to decrease water content and reduce volume. Will also remove some organic chemicals.	Lower porosity could be achieved without increasing waste loading by concentrating LAW through evaporation.	M Evaporation already part of Hanford flowsheet, but ability to generate tailored waste form from salt wastes unknown	M/L Need to take into account the costs of the evaporation campaigns and how much benefit is gained by the waste form	Unknown: Could be implemented by 2034 but dependent on evaporation approach (use existing units or build new)	M Resulting properties unknown with increasing concentration of LAW	(+) Increase waste loading in primary waste form but could result in a secondary waste form from the evaporation process	(0) Unknown. Concentrating stream may alter the waste stream by increasing key components (SO4, Cr, Al etc.) that may cause deviations from the known performance.	H Evaporators present at Hanford	4.5	4

Table 6-9: Grout formulation modifications considered to modify the bulk properties of the waste form.
Note: Dark grey rows indicate technologies the expert panel did not rank.

Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
4	Enhance microencapsulation by reducing porosity through **lowering water to cement ratio** of base case Cast Stone	Optimize the water-to-cement ratio to minimize the final porosity.	H/M Dependent on available data relevant to Hanford LAW as existing data only goes to 0.4 w/cm	M Lowering water to cementitious reagents will increase waste form volume which will add cost.	**NO** Could be implemented by 2034	M Existing data on Hanford LAW is from 0.4 - 0.6 w:dm ratio. Material properties are unknown outside this w:dm ratio. Potential Limits on mixability container filling. Processing admixtures for high ionic strength, high Na, and v. high pH solutions not highly effective and	(-) Less than current Cast Stone	(+) Unknown, but if successful could reduce release of multiple contaminants	H	5.5	2
5	Enhance microencapsulation reduce porosity by **increasing particle packing** of Base Case Cast Stone.	Optimize particle packing plus reduce water to solids ratio (cementitious materials or inert fillers) to densify the matrix and reduce porosity. Could use modeling software to design.	L It is possible to use the theories of interparticle spacing (IPS) but has not been demonstrated for salt waste grouts	H Conceptually simple and uses low cast additives. Would need R&D efforts and longer if modeling incorporated	**NO** Could be implemented by 2034	M Unknown if successful in high salt waste grouts and if model projections would lead to improved properties. Cast Stone reagents or novel reagents must be customized w/respect to particle size which will add cost.	(-) Unknown impact on waste loading, but likely less waste loading than base case	(+) Unknown, but if successful could reduce release of multiple contaminants	H Materials used would be the same as those currently used in cement industry	5	3
6	Optimize Cast Stone Formulation	Cast stone development to date has been limited to a relatively narrow range of compositions. This option would expand that envelope to potentially identify better performing formulations.	M Optimization of grout properties via formulation changes is well established. However, the extent of the possible gains is unknown and needs to be determined via testing.	H Likely little or no increase in cost from base case.	**NO** Could be implemented by 2034; 14 yrs	M Limits unknown, which is why testing is needed.	(0) Likely no impact on waste loading.	(+) Unknown, but if successful could reduce release of multiple contaminants	H Would employ commonly available materials	7	3
8	Geopolymer matrix	Cement like materials made by alkali activated polymerization of aluminosilicates. Basic process is simple and involves mixing of dry powders with an aqueous activator solution, typically at ambient temperature.	M Tested at up to engineering scale. Tested with various waste streams and with Tc. Waste form qualification work done for secondary wastes. Passes TCLP for secondary wastes	H Likely little or no increase in cost from base case.	**NO** Could be implemented by 2034; 14 yrs	M Limits unknown due to limited development for LAW since ~2012. Exiting work indicates comparable to cast stone. Need to show property improvement vs cost compared with Cast Stone Passing TCLP for high Cr LAW.	(0) Likely no impact on waste loading.	(+) Unknown, but if successful could reduce release of multiple contaminants	H Employs commonly available materials	7	2
9	Mineral incorporation or sorption of contaminants to matrix phases.	Calcium sulfoaluminate hydrate-based matrices made from calcium sulfoaluminocement or mixture of ingredients resulting in same matrix mineralogy. The phases generated could incorporate specific contaminants or increase interactions with the matrix.	L Tested at lab scale to prove incorporation mechanism. Effect in LAW is unknown. Need optimization studies to prove desired effect in LAW and upscaling	H Likely no increase in cost from base case.	**NO** Could be implemented by 2034; 14 yrs	M Some waste form cracking/expansion could occur without formulation optimization for controlled mineral growth.	(0) Likely no impact on waste loading.	+/- Could work for multiple contaminants but can also cause interference e.g. iodate, pertechnetate, chromate and other oxyanions expected to compete for mineral incorporation.	H Likely use common materials	5	1
10	Acid - Base Cement Matrix	Phosphate cements (e.g. $\mathrm{Mg\text{-}PO_4}$) could be used to immobilize the liquid wastes. Mg-phosphate materials were considered in previous secondary waste formulation down selections with documented technical challenges.	---	---	---	---	---	---	---	---	---
11	Hydroceramics	Generate a hydroceramic as a waste form designed to immobilize LAW that is made from metakaolin plus NaOH and/or NaOH rich LAW supernatant. Requires a calcining step and control of $\mathrm{Na/NO_2/NO_3}$ ratios to generate ceramic form	---	---	---	---	---	---	---	---	---
12	Bitumen	Original candidate in Hanford/SRS down-selection Used in the European Union. Is a thermal process that operates at ~ 140 C.	---	---	---	---	---	---	---	---	---
13	Sulfur polymer cement	Sulfur polymer cements use a sulfur source and commonly an aggregate to form a hardened material using a thermal process at ~140 C .	---	---	---	---	---	---	---	---	---
14	Steam reformed mineralized waste form or grouted mineralized waste form	Mineralized product or powder ingredient incorporated into cement or other ambient temperature monolith.	---	---	---	---	---	---	---	---	---
15	Review Hanford Grout original formulations	Original down selection process was focused on a pumpable mix and this is no longer a primary requirement. Assess if other mixes evaluated in the 1980's meet current needs for waste form qualification? The data can be reviewed to assess waste form qualification and assist with identifying promising alternate formulations.	Unknown Dependent on formulations and available data	Unknown Dependent on formulations	**NO** Identified formulations could be implemented by 2034	Unknown Dependent on formulations, waste form qualification data may be limited	Unknown	Unknown	Unknown Dependent on formulations	1	6
17	Evaporation of waste to decrease water content and reduce volume. Will also remove some organic chemicals.	Lower porosity could be achieved without increasing waste loading by concentrating LAW through evaporation.	M Evaporation already part of Hanford flowsheet, but ability to generate tailored waste form from salt wastes unknown	M/L Need to take into account the costs of the evaporation campaigns and how much benefit is gained by the waste form	Unknown: Could be implemented by 2034 but dependent on evaporation approach (use existing units or build new)	M Resulting properties unknown with increasing concentration of LAW	(+) Increase waste loading in primary waste form but could result in a secondary waste form from the evaporation process	(0) Unknown. Concentrating stream may alter the waste stream by increasing key components (SO_4, Cr, Al etc.) that may cause deviations from the known performance.	H Evaporators present at Hanford	4.5	4

Table 6-10: Engineered barrier technologies considered to modify bulk properties.

Line Number	Technology	Description	Development Level (H/M/L)	NOT Likely to Increase Cost (H/M/L)	Schedule Impacts (Yes/No)	Likely NO Technical Limits (H/M/L)	Waste Loading (+/0/-)	Effects on Other COCs (+/0/-)	Availability (H/L)	Score	Uncertainty
19	Waste form size	Varying the waste package size could lower overall release over time, whether through slowing the re-oxidation rate or through a longer migratory pathway	H Can pour containers of varying sizes up to SRS vaults	H/M Dependent on waste form size	Unknown; Dependent on waste form size, most could be implemented by 2034	M The bigger container, the higher temperature and risk of cracking or other pathologies. Retrievability concerns. Measurement of reoxidation rates as function of size	(0) Potentially smaller number of packages but with same loading	N/A	H Canisters of all sizes available commercially	5.5	1
20	Long term redox control achieved through changing monolith size	The retention of Tc in grout is heavily dependent on the maintenance of reducing conditions in the waste form. A larger waste form size would oxidize at a slower rate	H SRS vaults prove this concept.	H Going to larger waste forms unlikely to increase costs up to a certain size.	Unknown; Dependent on waste form size, most could be implemented by 2034	M Modeling exercises and scoping experiments have been done. Lysimeter test will assist with this. Need to resolve scrutiny on reduction capacity measurements.	(0) Does not increase or decrease waste loading.	(+) Larger volume form may slow release of other species	H No change to ingredients. Need to be cognizant of slag/fa supplies	7	1
21	Modification of current steel container with concrete or other material for macroencapsulation of the waste form (liner inside container) or container (around outside of container).	Pouring a low permeability, high strength grout around the emplaced waste packages in the IDF, or add a low permiable liner within the container. This would serve as a barrier to water ingress and migration of contaminants.	M Similar to design at WCS, grout design unknown	H Unknown for the effectiveness of the barrier grout, requires R&D Will require costly re-design of IDF.	**YES,** Long implementation time; Would require repermitting of IDF.	M Unknown for the effectiveness of the barrier grout; Requires R&D	Unknown Grout would need to be defined	(+)	Unknown	3	5
22	Use HDPE as a barrier to slow ingress of water, delaying leaching	Use HDPE as a barrier, either put grout in HDPE containers (HICs) or place grout in metal or concrete containers and cover containers with HDPE cover	H Similar to FWF at WCS, Saltstone SDF at SRS	H HDPE covers or containers readily available	**NO** Could be implemented by 2034; 3 yrs	H - included in SRS PA. Delays leaching for ~2000 years; Acceptability at Hanford is Unknown	N/A	(+) Will decrease water intrusion and reduce migration of all contaminants.	H	9	1

7.0 Conclusions

Use of a cementitious solidification process waste to treat low activity Hanford tank waste is currently impeded by three regulatory-driven constraints. First, Ecology currently applies the treatment-based standard of vitrification to waste codes D002 (corrosivity), and D004-D011 (metals). The solution to this impediment is founded in regulatory negotiations between the DOE and Ecology and is outside the scope of this effort. Second, LAW may contain RCRA LDR organics at levels that require removal to meet LDR concentration-based limits. Third, current estimates based on PA fate-and-transport modeling indicate that if the entire quantity of LAW currently designated for supplemental treatment were solidified as a cementitious material the regulatory limits for nitrate, 99-Tc, and 129-I could exceed groundwater standards after DOE's time of compliance (1,000 years) but within 10,000 years after IDF closure." It should also be noted that this third issue is only applicable to on-site disposal, not offsite at WCS. This last point is based on calculations that assume either a low-performing grout is used, or high-performance grout does not maintain its integrity (SRNL-RP-2018-00687). Asmussen et al. (2019a) estimated that for the inventory of grouted SLAW to meet IDF groundwater requirements requires the release rate of nitrate, 99-Tc, and 129-I to be reduced by a factor of approximately 3.2, 10, and 32, respectively.

This report is expected to provide guidance for planning technology maturation efforts to ensure that an inventory of SLAW cementitious waste forms, if pursued, would meet, or exceed regulatory requirements for disposal onsite in the IDF or at an offsite facility.

A team of subject matter experts from PNNL, SRNL, Vitreous State Laboratory, and WRPS was assembled by the WRPS Chief Technology Office to identify, assess, and describe a broad range of technologies that could potentially mitigate the second and third limitation. The regulatory requirement for LDR organics (second limitation) is concentration based and must be met by the waste when it is received at the disposal location, whether onsite or offsite. Only two general approaches were identified by the expert panel that could meet this requirement. The first approach, termed Sample and Send, relies on direct sampling of the retrieved, or in-tank waste, and only sending material to supplemental treatment if it meets the LDR concentration limits. It should be noted that data on the concentrations of LDR organics in Hanford tank waste are very limited (RPP-RPT-61301; SRNL-RP-2018-00687) and cannot be relied upon to provide process knowledge that would eliminate the need to sample the tank waste. Part of the work included making use of headspace vapor measurements to estimate the concentration of LDR organics in the waste as the waste sits in the tanks today, but this evaluation cannot be relied upon when sludge, saltcake, and supernatant are being mixed and retrieved to eliminate the need to sample. These studies have found that very few of the compounds appear to be present at levels above regulatory limits (RPP-RPT-61301; SRNL-RP-2018-00687). Hence, there may be a substantial quantity of LAW that can be shown through sampling and analysis with improved method detection limits that the waste can be solidified in a cementitious waste form without exceeding LDR and TSCA concentration limits. The Sample and Send approach was highly rated by the expert panel.

The second approach identified for mitigating LDR organics is the use of a pretreatment process. Three chemical and one electrical organic destruction process were identified and evaluated. Two physical removal process (evaporation and GAC adsorption) were also evaluated. Of these options, the use of evaporation to volatilize and separate organics from the LAW was identified as the most promising.

The solution to the third impediment; the need for reduced nitrate, iodine, and technetium release rates to meet groundwater standards was also assessed in this effort. Pretreatment technologies were found for iodine and technetium, but no viable pretreatment options were identified for nitrate. Direct sampling of the retrieved, or in tank waste, and sending only selected material to supplemental treatment (i.e., Sample and Send) was the highest rated option for both technetium and iodine. Note that Figure 2-12 suggests there may be waste streams low in organics, 99-Tc, and 129-I and thus amenable to a Sample and Send approach. The most promising methods to reduce nitrate release rates are associated with either amending the backfill or modifying the bulk properties of the waste form.

Four methods were identified to modify the waste formulation to enhance the retention of iodine and five similar methods were identified for technetium, respectively. For iodine, the use of a strong base anion resin as an adsorption additive and the addition of soluble species to facilitate iodine precipitation were the highest scored option, although both also have significant uncertainties. The two highest scoring technical approaches for technetium retention in the waste form are both associated with enhancing the reducing capacity of the waste form. It was also noted that the success of technetium retention methods may be hampered by our lack of understanding of the fraction of tank waste technetium that is present in the reduced Tc(I) form.

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APPENDIX A
TECHNOLOGY DESCRIPTIONS

APPENDICES TABLE OF CONTENTS

ACRONYMS AND ABBREVIATIONS

CAC	calcium aluminate cements
CASH	calcium sulfoaluminate hydrate
COC	constituent of concern
CSA	calcium sulfoaluminate cements
DOE	U.S. Department of Energy
DWPF	Defense Waste Processing Facility
EMF	Effluent Management Facility
GAC	granular activated carbon
HDPE	high-density polyethylene
IDF	Integrated Disposal Facility
LAW	low-activity waste
LDH	layered double hydroxide
LDR	Land Disposal Restriction
OPC	ordinary Portland cement
PA	performance assessment
RCRA	Resource Conservation and Recovery Act
redox	reduction and oxidation
RL	Richland Operations Office
SBAR	strong base anion exchange resins
SLAW	supplemental low-activity waste
SRS	Savannah River Site
WTP	Waste Treatment and Immobilization Plant
ZVI	zero valent iron

1.0 IODINE

It should be noted that methods that rely on chemical properties of iodine to remove or retain 129-I must be scaled appropriately to address both 127-I and 129-I present in the waste since the chemical properties of the two isotopes are identical.

1.1 Pretreatment Technologies

1.1.1 Line 4 – Silver Based Capture Media

This pretreatment technology would use a column packed with a silver-containing media through which filtered and ion exchanged (for cesium removal) low-activity waste (LAW) would be passed. The media would remove iodine species through precipitation (e.g., as silver iodide). An example media is a silver impregnated zeolite and other similar commercially available materials. The effluent LAW would be moved to a grout treatment and the resulting iodine (including 129-I) loaded media would be disposed of as a secondary waste (e.g., direct densification of the loaded media [Bruffey and Jubin 2015] or solidified in grout [Seitz 2017; Asmussen et al. 2019]).

The silver based media pretreatment approach is promising since media have demonstrated iodide removal from LAW in batch tests on the bench scale (Asmussen et al. 2016c). Several potential benefits of the technology are:

- It would decrease the iodine inventory in the primary LAW grout in the Integrated Disposal Facility (IDF).
- Potentially place the iodine in a more stable form in the IDF if the iodine-loaded media is disposed as secondary waste in the IDF as a densified waste form or grouted itself in a slag-free grout.
- The removal of iodine from LAW would allow reduction chemistry to be used in the primary LAW form. The addition of a Ag-based iodine getter to a slag-containing waste form can destabilize the AgI or use up reduction capacity that is available from the slag to immobilize reduction oxidation (redox) active species such as 99-Tc or Cr. By decreasing the iodine inventory in the LAW, improved iodine retention in the primary LAW form using getters would not be required.

These potential benefits cannot be realized without overcoming technical challenges with implementation of the technology. Silver carries both a high cost and a Resource Conservation and Recovery Act (RCRA) designation. The common zeolite substrate used in these materials is likely not stable over long service times in the column due to the high pH environment and has been shown to release silver in LAW (Asmussen et al. 2016c). Although, other substrates such as carbon have shown evidence of retaining Ag in LAW (Asmussen et al. 2016c). In order to be successful, a more stable substrate for the Ag would be required and will need to be demonstrated using LAW.

1.1.2 Line 5 – Superlig 639 Elutable Resin as a Capture Media

SuperLig® 639 is a resin that is highly selective for removing the pertechnetate ion (TcO_4^-) from high ionic strength solutions. The use of SuperLig 639 to remove pertechnetate from Hanford tank waste was in the initial baseline flow-sheet and no significant technical issues were identified for 99-Tc (additional

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description in the 99-Tc section). In prior testing the SuperLig 639 selectively removed 99% of pertechnetate from LAW but was also shown to remove some iodine from actual tank waste (Nash et al. 2013).

In this application, the resin would be loaded into ion exchange columns, and the LAW pumped through the columns to remove the 129-I. Typically, resin is eluted to remove the ions, regenerated, and returned to service. The eluate, containing the radionuclide of interest, is then treated as a secondary waste and disposed of separately from the parent LAW stream through another immobilization process. This second immobilization process would entail development of an iodine-specific waste form with unknown performance if disposed of in the IDF. In this application, direct disposal of the resin loaded with the radionuclides without elution in the IDF is not viable because the 129-I (and 99-Tc) would presumably leach readily from the resin and have no retention, even in the short-term. However, direct disposal of loaded resin at an offsite facility would be technically plausible.

The primary development requirement would be to develop a resin with improved iodine removal properties and less frequent elution. The distribution coefficient for 129-I capture on SuperLig 639 in actual Savannah River Site (SRS) tank waste was 49 mL/g, compared to 582 mL/g for 99-Tc for the same sample. This indicates that the breakthrough of iodine in a column would occur at <100 bed volumes before elution was needed. Frequent elution would make this process for iodine capture very challenging to operate. Note that if this resin is chosen for use to remove technetium, it will also remove ~10% of the iodine. This decrease in iodine inventory may have a beneficial impact on iodine release from the IDF. It is assumed, but not demonstrated, that iodine will subsequently be removed from the resin during elution.

1.1.3 Line 6 – Strong Base Anion Exchange Resins as a Capture Media

In this application, ion exchange resins would be loaded into columns, and the LAW pumped through the columns to remove the 129-I. The resin is either eluted or disposed directly. Strong base anion exchange resins (SBAR) are commonly available from many commercial suppliers. They are typically used to deionize water by removing the anions, replacing them with hydroxide or a spectator anion (e.g., chloride). Generally, these have a styrene divinylbenzene or polyacrylic backbone structure as either a macroreticular or gel-type material with the functional group as a quaternary amine ($R\text{-}NR'_3{}^+$ where R = polymer backbone and R' = alkyl groups or hydrogen atom). Typically, resin is eluted to remove the ions, regenerated, and returned to service. The eluate, containing the radionuclide of interest, is then treated and disposed as secondary waste through another immobilization process. This second immobilization process would entail development of an iodine-specific waste form with unknown performance if disposed of in the IDF. In some specific applications, the resins are not eluted, but are instead disposed directly as solid waste.

Testing on SBAR has been performed with SRS tank waste (Walker et al. 1985). The styrene-divinylbenzene resins Dowex® 1X8 and 1X10 (gel) and Amberlite® IRA-904 (macro) were tested for removal of technetium from SRS tank waste, but the removal of iodine was also measured. Both of these resins have quaternary amine functional groups. The 50% breakthrough of iodine for these resins was observed at 15 bed volumes. The elution of these resins requires high concentrations of nitric acid, so the low loading and frequent elution of these resins is not practical as an iodine removal process. Likewise,

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loading the iodine onto these resins and discarding the resins after processing only ~15 bed volumes indicates that direct disposal is also not practical because of the extremely high volume of loaded resin for disposal. Improved resins would need to be developed, tested and deployed.

1.1.4 Line 13 – Low Iodine Inventory Tanks Selected Specifically for SLAW Grout (Sample and Send)

Concentrations of contaminants in the waste feed heavily influence the resulting waste form's contribution to release in the IDF (e.g., 99-Tc, 129-I) and acceptance in a disposal facility (organics). Complex waste feed profiles that combine retrieved wastes from a range of tanks have been developed in projections for Hanford Waste Treatment and Immobilization Plant (WTP) operations (Anderson et al. 2017). An initial evaluation of the LAW feed profiles highlights the possibility that many years of this projected feed would have very low contaminant contents and thus be strong candidates for a cementitious waste form, Section 2. A thorough evaluation of tank contaminant inventories after retrieval would build on this work and provide targets for a supplemental immobilization pathway where the waste feed has one or more of the following characteristics:

1. Low 99-Tc content,
2. Low iodine content, or
3. Land Disposal Restriction (LDR) listed organics at concentrations low enough to produce a final waste form that is below the listing levels.

The findings of the evaluation can identify tanks or tank farms where updated sampling and analysis can confirm inventories and select those identify tank(s) that meet the above criteria. These tanks would then be candidates for supplemental LAW (SLAW) grout. This sample and send approach would be especially beneficial in 200 West Area tanks where cross-site transfer will be required to deliver feed to the WTP. The inventories of 99-Tc have been well documented (Serne et al. 2014), recent efforts have attempted to document the process history and sampling information related to organics (Lindberg et al. 2019), while published iodine information is limited but available through the Tank Waste Information Network System (TWINS) and evaluations of this inventory are ongoing at Pacific Northwest National Laboratory. This approach to waste treatment is low-risk as it requires no technology development and could be implemented immediately, although it is not known how many tanks may contain low enough concentrations of these contaminants to meet the objectives, but could provide significant value to the development of flow-sheets for SLAW if a sufficient number are identified.

1.2 Waste Form Improvement Technologies – Targeting Both Slag and Slag-Free Systems

The stability of iodine in a grout waste form can be improved through the generation of low solubility iodine phases (e.g., AgI, BiI_3, PbI_2) (Scheele et al. 1984). The generation of these phases can be achieved using the approaches described below where the cation that captures the iodine (e.g., Ag^+, Bi^{3+}, Pb^{2+}) is introduced into the LAW either with an inorganic substrate (i.e., as a getter) or directly added as a soluble salt.

1.2.1 Lines 15 and 16 - Ag or Non-RCRA Particulate Getters on Inorganic Substrate

The addition of commercially available Ag-containing getters to grout has been demonstrated to enhance iodine retention in waste forms fabricated from LAW (Crawford et al. 2017), and secondary wastes (Lockrem 2005a; Saslow et al. 2017a). In these examples the getter was added at ≥5 wt% in the formulation. The improved retention arises from precipitation of AgI from the waste that is incorporated on the getter in the grout slurry. An abundance of Ag in the waste form prevents further release. Other examples adding <0.5 wt. % of the getters have not been successful (Asmussen et al. 2016a; Gong et al. 2019). The minimum waste form getter loading that leads to improved iodine retention is unknown. Minimizing getter loading is important to minimize cost since Ag-based getters are more costly than most other getters.

All examples to date related to liquid waste immobilization have used formulations which contain blast furnace slag. There is a known interference on AgI stability from slag, resulting from the sulfide content and reducing environment in the waste form (Asmussen et al. 2017a; Kaplan et al. 2018a; Kaplan et al., 2018b; Asmussen et al. 2019) but this interference has been demonstrated to be overcome with an excess of Ag in the waste form. Although the impact on other species that rely on reduction to be sequestered (Cr(VI), Tc(VII)) is unknown. This interaction needs to be considered when defining optimal Ag getter loadings or other metal cation-based getters.

Use of Ag-getters has the disadvantages that Ag is an expensive reagent and is also a RCRA regulated metal. Both challenges can theoretically be overcome by replacing the Ag on the getter with a lower cost, non-RCRA species (e.g., Bi^{3+}). Such materials have been developed and tested for iodine capture in liquid environments (Leonard et al. 2019). As other cation species can generate stable iodine phases, the likelihood of similar improvements in iodine retention with the non-RCRA materials is high. Historical work on the stability of non-RCRA in grout is available, albeit limited (Scheele et al. 1984).

1.2.2 Line 18 – Ion Exchange Getter Using Strong Base Anion Resins

In this application, SBARs are blended with the grout-formers and LAW to produce a waste form or mixed with the LAW to capture 129-I and then the mixture is incorporated into the grout. The SBAR may also be added first to the LAW to sequester the 129-I then fabrication of the grout slurry. SBARs are common ion exchange resins available from many commercial suppliers. They are typically used to deionize water by removing the anions, replacing them with hydroxide or a spectator anion (e.g., chloride). Typically, these have a styrene divinylbenzene or polyacrylic backbone structure as either a macroreticular or gel-type material with the functional group as a quaternary amine ($R\text{-}NR'_3{}^+$ where R = polymer backbone and R' = alkyl groups or hydrogen atom). Testing has been performed with SRS tank waste using SBARs in a column configuration (Walker et al. 1985). That testing indicated 50% breakthrough of 129-I in 15 bed volumes, indicating that the capacity for selective removal of iodine is low and would require an unknown but potentially large quantity of resin. Instead of column adsorption, this application would blend the SBAR directly with the grout, improving 129-I (and 99-Tc) retention in the solid waste form. Although the selective capacity of the resin is low, the sorbed species are only eluted using acid. In the alkaline grout environment, the species would likely remain sorbed to the resin, in turn, slowing the release of 129-I from the waste form. As discussed in the section on distribution coefficients, a ~10× decrease in the iodine release rate would be ensure the contribution from an SLAW inventory in the IDF would be below regulatory limits. Testing and calculations would be needed to determine if this is a viable strategy for retaining 129-I in the final waste form. Testing would also be needed to identify the optimal resin, methods to incorporate it into the final waste form, stability in the grout and its impact on other constituents of concern (COCs), especially 99-Tc and RCRA metals.

1.2.3 Line 19 – Addition of Chemicals to Grout to Generate Iodine Phases with Low Solubility

In this approach the functional cation would be added to the LAW or grout dry mix in a soluble form to allow dispersion in the liquid/slurry to precipitate low solubility iodine phases. Upon curing the precipitated iodine phase would be present / microencapsulated in the grout matrix and the release of iodine controlled by the solubility of the phase. A number of iodine species have been evaluated previously in ordinary Portland cement (OPC) systems (Scheele et al. 1984).

This approach would present a simpler, lower cost method for generation of stable iodine phases than solid substrate getters, especially if it used commercially available chemicals. However, this approach has not yet been demonstrated in LAW and the likelihood of success is similar to that for the success of substrate-based getters as both approaches generate identical stable iodine phases (e.g., AgI, BiI_3). Many cations capable of generating low solubility iodine phases are RCRA listed and their release must also be evaluated (e.g., Ag, Pb). The resulting metal-iodide phases may be destabilized by the reducing environment in slag containing systems.

1.3 Engineered Barrier Technologies

1.3.1 Lines 21, 22, and 24 – Amend IDF Backfill with Sorbents

Following closure of the IDF the migration of species away from the waste forms can be retarded by amendments that were added to the backfill as the backfill was placed (i.e., during waste disposal operations). Two types of backfill are identified for use between containers within the IDF (RPP-RPT-59958, Rev.1). Both backfills are a mixture of clay and native fill material. Amendment materials would be incorporated into the IDF backfill before being packed around the waste forms. As infiltrating water moves contaminants (e.g., iodine) away from the waste form the contaminants would be exposed to and be sequestered by the backfill through a sorption or precipitation process. In turn, a higher distribution coefficient, K_d, can be assigned to the backfill and be represented in simulations to demonstrate better retain iodine in the IDF. A similar concept was investigated previously for iodine at the Yucca Mountain Repository (Krumhansl et al. 2006) and the SRS (Kaplan 2001). The primary challenges with this technology are:(1) ensuring the activity of the amendments is retained beyond the 500-year cap failure scenario in the IDF, (2) effect the development of porosity/permeability and preferential pathways as the result of as-installed inhomogeneity, (3) developing and testing the amended backfill that balances the need for contact between 129-I and Ag and (4),(4) cost, preparation, and quality control of the amended IDF fill. If successfully demonstrated, the approach could offer a low cost means of ensuring slowed iodine migration over long times.

Several material types have been identified that could serve as amendments. In addition, Strickland et al. (2017) have reviewed amendments that could be applicable to the Hanford subsurface. Specific promising amendments include:

- **Iron based minerals:** Naturally occurring iron minerals and artificially added iron (e.g., zero valent iron [ZVI], a corroding steel container) can provide iron oxides within the backfill. Iron oxides have been shown to sorb iodine species in subsurface environments (Wang et al. 2019).
- **Clay minerals:** A naturally occurring or synthetic clay can be functionalized to enhance stability and iodine capture in the IDF backfill (Kaplan 2001).
- **Granular activated carbon (GAC):** Activated carbon can be used for iodine removal from aqueous environments by a physisorption mechanism. Removal efficiency is dependent on the

iodine species present (Parker et al. 2014). The impregnation of the GAC with Ag would increase the capture of iodine.

- **Ag-based Amendments:** The backfill can be modified with colloidal AgCl, $AgNO_3$, or Ag nanoparticles coated on inorganic substrates. These types of Ag treatments have been developed and deployed for 129-I sequestration.

1.3.2 Lines 25 and 26 – In Situ Chemical Treatment to Form Reactive Barrier

Permeable reactive barriers have been widely used for in-situ treatment of contaminated groundwater. A permeable reactive barrier can be constructed by:

1. Injecting a solution containing a dissolved chemical into a permeable subsurface saturated or unsaturated formation so the chemical can react with the soluble COC to form an insoluble precipitate.
2. Injecting colloidal or small solid particles into a permeable subsurface formation in the pathway of migrating contaminated water which react with the groundwater or pore water (vadose zone) to produce a geochemical environment conducive to precipitating or destroying the target contaminants.
3. Installing a permeable ground water barrier in a subsurface trench. The barrier material is designed to allow water to flow through it and to include reactive solid particles that will precipitate or destroy the targeted contaminants.
4. Installing a funnel and gate system in which a low permeability barrier is installed in the subsurface which directs water to flow through short permeable sections referred to as gates, which contain reactive material for removing or destroying the targeted contaminant. The gate sections of this barrier can be replaced or modified as needed.

Permeable reactive barriers could be applied below the IDF to mitigate iodine transport. Technologies for the formation of the barriers are commercially available. The long-term performance of such barriers is unknown.

Injection of AgCl Particles (Line 25): The SRS has successfully injected AgCl particles into groundwater contaminated with 129-I (Denham et al. 2010b). AgCl has very low solubility, $K_{sp} = 10^{-9.7}$, and is not soluble in ambient groundwater at the site where it was deployed. The solubility of AgI ($Ksp = 10^{-16.1}$) is lower than AgCl and in the presence of 129-I the Cl is displaced by the I and AgI forms sequestering the 129-I in the subsurface. This has been demonstrated in laboratory and field-scale studies. Unlike most other permeable reactive barriers AgCl has minimal effect on prevailing groundwater chemistry and does not noticeably affect chemical equilibrium in the aquifer. AgCl is highly effective at treating iodide but less effective for iodate and organo-iodine species (Denham et al. 2010a; Denham et al. 2008).

Injection of Dissolved $AgNO_3$ (Line 26): More recently at the SRS, laboratory work has been completed to test the emplacement of silver by injecting dissolved $AgNO_3$ (Denham et al. 2015a; Denham et al. 2015b). Once injected the Ag^+ reacts with 129-I sequestering it in-situ or it reacts with Cl^- precipitating AgCl in the aquifer where it will later react with 129-I migrating into the treated zone. Injection of $AgNO_3$ has the benefit of treating a larger zone with silver due to migration of dissolved silver and preferentially treats higher permeability zones where 129-I is likely to migrate (Denham et al. 2008). Although Ag is a RCRA regulated compound and NO_3^- is regulated as a drinking water contaminant, potential regulatory issues of injection of AgNO3 were resolved at the SRS.

1.3.3 Line 29 – Incorporate an Amended Grout Barrier in the Waste Package

The current concept for a SLAW grout is to pour the waste form into a steel container. Enhancements of this basic waste package could include lining the waste container with a (1) low permeability reactive barrier containing colloidal AgCl, or Ag impregnated porous substrates or coated particles or (2) a relatively low permeability reactive barrier containing one or more of the Ag or other non-RCRA metals that react with I^- to form low or very low solubility Ag compounds (see Lines 25 and 26 - In-situ Treatment and Waste to Form Reactive Barriers, above).

Diffusion of I^- through the waste form pore solution and into the reactive barrier liner is expected to result in precipitation of AgI (or other low solubility metal iodide compound). The stabilization reactions, mechanisms and time frames for TcO^-_4 (relatively fast chemical stabilization during early curing by reaction with sulfide in the waste form but subject to oxidation and solubilization from reaction with oxygen over time) and for I^- (physical stabilization related to ion diffusion through the waste form and liner pore fluids – years to decades) require separate amendments that do not interact with one another.

Feasibility of this concept can be evaluated in a simplified manner by bench-scale testing. However, sensitivity simulations (parameters, conditions, assumptions) may be the most useful means of evaluating the potential for long term benefits. A modeling evaluation of this approach was presented during the National Academy of Sciences Review of Hanford SLAW (Alexander 2018).

1.3.4 Line 30 – Reduced Mobility by Iodine Reactions with Corroding Fe and Naturally Present Fe Minerals

See the technology description for Line 21 in the iodine section of this appendix.

1.3.5 Line 31 – Gaseous CO_2 Injection to Precipitate with Carbonates

Incorporation of contaminants into subsurface minerals would provide a stable barrier that further limits COC migration as release would be dictated by the dissolution of the host mineral. Iodine migration is a particular challenge due to the high mobility of anionic iodine species. Within the Hanford subsurface, regardless of source, the predominant iodine species is iodate. Recent characterization studies of the Hanford vadose zone have identified iodine association with carbonate precipitates (Truex et al. 2017). This interaction has been further characterized in recent laboratory evaluations that iodate can be co-precipitated with carbonate minerals (Truex et al. 2016). The precipitation of carbonate containing minerals below the IDF could be achieved through gas phase injections of CO_2.

However, there are three critical unknowns for using this process in perpetuity at the IDF. These unknowns are (1) how long the growth of carbonate minerals can be sustained, (2) the longevity of any captured iodine, and (3) the rate of conversion of released iodide from the waste form to iodate. These topics are under investigation by the U.S. Department of Energy (DOE) Richland Operations Office (RL) funded programs.

A similar concept is currently being evaluated under field conditions at the SRS. A solution containing 3.5 mM NaOH and 7.9 mM $NaHCO_3$ is injected to raise the pH of the soil to sorb U-238 and Sr-90 in F Area. The 7.9 mM $NaHCO_3$ was a buffering agent in this case limiting the pH increase and preventing subsequent release of other COCs and dispersion of mobile colloids. The injection is currently ongoing. The frequency of periodic injections and duration of the treatment depends on natural reduction of pH from acidic groundwater migrating into the treated zone. Eventually the natural pH of the aquifer and sediments will be restored from their current acidic condition and additional base injection will not be necessary to maintain sorption of Sr-90 and U-238 onto aquifer solids.

2.0 TECHNETIUM

2.1 Pretreatment Technologies

The removal of 99-Tc from LAW in a pretreatment step was previously included in the Hanford LAW WTP flowsheet and removed in 2003 (Burgeson et al. 2005; Serne et al. 2016a). The proposed approach at WTP was the use of an ion exchange resin to remove TcO_4^-. A previous summary of alternate 99-Tc capture approaches within the Hanford flowsheet is presented elsewhere (Serne et al. 2016a). The most promising and technically mature technologies available to date are discussed below.

2.1.1 Line 4 – Elutable Ion Exchange/Sorbent Molecular Recognition Technology

In this application, ion exchange resins would be loaded into columns, and the LAW pumped through the columns to remove the 99-Tc. The absorbed 99-Tc is subsequently removed from the resin by elution and immobilized separately. The columns are regenerated and returned to service for another loading-elution cycle. SuperLig 639 is a resin that is highly selective for removing pertechnetate ion (TcO_4^-) from high ionic strength solutions. It can selectively remove 99+ % of the pertechnetate from LAW and is eluted with warm water. The use of SuperLig 639 to remove pertechnetate from Hanford tank waste was in the initial baseline flowsheet and design for the Hanford WTP but was removed in 2003. The process was highly effective, and no significant technical issues were identified. The technical maturation activities that were completed included testing with several actual tank waste samples, full-height columns with simulants, and manufacturing scale-up (Burgeson et al. 2005; Hassan et al. 2002; McCabe et al. 2001). The resin can extract the technetium from approximately 300 bed volumes of waste for each loading cycle. After absorbing the pertechnetate, the resin is eluted with about 20 bed volumes of warm water, re-conditioned, and then returned to service for reuse multiple times. The water eluate contains the pertechnetate and the original flow-sheet was to concentrate the eluate by evaporation, and then incorporate it into the HLW melter feed. It is also possible to immobilize the eluate as a secondary waste.

For the current potential application of using a grout waste form to immobilize SLAW, the SuperLig 639 could be used to remove pertechnetate from the aqueous LAW. It is reasonable to assume that 99% of the pertechnetate can be removed, but none of the non-pertechnetate would be removed without prior treatment to convert it to pertechnetate. The eluate containing the technetium could be treated in several ways for the current application. Similar to the original WTP baseline approach, the eluate could be evaporated in WTP and then vitrified in the HLW melter. (In the interim until HLW vitrification begins operation, evaporation could be performed in the Effluent Management Facility (EMF), which is no longer utilized in this flowsheet; and stored.) Alternatively, the technetium could be precipitated from the eluate with a reducing agent and removed by filtration, sorbed onto another ion exchange resin and disposed, or it could be evaporated and immobilized as a separate grout waste form. Vitrification with Hanford HLW was demonstrated using concentrated eluate from the actual tank waste testing, but the other potential disposal paths were not.

2.1.2 Line 5 – Strong Base Anion Exchange Resin

In this application, ion exchange resins would be loaded into columns, and the LAW pumped through the columns to remove the 99-Tc. The resin is either eluted or disposed directly. SBARs are commonly available from many commercial suppliers. They are typically used to deionize water by removing the anions, replacing them with hydroxide or a spectator anion (e.g., chloride). Generally, it is a styrene divinylbenzene or polyacrylic based backbone structure as either a macroreticular or gel-type material with the functional group as a quaternary amine ($R\text{-}NR'_3{}^+$ where R = polymer backbone and R' = alkyl groups or hydrogen atom). Testing has been performed with SRS tank waste (Walker et al. 1985) and Hanford tank waste. The styrene-divinylbenzene resins Dowex 1X8 and 1X10 (gel) and Amberlite IRA-904 (macro) were selected for SRS column tests. Both of these resins are quaternary amine functional groups. The 50% breakthrough of technetium for these were observed at 173 (Amberlite) and 125-145 (Dowex) bed volumes. Hanford LAW was tested in 222-S laboratory using Purolite® A530E (Duncan et al. 2012), which is a bifunctionalized alkylamine resin with improved selectivity for pertechnetate. (This resin is currently used at Hanford at large scale to treat groundwater in a pump-and-treat system.) In the testing of LAW treatment, the initial batch contact testing indicated that the 50% breakthrough point in a column should be reached at ~3000 bed volumes. However, the column testing with 6.5 M [Na^+] simulant indicated a 50% breakthrough at 284 bed volumes. During storage of resin in another bottle of simulant, the liquid phase turned distinctly yellow within seven days at ambient temperature. These two observations suggest that the resin degraded. This is also consistent with vendor literature that although it is stable to pH 14, the operating range for A530E is 4.5-8.5 (Purolite 2020). Another anion resin, Reillex® HPQ, has also been tested using actual AW-101 waste and simulated Hanford LAW (Blanchard et al. 1996). That resin is a divinylbenzene and methylated 4-vinylpyridine. The 50% breakthrough was at 130 bed volumes. Overall, it is very challenging to elute these SBARs, typically requiring 4-8 M nitric acid or other special conditions and it is also inefficient to discard them due to low loading.

For the current process to immobilize SLAW in grout, the technetium could be removed using a SBAR, although the capacity of the resins for technetium is not very high, requiring a large amount of resin. There are two options for handling the technetium stream. One option is to elute the technetium from the resin with concentrated acids, and further treat or immobilize it in a second waste form with unknown impacts. The second would be to load the SBAR resins with technetium and dispose the loaded resin directly without elution, although this also has unknown secondary impacts. For example, it is not known if the spent resin could be disposed directly, or if secondary processing or solidification as a monolith are needed, whether other COCs would cause issues, etc. Among the challenges are the estimated volume of spent resin to be disposed. Based on the testing, the phase ratio is ~200:1 (i.e., 200 gal of treated LAW liquid generates 1 gal of spent resin). At this loading ratio, the Tc-loaded spent resin would likely be Class C for some LAW tanks and greater than Class C for others. To improve this, development of more selective media with higher capacity would be beneficial. Another challenge is the stability of the resins in the highly alkaline LAW solution. Similarly, performance of the spent resin during disposal is also unknown. None of the non-pertechnetate would be removed from the LAW without prior treatment to convert it to pertechnetate.

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2.1.3 Line 6 - Redox Strike/Co-Precipitation

The reduction of 99-Tc from its soluble $Tc(VII)O_4^-$ form to a low solubility Tc(IV) species can be utilized to extract 99-Tc from liquid waste streams (Serne et al. 2016a). Several candidate reductants exist which can drive the reaction of Tc(VII) including Sn(II) (Asmussen et al. 2016b; Duncan et al. 2016; Taylor-Pashow et al. 2018), Fe (0 or II) (Del Cul et al. 1995) and S(0) (Pearce et al. 2018). In this approach a simple chemical form of the reductant would be introduced to the LAW to precipitate 99-Tc. The precipitated 99-Tc would then be filtered out and disposed of in a secondary waste form (for which multiple candidates exist [Serne et al. 2016a), or remain in the LAW feed and blended into a grout.

The primary challenges with the approach are the presence of excess Cr(VI) in the LAW (along with other oxidants such as NO_3) that would compete for reductants (~50 × higher than 99-Tc), decreased efficiency of some reductants in alkaline conditions (Asmussen et al. 2016b), and the potential need for a filtration step to collect the precipitated 99-Tc. The strike could be performed as a direct chemical addition or in a column. Disposal of the 99-Tc-containing solid phase would require a secondary waste form or inclusion in the HLW or LAW glass.

2.1.4 Line 9 – Sample and Send Only Low Technetium Waste to SLAW

See technology description for iodine, Line 13.

2.2 Waste Form Improvement Technologies

2.2.1 Line 11 – Chemical Reduction of Tc(VII) by Slag Glass Containing Sulfur in Caustic Solution

Tank wastes at Hanford and the SRS are alkaline salt solutions. Two redox sensitive contaminants, pertechnetate and chromate, are present in these waste solutions. TcO_4^- and CrO_4^{2-} are the most prevalent forms of these contaminants in the tank waste. In these oxidized forms, both contaminants are soluble and have transport properties similar to mobile, highly soluble, non-sorbing species (e.g., , nitrate, nitrite, and iodide) which are not chemically stabilized in Portland cement- or slag cement-based waste forms. However, chemical reduction of these contaminants (Tc^{+7} to Tc^{+4} and Cr^{+6} to Cr^{+3}) can result in precipitation of low solubility phases, TcS, TcO_2 and technetium oxy-hydroxides (Allen et al. 1997; Li and Kaplan 2012) and $Cr(OH)_3$ (Langton 1986; Langton 1987) and thereby are chemically stabilized by the waste form resulting in slowed release.

Both Cast Stone and Saltstone are slag cement waste forms containing (1) fly ash that serves as a supplementary cementitious material and initial heat "sink" and (2) a small amount of cement or $Ca(OH)_2$ that provides a source of calcium for fresh property enhancement and formation matrix phases (calcium silicate hydrates and calcium aluminosulfate hydrates). The primary features of both the Cast Stone and SRS Saltstone mix designs are (1) chemical stabilization of redox sensitive contaminants via chemical reduction of these ions and precipitation of low solubility phases and (2) physical stabilization of soluble and solid phases and by microencapsulation in the waste form matrix. Sulfide, in the slag cement is responsible for the chemical reduction in both formulations, and the waste form matrix microstructure is responsible for restricting transport of water, air (oxygen) and ions.

However, Portland and slag cement waste forms are porous and have pore volumes between 40 to 65% of the total volume. Most of the porosity is transmissive to water and gas (Phifer et al. 2006; Cantrell et al., 2016). Therefore, TcS, TcO_2 and technetium oxy-hydroxides are susceptible to re-oxidation when exposed to oxidizing environments (laboratory testing, field testing and shallow land disposal) (Almond et al. 2012; Langton and Almond 2013; Langton 2014; Langton 2015). Consequently, long-term performance with respect to both chemical and physical stabilization of redox sensitive contaminants present as precipitated nano/microparticles or sorbed ions in the cementitious waste form matrices requires mechanistic understanding, source term data, and sensitivity case modeling to evaluate potential long-term risks.

Some observations of 99-Tc geochemistry in reducing grout are provided below:

- Spectroscopic evidence showed that when Tc(IV) in a slag-cement was exposed to an oxidizing environment, it will convert to the more mobile Tc(VII) species within a short timeframe, 2.5 years (Allen et al. 1997; Lukens et al. 2005; Shuh et al. 2000).
- Reducing grout will reduce Tc(VII) in the absence of Na_2S or sodium dithionite when maintained in a reducing atmosphere (Shuh et al. 2000).
- Only trace concentrations of atmospheric oxygen (30 to 60 ppm O_2; Eh 120 mV) at the high pH levels of cementitious systems is required to maintain 99-Tc as Tc(VII) (Kaplan et al. 2011).
- Measured reduction capacity values for Cast Stone and Saltstone of about 800 μeq/g are in the realm of literature values and are consistent with theoretically based thermodynamic calculations (Um et al. 2015).
- Tc(VII) reductive precipitation with sulfur phases has an appreciably lower solubility than $TcO_2 \bullet x\text{H2O}$ phases (Pearce et al. 2018).

Unlike sulfide based reductants, increasing the reduction capacity of grout through the addition of Fe(0) is not effective in reducing pertechnetate or chromate in high pH solutions. However, Fe(0) can serve as an O_2 scavenger and retard oxidation of chemically reduced 99-Tc and Cr. Sn(II)-treated apatite and Sn(II) chloride showed very small capacities to remove 99-Tc from simulated Cast Stone pore water conditions, whereas a potassium-metal-sulfide (KMS) getter showed promise with extraordinarily high 99-Tc Kd values of 10^4 mL/g (Neeway et al. 2016; Mattigod et al. 2003; Qafoku et al. 2015).

2.2.2 Line 12 – Redox Getters for Improved Technetium Retention

Getters have long been proposed as additions to grout waste forms for the improvement of contaminant retention, with surprisingly limited testing (Mattigod et al. 2003; Pierce et al. 2010a; Qafoku et al. 2014). Targeting improved 99-Tc retention with redox getter additions would be achieved through the getter reducing the $Tc(VII)O_4^-$ to a low solubility Tc(IV)-oxide or -sulfide species. The getter could be added to the LAW prior to dry mix addition or included into the dry mix. Once incorporated into the grout waste form the 99-Tc would theoretically have increased stability and a slowed release. Resistance to long-term re-oxidation in this technology is imperative and requires evaluation.

This approach has shown evidence of success in waste forms fabricated from simulated LAW (Asmussen et al. 2017b), and in the immobilization of ground water (Duncan et al. 2009). Example candidate redox getters include tin-based (Levitskaia et al. 2016), sulfide-based (Neeway et al. 2016), and iron-based materials (see the discussion under Line 13 – Addition of Redox Getter, below).

2.2.3 Line 13 – Increasing Reduction Capacity of Cementitious Components to Increase the Amount of Oxygen Required for Complete Oxidation

Controlling technetium and chromium release through controlling the reduction potential (redox) of the bulk waste form can be enhanced by increasing the overall reduction capacity of the waste form. The result is expected to be extending the time frame of redox sensitive contaminant stabilization by increasing the overall waste oxygen consumption capacity. In Hanford Cast Stone and SRS Saltstone, Tc is present as pertechnetate, TcO^{-}_{4}, i.e., Tc^{7+} (Another redox sensitive contaminant in tank waste is chromate, CrO^{2-}_{4}, i.e., Cr^{6+}). These ions are chemically reduced to Tc^{4+} and Cr^{3+} and precipitated as TcS, TcO_2 or oxyhydroxides, and $Cr(OH)_3$ by sulfur in the slag component of the waste forms. Increasing the reduction capacity of the bulk waste form would slow re-oxidation and consequently solubilization of these phases in a near surface oxidizing environment. Additional reduction capacity can be provided to the waste form through increased slag content, Fe amendment, or incorporation of Sn_2Cl or other suitable materials. However, due to the porous nature of cement and slag waste forms and the presence of fast pathways, a discrete oxidation front has not been observed and once the surfaces of reductant additive are oxidized they are kinetically less effective in further chemical reduction reactions. It should also be noted that increasing the waste form reduction capacity does not directly improve the retention of non-redox sensitive anions (e.g. I^-). The species which contribute to reduction capacity (e.g. sulfides) may destabilize getters used for iodine retention (e.g. AgI) via substitution of the iodine or reduction of the supporting metal cation.

2.2.4 Line 14 – Iron Oxyhydroxide Treatment

The use of Fe in its zerovalent or oxyhydroxide forms as a low-cost option for 99-Tc sequestration from liquid nuclear waste streams has been studied previously in various waste compositions similar to LAW and in the presence of competitive Cr (Darab et al. 2007; Skomurski et al. 2010; Lee et al. 2016; Xiao et al. 2019; Emerson et al. 2020). The reduction of Tc(VII) to Tc(IV) occurs via the oxidation of Fe(0) or Fe(II) species that are added to the waste stream. The Fe source (simple Fe-oxides or ZVI) would be added directly to the LAW stream to sequester the 99-Tc followed by addition of the grout dry mix. A potential benefit of this approach is that during the oxidation of Fe the resulting reduced 99-Tc (and Cr) can be incorporated into the resulting Fe(III) oxide, which would add an additional stability barrier to release of the 99-Tc (Heald et al. 2012; Smith et al. 2016; Saslow et al. 2017b; Saslow et al. 2019b). In some of the available data on 99-Tc sequestration, $Fe(II)(OH)_2$ materials were used which have limited stability in aerobic environments. However, ZVI is aerobically stable. While evidence to date is promising no data on the subsequent leachability of Fe-Tc complexes from a grouted waste form is available. . However, due to the porous nature of cement and slag waste forms and the presence of fast pathways, a discrete oxidation front has not been observed and once the surfaces of reductant additive are oxidized they are kinetically less effective in further chemical reduction reactions. Field testing over time is required.

2.2.5 Line 15 – Use of Ion Exchange Resins as Getter for Incorporation into Waste Form

This retention approach for 99-Tc would use commercially available ion exchange resins to selectively capture anions, including pertechnetate (TcO_4^-) and use them for direct incorporation into grout. Documented ion exchange resins for 99-Tc capture include Purolite A530E and SuperLig 639 with Purolite A530E showing secondary selectivity for other contaminants such as IO_3^- (Valenta et al. 2010;

Saslow et al. 2019a). Initial work has been performed to identify formulations capable of solidifying ion exchange resins that were exposed to simulated Hanford wastes drawing on the previous work on resin solidification by the French Atomic Energy Commission (Neji et al. 2013; Neji et al. 2015).

While contaminant selectivity and preliminary stabilization studies of ion exchange resin in cementitious waste forms yield promising results, when and where to add the resin to the grout processing flow-sheet (before or after slurry fabrication, pre-treat vs. treat upon mixing to sequester the 99-Tc) remains unknown. The cost of incorporating ion exchange resin into grout formulations must be offset by long-term improvements to waste form performance/contaminant retention. As such, knowledge gaps pertaining to (1) resin impact on contaminant release from the cured waste form, (2) the stability of the selected resin in grout, and (3) the impact of dehydration/expansion processes on the resin in the cured waste form must be addressed

2.3 Engineered Barrier Technologies

2.3.1 Lines 17 and 18 – Sorbent Added to Backfill Around Waste Packages

Following closure of the IDF the migration of species away from the waste packages could be retarded through amendments of the backfill within the IDF. Amendment materials would be incorporated into the IDF backfill before being packed around the waste package. As infiltrating water moves contaminants (99-Tc, 129-I, nitrogen species) away from the waste form the contaminants would interact with the amendment and be sequestered onto the backfill through a sorption or precipitation processes. This behavior would likely be described in fate-and-transport models as a higher distribution coefficient, K_d, than is currently used in the IDF PA and would result in simulations showing longer 99-Tc retention in the IDF. The primary challenge with this technology is ensuring activity of the amendments is retained beyond the 500-year cap failure scenario in the IDF, particularly the maintenance of reducing capacity of the amendment with infiltrating oxygen. If successfully demonstrated, the approach could offer a low cost means of ensuring slowed 99-Tc release over long times.

Several material types have been identified that could serve as amendments. A summary of candidate materials have been fully reviewed elsewhere (Pearce et al. 2020). Specific promising amendments include:

- **Iron based minerals:** Naturally occurring iron minerals and artificially added iron (e.g., ZVI, a corroding steel container) can provide reducing sources within the backfill. Iron oxides have been shown to successfully sequester 99-Tc (Xiao et al. 2019) and 99-Tc will interact with corroding steel (Heald et al. 2012). 99-Tc/Fe-oxyhydroxides form in nature, especially wetlands, but the chemical conditions of formation are limited, and typically do not tolerate high concentrations of competing anions (Burke et al. 2006; Icenhower et al. 2010).
- **GAC:** Some activated carbons are impregnated with sulfur, which is a known reductant for 99-Tc (Pearce et al. 2018; Asmussen et al. 2019). The placement of sulfur impregnated GAC around the waste packages may provide a stable sulfide source to retard 99-Tc migration, but has not been proven experimentally.
- **Silicate Based Sulfur Impregnated Substrates:** Other sulfur/sulfide/thiosulfite, impregnated inorganic substrates have been developed for incorporation in cementitious waste forms and backfills (Langton et al. 2020).

2.3.2 Line 26 – Polysulfide Injection for Redox Control Followed by Phosphate Sequestration

Contaminant species which are redox sensitive can be manipulated within the subsurface to slow their mobility using injections to convert the contaminants to reduced (less-mobile) states that are incorporated into mineral phases. *In-situ* manipulation of redox conditions has been tested and implemented in bench-scale simulated Hanford conditions to reduce contaminant species. Calcium polysulfide has been demonstrated to reduce 99-Tc (Dresel et al. 2008; Saslow et al. 2018; Petersen et al. 2006). This treatment is followed by a calcium phosphate injection to promote the growth of apatite in the subsurface to sequester the reduced contaminants. Bench-scale experiments have shown that the combined application of calcium polysulfide and apatite in these experiments resulted in almost complete immobilization of uranium and 99-Tc with no reoxidation; however, the full duration of the reoxidation resistance is unknown.

2.4 Other Technology Approaches

2.4.1 Line 29 – Conversion of Non-Pertechnetate to Pertechnetate

Technetium Inventory and Speciation
The speciation of technetium in tank waste is vital to its management and disposition in grout waste forms. The predominant form of technetium in tank waste is pertechnetate ion, TcO_4^-. There is also a form of technetium in Hanford tank waste known as “non-pertechnetate” (Schroeder et al. 1995), which is believed to be predominantly a Tc(I) carbonyl moiety (Lukens et al. 2003; Lukens et al. 2004), although other forms may also exist (Levitskaia et al. 2015; Hall et al. 2015). However, the full speciation and distribution of technetium in Hanford tank waste is not known. Of the 177 tanks, only 10 have ever been tested for the presence of non-pertechnetate, and it is the predominant form (>60%) in five of those, with 0 to ~10% in the others (Rapko et al. 2013), but has been estimated in the remaining tanks (Serne et al. 2014). It is not known if the non-pertechnetate fraction in waste tanks changes over time but is most likely influenced by the presence of complexants in the tank waste and irradiation of the waste.

Historical Determination of Non-Pertechnetate Species
The method used to measure the non-pertechnetate fraction was by processing the waste sample through an ion exchange column that removed only pertechnetate; any remaining technetium in the effluent liquid was “non-pertechnetate.” But the purpose of the testing was not strictly to quantify the amount of non-pertechnetate, it was to test the effectiveness of various ion exchange resins and often to prepare samples for further decontamination testing and immobilization. There were several discrepancies in the data, where results from test to test with samples from the same tank yielded different results. Generally, if a waste tank sample contained <10% of total technetium as non-pertechnetate, no further testing or analysis was done to elucidate the exact amount. It is possible that these tanks actually have little or no non-pertechnetate, but there is no way to know. Most of the tanks that were found to contain >60% non-pertechnetate are “complex concentrate” tanks, or otherwise high organic tanks.

A specific method for quantifying non-pertechnetate has been developed that is compatible with test procedures used in 222-S Laboratory (DiPrete et al. 2016), but the method has not been implemented. Another method has also been developed that uses a special sensor to measure the Tc(I) moiety, but would not sense if other forms of non-pertechnetate exist (Bryan et al. 2017). More information on the topic of non-pertechnetate chemistry can be found in a summary report (Nash et al. 2017).

Impacts of Non-Pertechnetate Presence

The presence of non-pertechnetate may have significant impacts on potential use of a grout waste form if a significant inventory exists universally across the tanks. First, if technetium is to be removed to enable a grout waste form, the known removal methods have only been shown to be effective for the pertechnetate ion. No removal methods have been developed specifically for the non-pertechnetate form(s) and no methods have been developed to convert it to pertechnetate. Second, the long-term stability of technetium in grout waste forms requires that the technetium is originally present as pertechnetate in the waste when it is mixed with grout-forming materials. The grout immobilization relies on the reaction chemistry of ingredients in blast furnace slag to convert the Tc(VII) pertechnetate into an insoluble Tc(IV) species (Angus et al. 1985; Langton 1987; Allen et al. 1997). Once completely converted to Tc(IV), it is present as either an oxide or sulfide species that is integral to the grout matrix (Lukens et al. 2005; Um et al. 2011; Arai et al. 2015). That redox reaction of Tc(VII) to Tc(IV) is key to sequestering the ^{99}Tc in the matrix, thus it is not easily leached with water (Cantrell et al. 2016; Kaplan et al. 2011; Kaplan 2016). The assumptions for the SRS performance assessment (PA) is fundamentally based on the complete conversion of soluble Tc(VII) to the insoluble Tc(IV) during the initial formation of the grout waste form and is supported by experiments (SRR-CWDA-2013-00062). With Hanford waste that contains non-pertechnetate, it is not known if the non-pertechnetate species will undergo this conversion, but is unlikely because it is already in a reduced oxidation state (i.e., Tc(I) vs. Tc(IV)). Indeed, simulant testing of a grout waste form containing a synthesized Tc(I) carbonyl complex indicated a ~10× increase in observed diffusivity for a Tc(I) species compared with Tc(VII) (Serne et al. 2016b). It is key to the assumptions in this report that the 99-Tc speciation in tank waste is known and is present entirely as Tc(VII) pertechnetate. Although, the only exception to this is if the waste form is disposed off-site where a groundwater path is not plausible and the insoluble property of technetium in grout is not a fundamental assumption.

For potential onsite disposal of tank waste as a grout waste form, the quantification of the non-pertechnetate fraction of the Tc-inventory would assist with accurately projecting 99-Tc fractional release rates from the IDF with contributions from both pertechnetate and non-pertechnetate forms. The higher degree of confidence in the inventory splits between the two forms, and their corresponding release mechanisms, would ensure the risk associated with disposal of the non-pertechnetate inventory in the IDF can be accurately assessed.

Non-Pertechnetate Roadmap

If the use of a grout waste form for SLAW is pursued for onsite disposal, to the following steps are recommended to address the non-pertechnetate issue.

Recommendations:

1. Implement an analysis method in 222-S Laboratory to enable quantitation of the non-pertechnetate fraction down to 1% of the total technetium
    a. Measure the non-pertechnetate fraction in tanks considered for grout immobilization using the Sample and Send approach.
2. Calculate the impact of a partial non-pertechnetate inventory of 99-Tc contributing to the overall 99-Tc fractional release from the IDF in a PA-type simulation.
3. Produce grout waste forms containing non-pertechnetate and perform leach testing to determine if non-pertechnetate is immobilized by a grout waste form; measure the K_d and diffusivity in leach tests to compare to the single dataset currently available.

4. If calculations show that the fraction of non-pertechnetate present in the tanks of interest impacts the PA limits, develop a treatment method to convert non-pertechnetate to pertechnetate (may be combined with organic destruction methods, if feasible).

At this time, it is not recommended to pursue non-pertechnetate removal methods, since that would add another process step and result in another secondary waste stream. It would be more straightforward to destroy the non-pertechnetate and convert it to pertechnetate, followed by pertechnetate removal or immobilization. Similarly, additional characterization and synthesis of the non-pertechnetate species is needed if a simulant is required for testing.

3.0 NITRATE

3.1 Waste Form Improvement Technologies

To date, no examples of getter use for improved nitrate retention have been evaluated. The getter strategies for other species – iodine through solubility (nearly all nitrate salts are soluble) and technetium through redox (if reduced to nitrite the mobility is not improved) – are unlikely to be successful for improving nitrate retention in cementitious waste forms. The remaining candidate getter approaches for nitrate would be the presence of a media with an affinity for nitrate sorption or entrapment in a matrix phase of the grout. The primary challenge of these approaches would be the competition from other anions in LAW or the grout pore water within the waste form. Candidate materials for nitrate getters include carbon based materials and layered double hydroxide (LDH) phases.

3.1.1 Lines 11 – Carbon Based Getters

A review of nitrate sorbing materials is available and highlights that the highest sorption was attained through carbon-based materials in near neutral conditions (Bhatnagar and Sillanpää 2011). Carbon materials (e.g., activated carbons, hierarchal materials) are low cost and highly tailorable. Amberlite IRA 400 resin has been shown to effectively sorb nitrate and other oxyanions (Chabani et al. 2007).

However, several issues related to the suitability of carbon-based getters similar to GAC added in cementitious waste forms include (1) high concentration of nitrate in SLAW requires a relatively large amount of carbon sorbent, (2) vast anion competition in the concentrated LAW, (3) potential reaction between carbon and nitrates to form ammonia (observed in SRS Saltstone and resulting in specification of carbon burn-out fly ash), (4) nitrate loaded resin secondary waste disposal pathway must be specified, (5) Effectiveness of carbon based getters for nitrate in high sodium (3-7 Molar) highly caustic salt solution has not been demonstrated, and (6) performance of carbon based material included in the cementitious waste form is unknown as a function of time under disposal conditions.

3.1.2 Lines 14 – Layered Double Hydroxides

In this technology, cementitious or geopolymer formulations would be specifically tailored to promote the formation of LDH phases that are capable of sequestering and immobilizing nitrate from waste waters (Ivánová et al. 2018; Halajnia et al. 2016); it is noted that such phases also have the potential to sequester other anions such as iodide and pertechnetate. These phases are thermodynamically stable and compatible with other phases in these waste forms.

LDHs are composed of positively charged host layers and guest interlayer anions, which can be widely variable (Goh et al. 2008; Wang and O'Hare 2012; Theiss et al. 2014). The preparation, properties, and applications of LDH materials have been extensively studied (Rives 2001; Thomas and Daniel 2019).

LDHs associated with a range of cementitious materials are so-called AFm phases, a shorthand for a family of hydrated calcium aluminate phases structurally related to hydrocalumite and/or hydrotalcite occurring mainly in hydrated cement paste. Hydrocalumite is a major phase in SRS Saltstone. LDH phases that form in Ca-Al-Si-S-H cementitious systems and engineered LDH phases added to cement reagents used to stabilize DOE tank waste solutions have not been selective for radionuclide COCs because of competition for lattice sites from other anions such as sulfate and carbonate that are present at much higher concentrations in the waste solution (Kubilius 2018). Another important secondary phase in cementitious systems is the AFt phase ettringite. AFt and AFm phases are well known as secondary phases present in calcium sulfoaluminate cements (CSA), hybrid calcium aluminate cements (CAC)/hydrated lime/gypsum systems, and systems involving OPC blended with CSA, CAC, and gypsum. The compatibility of these phases with cementitious systems and their ability to sequester certain anions could be used to guide the design of alternative waste form formulations that promote the in-situ formation of these phases. An alternative approach could be to promote the formation of these phases in the LAW solution to form a slurry before addition of the grouting mix by addition of suitable reagents. A further alternative would be to include LDH phases as additives in the dry mix.

If successful, this technology could be relatively simple to implement since it essentially involves modifications of the grout formulation and/or addition of reagents prior to the addition of the dry mix. Testing would be required to define such formulations and to demonstrate an improvement on nitrate retention while still meeting other performance requirements. Since the nitrate concentrations in the LAW are relatively high (often several molar) the ability to form sufficient quantities of LDH phases with sufficient nitrate capacity would need to be demonstrated. The long-term stability of the LDH phase in the waste from would also need to be investigated.

3.2 Engineered Barrier Technologies

3.2.1 Line 16 – Iron Amended Backfill in IDF

Naturally occurring iron minerals and artificially added iron (e.g., ZVI, a corroding steel container) can provide reducing sources within the backfill, similar to other Fe-driven approaches described previously for 99-Tc (see Line 14 - Iron Oxyhydroxide Treatment in the 99-Tc section of this document). The use of ZVI could provide a reduction/sorption zone for nitrate in the near field around waste packages. This technology has been previously explored as part of the fiscal year 2018 DOE-RL deep vadose zone program at Pacific Northwest National Laboratory for the treatment of the Hanford perched water zone, which contains elevated concentrations of nitrate, technetium, and uranium (Saslow et al. 2018). ZVI, among other Fe sources, may be commercially purchased at low cost, facilitating the jump from laboratory to field scale demonstration with relative ease. However, the effectiveness of ZVI added as an amendment to the IDF backfill remains unknown, especially considering the sensitivity of this technology to changes in aqueous conditions (pH, E_h, water velocity/reactivity), and co-mingled contaminants (Liu and Wang2019). Understanding the reduction and conversion mechanism under changing field conditions is also necessary to address secondary product formation (NH_4^+, N_2) and long-term performance.

3.2.2 Line 18 – Gas Phase Bioremediation

This technology approach targets creating a reactive barrier for denitrification below the IDF. The use of gas-phase carbon amendments has been successfully demonstrated for degradation of perchlorate and nitrate in the vadose zone (Cai et al. 2010). Preliminary laboratory testing (Bagwell et al. 2018) has evaluated gas-phase bioremediation as an effective treatment to decrease the amount of nitrate present in the subsurface. If implemented to a theoretical nitrate plume below the IDF it could lead to a significant decrease in projected nitrate levels present in the subsurface. However, obvious challenges in implementation and effectiveness duration would need to be overcome, especially considering the need to have active treatment for more than 1000 years.

4.0 ORGANICS

4.1 Pretreatment

4.1.1 Line 4 – Contaminant Inventory Management Sample and Send Waste with RCRA Organics to Vitrification

See the technology description for Line 13 in the iodine section of this appendix.

4.1.2 Line 5 – Chemical Destruction of Organics Using Permanganate

The SRS Defense Waste Processing Facility (DWPF) is currently planning to implement an ambient temperature alkaline permanganate process for the oxidation of glycolate and this proposed approach is based on a similar process (Zamecnik et al. 2019; Siegfried et al. 2019; Nash and Siegfried 2020). Permanganate is a strong oxidant that reacts with a wide range of water soluble organic chemicals throughout the pH range including phenols, ketones, aldehydes, organic acids, glycols, sugars, cyanide, and hydrocarbons (Ladbury and Cullis 1958; Szammer et al. 1992; Chow and Leung 2019; Dash et al. 2009; Kuhn and Sontheimer 1979). Neither ammonia nor ammonium ion react with permanganate (Brandt et al. 2017; Xue and Chen 1999).

Permanganate should remediate most of 22 organic species that may exceed LDR limits for SLAW compiled recently (Bates et al. 2019, Table 3). Most of the species listed are in the categories shown above, especially phenols, ketones, and aldehydes. Pyridine and dichloromethane are notable exceptions. Both of these have been used in chemical research as solvents for permanganate, demonstrating their inertness to oxidation (Nace and Rieger 1970; Marques et al. 1998).

Permanganate is easily available as the sodium and potassium salts, with the sodium salt being more water soluble of the two. The cited DWPF studies of the process at pH 13 found that glycolate and formate were removed from solution within a few hours, and that nitrite did not react with the added permanganate. Excess permanganate and manganate oxidants at the end of processing could be removed from solution using added sulfite, as practiced for samples in the three test reports cited above for DWPF.

Separately, Hanford wastes and simulants were studied using added permanganate to precipitate and adsorb strontium and actinides (Duff et al. 2002; Wilmarth et al. 1999; Krot et al. 1998). The objective of using permanganate was to oxidize organic chelants in the waste that kept strontium and actinides soluble in the alkaline solution. In all cases the permanganate was reacting with organic chemicals already present in the waste, though Duff et al. 2002 added formate to caustic Hanford simulant. That simulant had no other reductants and formate worked well in reacting with the permanganate.

The extensive body of information for industrial waste and nuclear waste cited here show that permanganate is promising for destruction of certain organic chemicals in caustic sodium salt solutions at ambient temperatures. Reactions are relatively rapid (i.e., within hours). The DWPF application is at a high Technology Readiness Level and is to be applied at the SRS in the next 2 years. One drawback of the permanganate process is the formation of manganese precipitates (i.e., chemical solids). These reaction byproducts may necessitate filtration or decantation and disposal. Slurries require mixing to suspend solid particles and to keep them suspended during downstream processing. Consequently, managing the resulting slurry for grouting LAW treated in this way requires process design and instrumentation enhancements compared to the current SLAW solution solidification concept.

4.1.3 Line 6 – Chemical Destruction of Organics Using Hypochlorite

Chlorination has been used worldwide for decades for drinking water treatment and also wastewater treatment, with disinfection being the first priority (White 1986; Kuhn and Sontheimer 1979). Chlorine and the easier to handle hypochlorite salts react with a wide range of chemicals. However, this reactivity also has drawbacks. Chlorine is reactive with nitrite, which is an undesired target when other species are to be oxidized (Lee and von Gunten 2010). If active chlorine addition to an organic bearing aqueous stream is not carefully controlled, chlorinated organics such as trihalomethanes can form. Chloroform is one example of a regulated product that can be produced. Active chlorine will also react with ammonia to form chloramines. Chlorination of wastewater often uses sulfur dioxide or sulfite to remove residual active chlorine from the finished product (Severn 2005).

Lee and von Gunten 2010 compared chlorine performance against ozone, hydroxyl radicals created from hydrogen peroxide photolysis, chlorine dioxide, and ferrate for specific target organic species in water. Active chlorine often had poorer kinetics than the other oxidants outside of its ability to attack primary amines.

Overall, the use of active chlorine for the SLAW aqueous stream is not likely the best option for removal of organic content.

4.1.4 Line 7 – Chemical Destruction of Organics Using Ozone

Ozone gas is a powerful oxidant that must be generated at the point of use because of its instability. It is most often generated within a device applying a high voltage corona discharge to a stream of either air or oxygen gas (Kuhn and Sontheimer 1979; Klasson et al. 2002). The ozone-bearing gas can then be entrained in an aqueous liquid to be treated. The chapter provided by Hoigne and Bader to Kuhn and Sontheimer 1979 lists a wide range of organic chemicals that are remediated, often in the pH range of 5 to 9. Methylmercury is one of the chemicals that Hoigne and Bader reported as being reduced to inorganic mercury. Organic feed levels in the 100 mg/L carbon range are reported.

One special benefit of ozone is that it adds only oxygen to a waste stream, thus it does not increase the load of solids-forming elements to waste. On the cautionary side, ozone is very toxic such that exposure to 1% ozone in air for less than a minute can be fatal (Rice et al. 1986). It will also degrade organic polymers, notably rubbers. In addition, emissions of unused ozone, such as the gas not absorbed in the treatment process, are subject to environmental regulation. Means to reduce ozone emissions are available as part of process design.

In the last 10 years an ozone process for organic remediation has been examined at the SRS (Ketusky et al. 2012; Ketusky 2018). A tank cleaning option with oxalic acid would produce much spent oxalate, and the references cited here show that ozone was able to treat 1 wt% aqueous oxalate. In addition, the fire safety of ozone and hydrogen was examined (Osterburg et al. 2013).

Winters 1981 reports 80 to 99% removal of 0.03 M EDTA from a Hanford simulant in 4-6 hours at varied pH values from 4.5 to 14 using ozone. Nitrite started at about 1.35 M, and most of that was oxidized.

Since nitrite is an undesired target, lower nitrite feeds should do much better than these results. Runs below pH 4.5 were also reported, but performance was not as good.

Delegard et al. 1993 demonstrated organic species oxidation with ozone using both simulated and actual Hanford tank 241-SY-101. High shear agitation was found to be essential and effective in utilization of the supplied ozone. About 90% of organic carbon representing many species was oxidized, and the destruction of organic complexants was indicated by precipitation of calcium, nickel, strontium, and americium. Oxalate was found to be refractory to ozone oxidation.

White 1999 lists industrial wastewater plants operating in the mid-1980s along with comments on experiences with the use of ozone. One problem mentioned in this work was nitrite in the waste, which is an undesired consumer of ozone. In addition, good gas-liquid contactor design is essential for the success of the process.

Ozone should be considered in cases where the total organic carbon is low (on the order of 10 of mg/L) but of high regulatory concern. Nitrite would make ozone processing difficult as noted above. Optimal pH is neutral to mildly alkaline. Residual ozone in the liquid phase is short lived (an hour or less) and thus should have minimal downstream impact, such as in grout chemistry.

4.1.5 Line 12 – Electron Beam Exposure for Organic Destruction

This process uses an electron beam to irradiate the LAW, destroying the regulated organic compounds. The waste would be dosed with the electrons, producing free radicals in solution that break down covalent bonds in the organic chemicals, converting them to less toxic species. Continued irradiation can break down the organics to carbon dioxide, but this is not common due to the very high dose needed. The irradiated LAW would then be mixed with grout forming chemicals and immobilized.

No reports of testing on this concept for waste streams similar to LAW have been found. Electron beam irradiators are very common and available from multiple vendors and are widely used for a range of applications. Testing has shown destruction of many organic chemicals in groundwater and industrial wastewater (Bibler 1993; Duarte et al. 2004), with the degradation products being lower molecular weight alcohols or phenolic compounds (Lin et al. 1995). Related applications are currently in use for treating municipal and industrial wastewater. Most applications are at near neutral pH, and competition and interference from many species in tank waste are likely, including the high pH, carbonate, nitrite, and dissolved metal ions. Partially destroyed organics are another possible interferant (Cooper et al. 2002). The effect on other COCs is unknown but would generally be expected to cause reduction of metal ions to a lower valence state (Djouider et al. 2018), perhaps causing precipitation of Cr, Hg, and 99-Tc. Testing of this technology would be relatively inexpensive, and very large-scale systems are commercially available. A proof-of-principle test would be needed to show if it is effective, and subsequent testing to determine dose and time needed for destruction of the intended contaminants.

4.1.6 Line 14 – Evaporation to Remove Organics

Removal of regulated organic compounds from a SLAW liquid stream to be grouted may be required before land disposal. The practicality of removing such compounds by evaporation is the option examined here. Evaporators are commonly used for waste concentration at both Hanford and SRS, but examination of organics that might follow the condensate (overhead) stream is an important part of flowsheet development. The process considered here would also be referred to as “steam stripping” or "steam distillation" (Hassan and Timberlake 1992). The process is very common in the chemical and petroleum industry for treating aqueous waste streams for organic chemical removal. The key to the separation, though, is that the relative volatility of the organic chemicals to be removed must exceed unity. If this is not the case, the evaporation process would remove water and would concentrate the organic chemical in the aqueous bottoms.

Lindberg et al. 2019 reviewed the case of regulated organic chemicals in the Hanford nuclear waste system and provided Henry's Law constants for specific organic COCs. Henry's Law constants (H) can be used to calculate relative volatility. The smaller the value of H, the more volatile the compound with respect to water. Table 17 of the Lindberg reference shows methanol having an H of 199 mol/atm*kg, bounding all other compounds except phthalates. Phthalates are noted to likely be an artifact of sample storage, thus not likely present at levels of concern in Hanford waste. Bennett 1929 reports that the relative volatility of methanol in water is 13.6 (unitless, and exceeding 1.0) at 370 K, so all compounds in Table 17 would be removed by evaporation except for the phthalates if present. Separately, Abramowitz et al. 2020 obtained literature values of the Henry's Law constants for acetonitrile and acrylonitrile as functions of temperature. The values indicate high volatilities with respect to water, allowing these two chemicals to be steam stripped from SLAW.

The overall benefit of evaporation is that nuclear waste evaporation has been a common unit operation, and facilities including the 242-A Evaporator will likely be available for this organic removal need. Organics of concern identified so far will theoretically follow the condensate given their high relative volatility (sufficiently low Henry's Law constants). If other organic COCs are identified, their volatility with respect to water will have to be evaluated. It should be noted that most of the wastes in the Hanford double-shell tanks have been through some level of evaporation previously and LDR organics may still be present in these wastes. The prior 242-A campaigns can be used as an initial assessment metric for the likely success of an evaporation approach.

5.0 MODIFICATION OF BULK PROPERTIES

5.1 Formulation Design

The current Cast Stone formulation is a modification of the SRS Saltstone waste form which was designed to be pumpable for disposal in large concrete engineered barriers at the SRS. The down-selection of the current Cast Stone (47 wt% blast furnace slag, 45 wt% class f fly ash and 8 wt% OPC) as a baseline formulation for use with Hanford wastes was performed by CH2M Hill at the 222-S Laboratory (Lockrem 2005b). The screening effort was small and only considered four formulations: the current Cast Stone along with three formulations containing OPC, fly ash and a mixture of Indian red pottery clay or attapulgite clay. These formulations were based on prior development of the Hanford grout vaults. The selected formulation had an immediate advantage over the other formulations as it was the only one to contain blast furnace slag, which facilitates passing the toxicity characteristic leaching procedure requirement.

The current SLAW Cast Stone concept is to solidify the waste and package the resulting waste form in steel containers which will be transported and disposed in the Hanford IDF or an offsite disposal facility with no groundwater pathway. The waste form requirements for each of these options are different than those for which the current Cast Stone formulation was designed. While Cast Stone for LAW has shown to qualify for nearly all waste acceptance criteria testing (Westsik et al. 2013) and could be utilized initially in the SLAW mission (especially if coupled with technologies in this report), areas for improvements in performance, processing and economics exist. Consequently, redesign of the SLAW/LAW cementitious waste form is expected to result in benefits for both the IDF and the offsite disposal options.

Improving/redesign of the current Cast Stone waste form to reduce contaminant release and physical/chemical alteration over time is warranted to support disposal in the IDF and is expected to address one or more for the following objectives:

- Improve chemical stabilization of selected contaminants (e.g., 129-I and 99-Tc).
- Reduce transmissive porosity for moisture and gas transport thereby reducing soluble ion transport and oxidation/solubilization of redox stabilized contaminants.
- Improve physical containment/microencapsulation of all contaminants.
- Improved resistance to chemical alteration by reducing exposure to external chemicals (e.g., CO_2, H_2O, SO_4^{2-}) and microbial degradation which is a function of porosity (i.e., surface area available for colonization and organic acid attack).

Technology for designing cement waste forms for IDF based on chemical stabilization include:

- **In-situ stabilization.** Incorporation of COCs in crystal structures resulting from hydration of commercially available cements and supplementary cementitious materials.
    - Incorporation of COCs in low solubility cement-water reaction products (e.g., 99-Tc substituted AFt or AFm phases or Sr substituted calcium silicate hydrate of portlandite).
    - Incorporation of COCs in phases that precipitate the waste form (e.g., TcO_2, HgS).
- **Stabilization by chemical addition.** Stabilization of COCs by chemicals added to waste or cement reagents to form low solubility phases.
- **Stabilization by getter addition.** Reaction of COCs with “getters” (substrates carrying reactants) blended with the cementitious materials or premixed with the waste.

Technology for designing hydrated cement waste forms for disposal at the IDF based on enhanced waste form microstructure/low porosity include:

- Reducing the water to cementitious material ratio by increasing the amount of cementitious material in the mix
- Reducing the water to cementitious material ratio by reducing the amount of water in the waste (evaporation)
- Optimizing the particle size and size distribution of the solid cementitious reagents to enhance particle packing in the waste form.

Additional needs include (1) cement waste form-specific test protocols to provide a more realistic evaluation of exposure conditions and/or (2) logic that better predicts actual performance using the current testing protocols for contaminant release.

Issues with each of the chemical and physical technology opportunities are discussed below. The greatest opportunity is expected to be realized by a combination of both chemical and physical waste form enhancements.

5.1.1 Line 4 – Porosity Reduction: Lower the Water to Cement Ratio

The potential to reduce soluble ion leaching by "densifying" the matrix via reducing the water to dry cementitious materials ratio (w/cm) in the Cast Stone has been demonstrated by (Cozzi et al. 2015; Westsik et al. 2013). This work was focused on a pumpable/flowable waste form rather than a containerized waste form. Testing formulations with an even lower w/cm is expected to result in lower porosities provided that the resulting waste form is processable and does not entrap air. Lower w/cm can be achieved by:

- Increasing the amount of cementitious reagent blend per unit mass of waste form with a given water content. For a given contaminant inventory, this approach may distribute the contaminants over a larger footprint which may be a positive factor for ground water modeling projections (dependent on model). Issues include optimizing container filling, an increase in the waste form disposal volume and an increase in the cement material blend reagent cost. Testing would determine whether lowering the w/cm from 0.4 to 0.25 or less by adding additional reagents significantly reduces leaching of the COCs.
- Concentrating the amount of salt in the aqueous waste stream by retrieval and/or evaporation. This approach may result in a lower waste form total volume and concentrate COCs in a smaller footprint which may have a negative factor in modeling projections. Potential issues with this approach could include difficulty in managing and solidifying elevated temperature caustic salt solutions and/or managing precipitated salts and scaling from as concentrated salts cool.

5.1.2 Line 5 – Particle Size Optimization: Lower water to cement ratio

Optimizing the particle size and particle size distribution of the solid cementitious reagents to enhance particle packing in the waste form is an approach for densifying the waste form thereby reducing porosity. This approach will result in also lowering the water per unit volume compared to an unoptimized particle size distribution mixture. This technique is one method of enhancing workability (rheology) and lowering the w/cm ratio.

The magnitude and consequences of optimizing the micron and submicron particle sizes and size distributions that make up cementitious materials is unknown for a SLAW form.

5.1.3 Line 6 – Optimize Cast Stone Formulation

See the discussion under Formulation Design at the beginning of this section.

5.1.4 Line 8 – Geopolymers

This technology involves the development of waste forms that are based on geopolymers or other alkali-activated cements. These are cement-like materials made by alkali-activated polymerization of aluminosilicates (Provis and Bernal 2014; Pacheco-Torgal et al. 2015). They generally do not contain any OPC, which is the primary ingredient in standard grouts. The basic process is simple and similar to that used for conventional OPC grouts and involves mixing of dry powders with an aqueous activator solution, typically at ambient temperature. For waste forms for the treatment of alkaline waste such as Hanford LAW streams, the waste itself can often function as the alkali activator, either alone or with suitable amendments (Gong et al. 2006; Gong et al. 2011; Gong et al. 2012; Gong et al. 2014; Gong et al. 2015; Papathanassiu et al. 2013; Papathanassiu et al. 2014; Xu et al. 2014a; Xu et al. 2014b). Such waste forms (e.g., DuraLith) have been developed and tested for Hanford WTP secondary wastes and LAW streams (Gong et al. 2006, Gong et al. 2011, Gong et al. 2012; Xu et al. 2014a; Xu et al. 2014b) as well as for SRS salt wastes (Gong et al. 2014; Gong et al. 2015; Papathanassiu et al. 2013; Papathanassiu et al. 2014). The latter so-called “cement-free saltstone” formulations are presently planned for full-scale implementation at the SRS Saltstone Facility.

Geopolymer waste forms developed for Hanford secondary waste have been tested with technetium and tested up to engineering scale (Josephson et al. 2011), and, even with limited optimization they have demonstrated comparable performance to Cast Stone (Mattigod et al. 2011). It seems likely, therefore, that further improvements in waste form performance should be possible. Testing would be required to assess whether such potential improvements can be realized. The process would be relatively simple to implement since it is very similar to the Cast Stone baseline and employs commonly available materials.

5.1.5 Line 9 – Mineral Incorporation

Mineral formation in cementitious waste forms is a direct product of the dry ingredients used during mixing and the composition of the liquid waste stream being solidified. Mineral incorporation of contaminants is a known and widely studied immobilization pathway for COCs in nuclear waste and in the environment. Calcium sulfoaluminate hydrate (CASH)-based matrices made from CSA or mixture of ingredients resulting in a similar matrix mineralogy have been shown to generate mineral phases capable of incorporating specific contaminants or increasing contaminant immobilization interactions (e.g., sorption) with the matrix (Chrysochoou and Dermatas 2006; Fernández-Martínez et al. 2008). For example, ettringite, commonly formed in CASH cements, has been shown to incorporate RCRA metal and radionuclide cations and anions into its structure (Gougar et al. 1996). In more recent studies, ettringite has been proposed as a natural pathway for immobilizing COCs to the Hanford Site TcO_4^-, as well as IO_3^- and CrO_4^{2-} (Aimoz et al. 2012a; Aimoz et al. 2012b; Um et al. 2017; Guo et al. 2020). Given the mineralogical evolution of the cement matrix as cementitious waste forms age, the ability to facilitate controlled early mineral formation through long-term mineral growth could provide a valuable pathway towards integrating a natural contaminant barrier into these waste forms. However, success in LAW of this approach has yet to be demonstrated. To this end, the unknown long-term mineral growth and impact on contaminant immobilization remains to be understood, especially while optimizing contaminant immobilization without straining other physiochemical properties influenced by mineral growth (e.g., late ettringite growth that causes cracking in waste forms).

5.1.6 Line 15 – Grout Vault Historical

In the initial development of the Hanford Grout Vault program and down-selection of Cast Stone, research was performed to identify candidate formulations for the immobilization of Hanford wastes (Serne et al. 1992; Lockrem 2005b). The data supporting these efforts should be summarized to provide information on candidate formulations which may be proposed for cost-efficient/improved formulations for Hanford LAW. The criteria for these formulations were different than the needs today, and formulations that meet current requirements may have been cast aside at the time because of characteristics that are no longer relevant. Doing so would also facilitate a link between prior Hanford grout development and improvements to future programs.

5.2 Pretreatment

5.2.1 Line 17 – SLAW Evaporation to Decrease Water Content and Waste Volume

In this technology, pretreatment of the LAW stream by evaporation would be employed prior to grouting in order to decrease the water content thereby potentially increasing the waste loading, decreasing the volume of waste form for disposal, and improving the waste form properties. There is also the potential for beneficial removal of some of the LDR organics during evaporation as discussed above. The waste loading, and therefore the volume of waste form produced per unit volume of waste treated, can be limited by the water in the waste or by the dissolved solids in the waste. In the former case, the loading and volume reduction can be increased by removing water from the waste stream by evaporation prior to stabilization. This process was employed in the development of geopolymer waste forms for the treatment of Hanford secondary wastes (Gong et al. 2006; Gong et al. 2011; Gong et al. 2012), where evaporation was used to vary the waste solids loading from about 3 to 20 wt%. The approach has also been examined for Cast Stone (Fox et al. 2014; Westsik 2013). In addition, it is well known that the water to dry mix ratio (w/dm) is a critical formulation parameter for cementitious and alkali-activated cement systems, affecting flow and rheological properties as well as cured properties. In particular, as the w/dm ratio is decreased, compressive strength typically increases and porosity and permeability typically decrease. There is therefore also the potential to improve the retardation of release of the COCs from the waste form by this approach. Furthermore, since this improvement would be achieved primarily via reductions in porosity and permeability, it should accrue to all COCs and have good longevity.

Evaporation of LAW is a standard and well understood process at Hanford and evaporators are incorporated into both the WTP Pretreatment facility and the EMF as well as the Effluent Treatment Facility. The WTP Pretreatment facility evaporator could be used to support this approach with few process changes. However, at present, the WTP EMF evaporator handles recycle streams from LAW vitrification, not LAW, so during direct-feed LAW operations, process modifications or a new evaporator facility would be required. For some tank wastes and high concentration factors, there is the possibility that the waste form radiological classification could be affected, which would need to be addressed. Testing would be required to develop appropriate waste form formulations, concentration factors, and the process operating envelope.

5.3 Engineered Barriers

5.3.1 Lines 19-20 – Waste Form Size Impacts and Resulting Redox Control

Waste form packages being considered for grouted waste that will be placed in the IDF range from 55-gal drums to B-25 boxes or intermodal containers. Increasing the waste form size could impart several benefits to contaminant retention. Two specific benefits are:

- **Redox Control:** One of the primary unknowns with grout waste form disposal at the IDF is the evolution of redox conditions within the waste form. Reducing conditions are imperative for maintaining sufficient retention of redox sensitive contaminants (e.g., 99-Tc, Cr, U). The reducing environment within a slag-containing waste form is slowly lost over time with the migration of excess oxygen into the waste form (coupled with other interactions with near-field species). A larger waste form has less surface area per volume of waste for oxygen to diffusion. Hence, larger waste forms retain reducing conditions over a larger portion of the contaminant inventory compared to smaller waste forms with equal inventory. This situation is best represented by the disposal vaults at SRS where a shrinking core model (e.g., moving smeared versus discrete oxidation front) is used to define conditions within the waste form monoliths over time (Kaplan et al. 2011). Preliminary assessments of waste form size impacts in the IDF have been carried out (unpublished). Lysimeter tests have previously provided information on this process (Langton 1988) and are ongoing at Hanford (Bacon et al. 2018). Improvements in accurately measuring reducing conditions of porous waste forms and the relationship of redox measurement to oxidation of redox sensitive contaminants in the laboratory and in disposal conditions samples is required beyond the traditional methods (Almond et al. 2012; Um et al. 2015).

- **Migration Pathway:** Increasing the waste form size also increases the length of the migratory pathway for contaminants and, for a given volume of waste reduces to surface area for diffusion. Both changes decrease the rate that contaminants move from the waste form to the environment. An important caveat to this assertion is that rapid that rapid transport pathways (e.g., cracks) need to be minimized.

5.3.2 Line 21 – Macroencapsulation Within or Around Waste Packages

Concrete culverts, cast-in-place reinforced concrete diffusion/isolation barriers structures are used at DOE and commercial radioactive waste disposal sites, e.g., SRS Solid Waste Disposal Facility, Idaho National Laboratory, Hanford Environmental Restoration Disposal Facility, and Waste Control Specialists (Nelson and Covert 2010; WCS 2016; WCS 2015). Reinforced concrete containers are used in Europe as primary waste containers and overpacks (IAEA 2002; IAEA 2009). Concrete mix design technology that combines features from these applications is mature and includes balancing very low water to cementitious materials ratio (<0.25), particle size optimization to enhance densification, multi-scale reinforcement to provide fracture toughness and tensile/flex strength, high-early strength for rapid fabrication, aggregate specified for application, and pre- or post-tensioning to achieve desired results. The most common cement matrices are Portland cement with and without slag cement (microstructure modification) and supplementary cementitious materials. However, other cements (ceramic cement or rapid set CSA) may provide specific benefits.

This approach would encapsulate individual containers or a group/stack of waste form containers in the IDF in concrete or other cementitious material to impede migration of water and contaminants. For the most part, strength of the encapsulation grout is not an issue but does correlate to porosity and is therefore used as an initial indicator of diffusion barrier and advective transport performance. Installation and quality control of grout layers placed between waste packages once they are in the IDF needs to be developed and confirmed. Rework is not desirable.

Prior to design and testing of special concrete barriers, PA modeling (parameters and assumptions about long-term performance) is required to identify hydraulic conductivity values that result in significant improvement in performance (e.g., lower and delay peak fractional release of contaminants). These values can then be used as specifications for design of concrete primary containers, overpacks, or IDF backfills for effective evaluation. A basis for long-term container properties and aging mechanisms is needed for defensible PA modeling.

Also see the technology description for Line 29 in the iodine section of this appendix for a description of an amended concrete barrier.

5.3.3 Line 22 – HDPE Containers or Landfill Cover

This concept is to use high density polyethylene (HDPE) as a barrier to groundwater contact with and contaminant release from a buried grout waste form. There are two options considered: (1) a slurry of grout formers and LAW could be poured into HDPE containers (or HDPE-lined steel containers) and the containers placed in the disposal facility, or (2) the grout slurry is poured into other containers (e.g., steel, concrete) and then a HDPE cap placed over the filled containers in the disposal facility. HDPE is selected because it is impermeable and resistant to environmental degradation, which lengthens its ability to function as a hydraulic barrier. High integrity containers made of HDPE are currently approved for use for disposing some waste types in the IDF waste burial ground (WHC-EP-0063), and the cell liners at the Environmental Restoration Disposal Facility are made of HDPE (Teachout 2018). HDPE can completely block advective and/or diffusive contaminant transport until embedded antioxidants are eventually depleted. A 2-mm thick geomembrane is estimated, based on testing and Arrhenius modeling, to have a service life of approximately 2000 years before failure due to oxidation and leachate exposure (Tian et al. 2017). If the barrier is above the waste and in contact with only infiltration water and not leachate, the barrier should last even longer. Further, a thicker HDPE layer should further delay oxidation and the onset of degraded performance. Challenges to this approach are demonstrating quality control in material manufacturing and placement to ensure defects are eliminated or too infrequent to be significant, and accurately forecasting material oxidation and thus service life through accelerated laboratory testing. This would be a significantly longer assumption of barrier integrity than typically used for HDPE containers, so significant material science work to accurately project service life would be needed. PA calculations would be needed, based on the approach, design, and assumptions.

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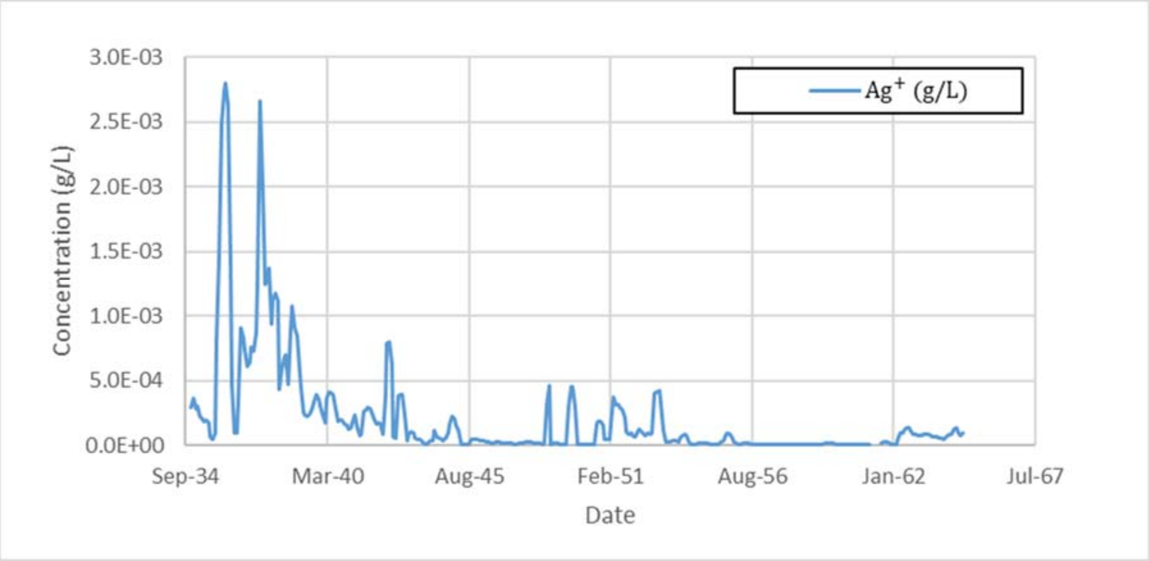
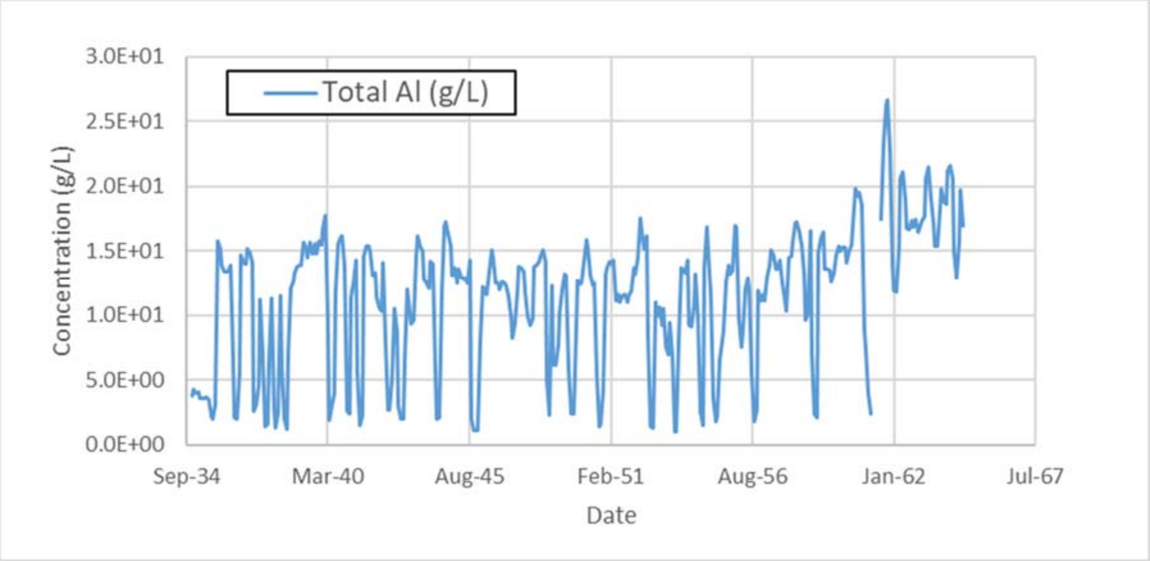
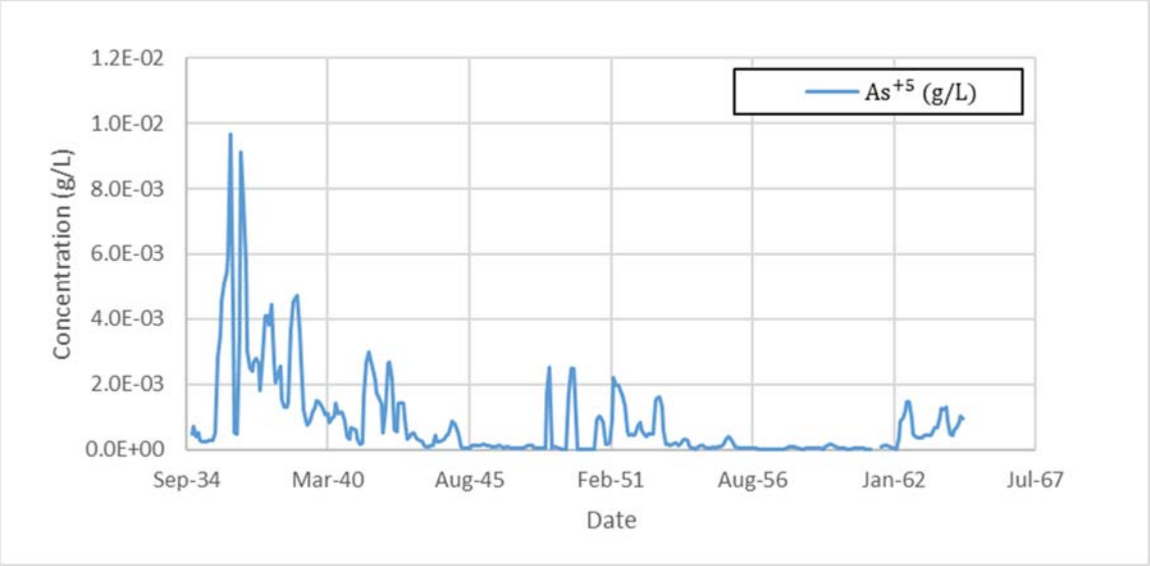
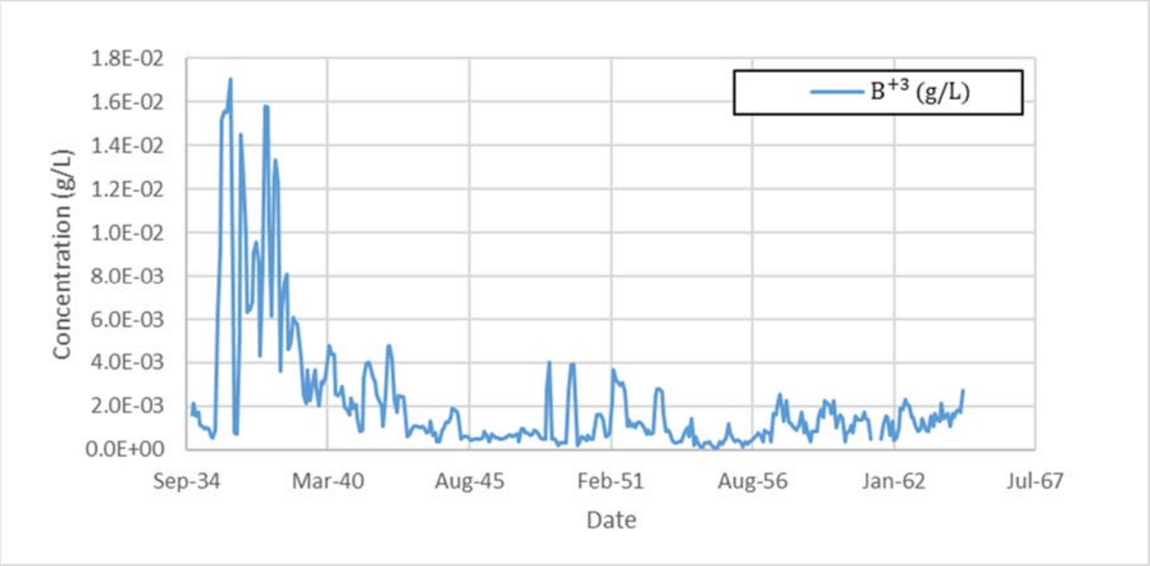
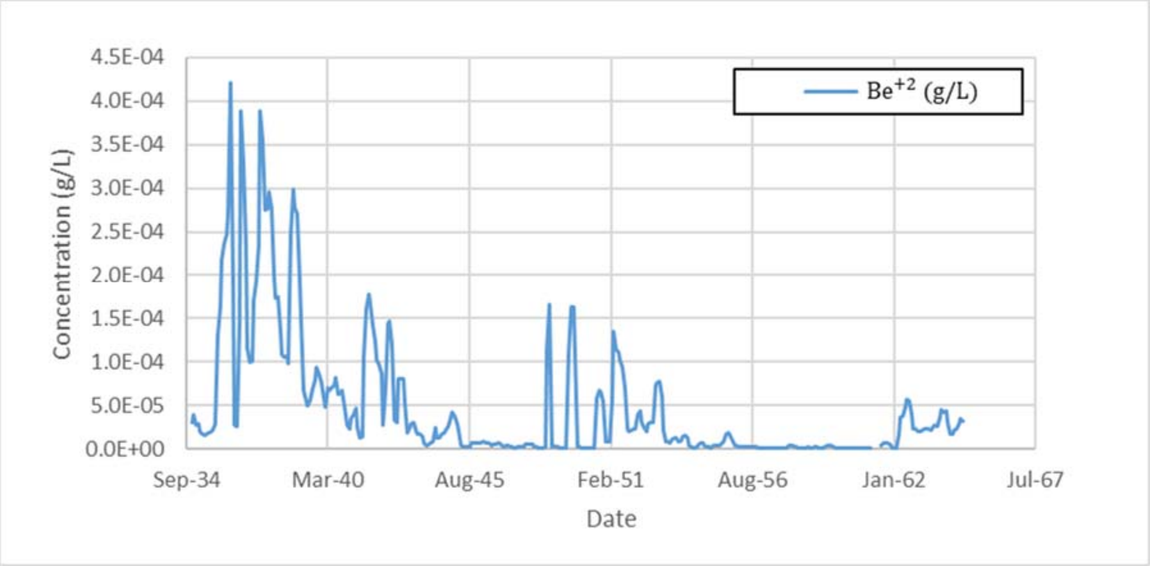
APPENDIX B
FEED VECTORS DISSOLVED NON-RADIOLOGICAL CONSTITUENT

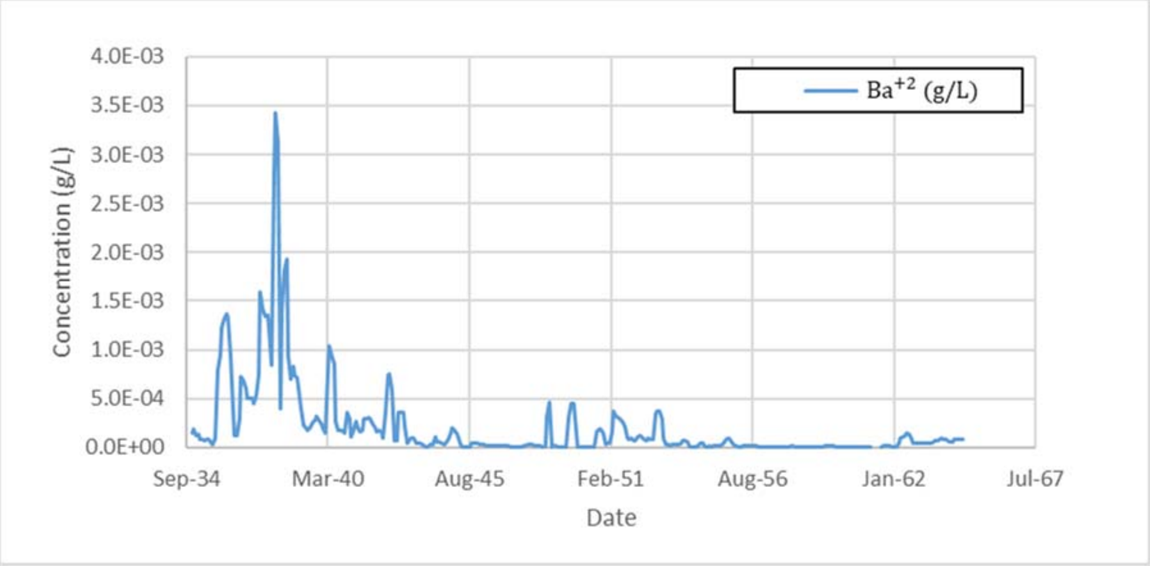

Ag$^+$ (g/L)
Concentration (g/L)
3.0E-03
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

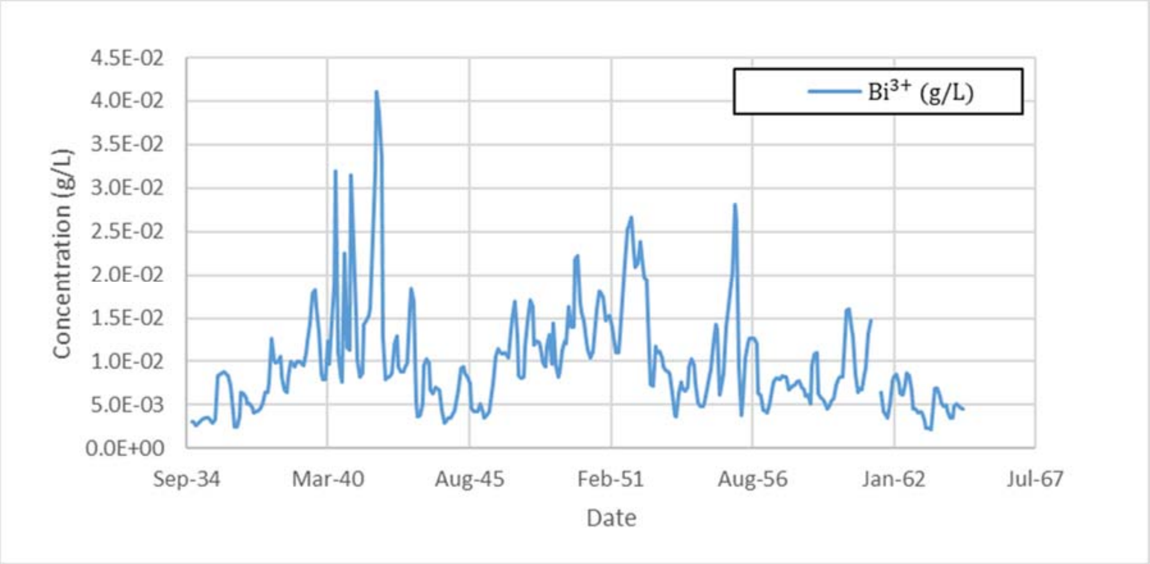

Total Al (g/L)
Concentration (g/L)
3.0E+01
2.5E+01
2.0E+01
1.5E+01
1.0E+01
5.0E+00
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

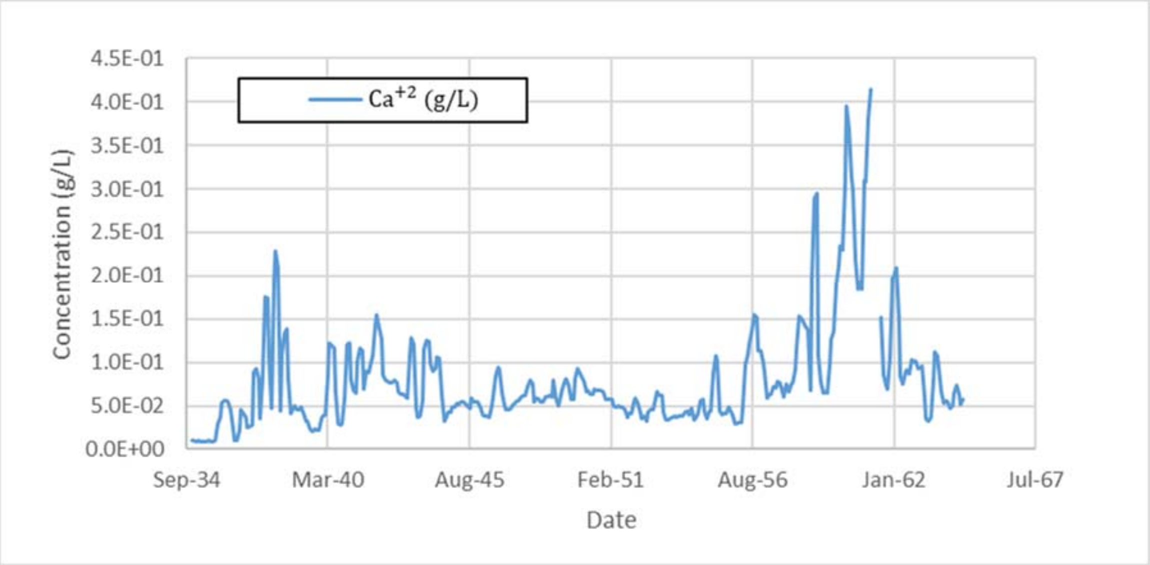

1.2E-02
1.0E-02
8.0E-03
6.0E-03
4.0E-03
2.0E-03
0.0E+00
Concentration (g/L)
As^{+5} (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

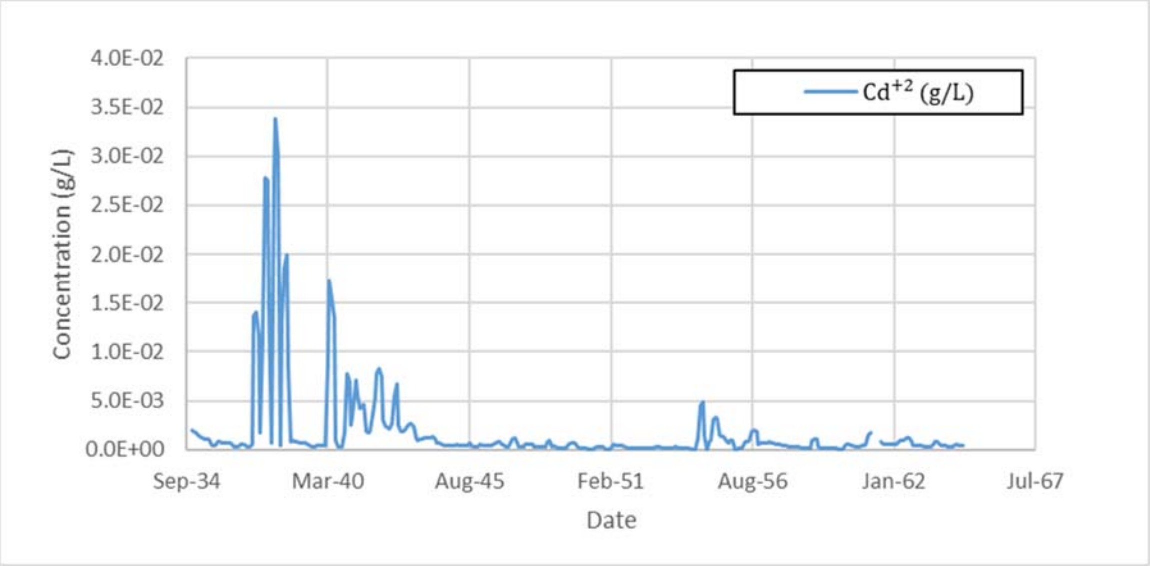

1.8E-02
1.6E-02
1.4E-02
1.2E-02
1.0E-02
8.0E-03
6.0E-03
4.0E-03
2.0E-03
0.0E+00
Concentration (g/L)
B^{+3} (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

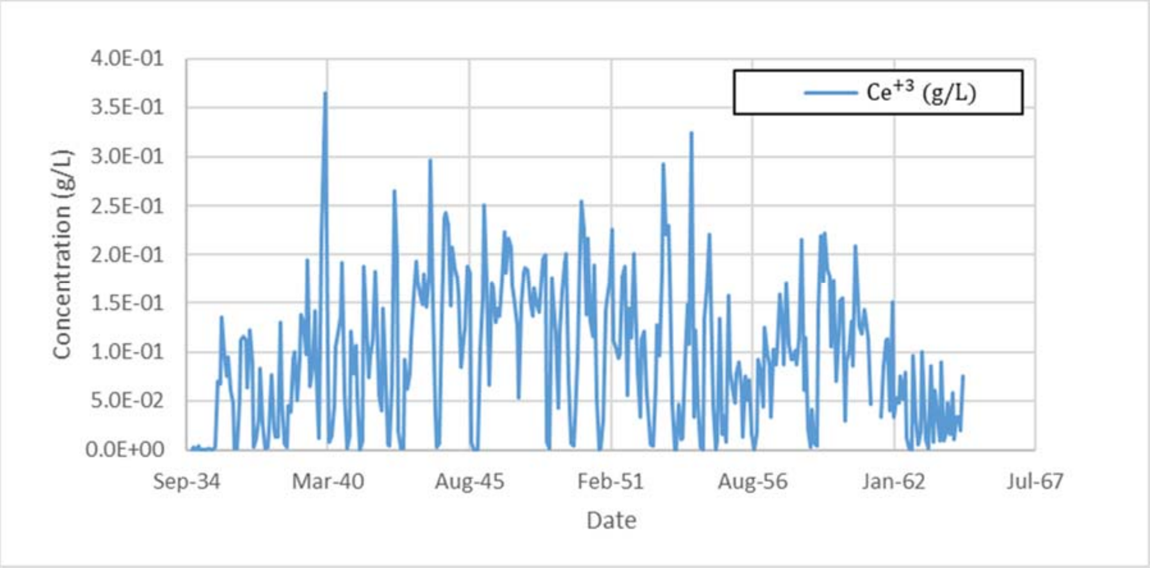

4.5E-04
4.0E-04
3.5E-04
3.0E-04
2.5E-04
2.0E-04
1.5E-04
1.0E-04
5.0E-05
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Be^{+2} (g/L)

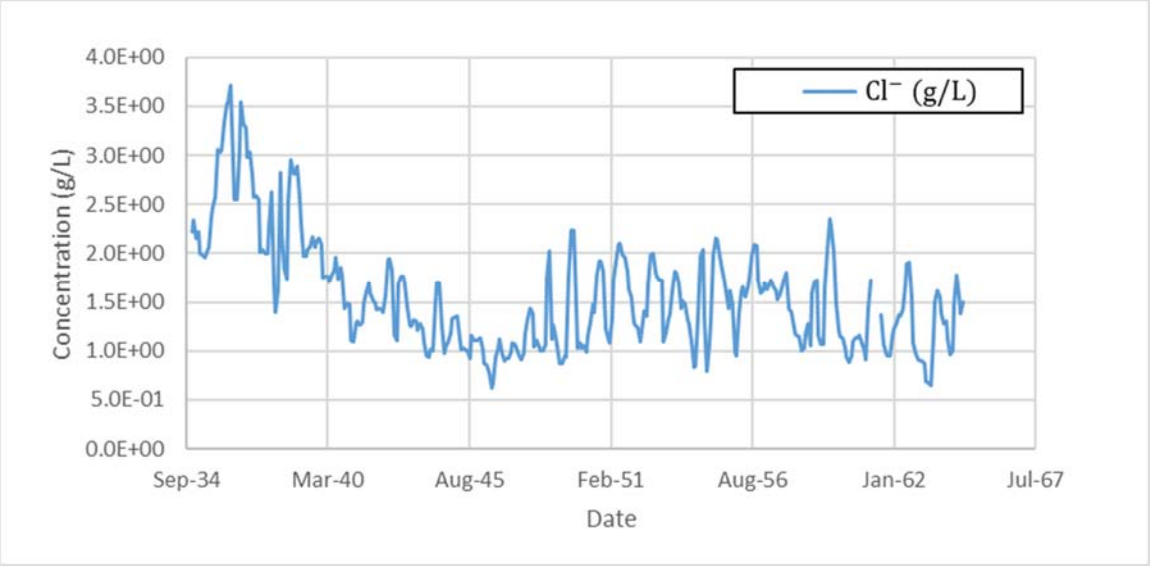

4.0E-03
3.5E-03
3.0E-03
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Ba^{+2} (g/L)

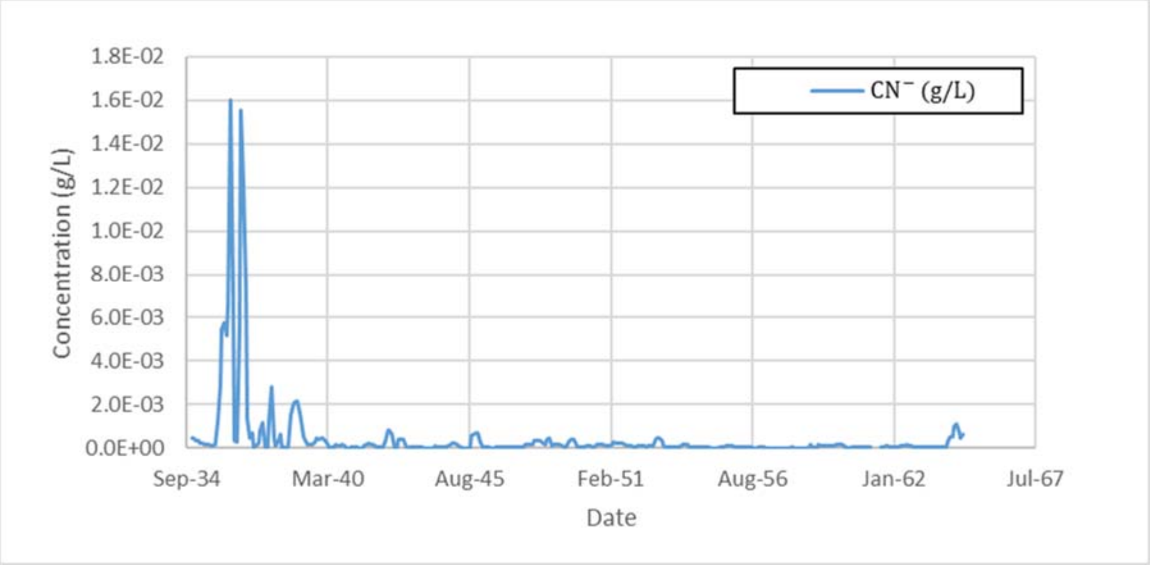

Bi^{3+} (g/L)
Concentration (g/L)
4.5E-02
4.0E-02
3.5E-02
3.0E-02
2.5E-02
2.0E-02
1.5E-02
1.0E-02
5.0E-03
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

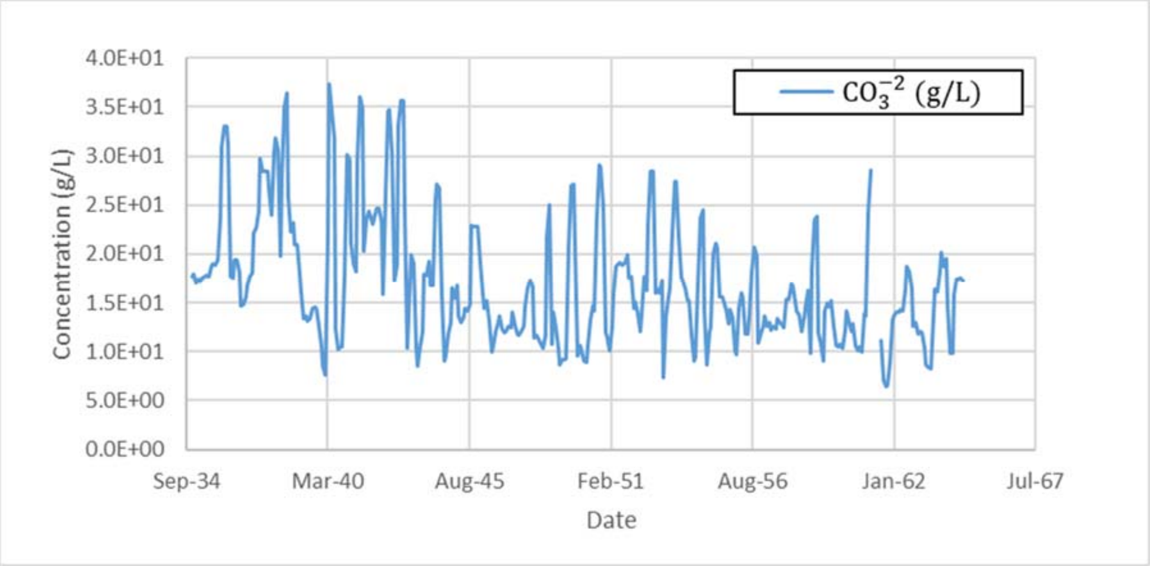

Ca^{+2} (g/L)
Concentration (g/L)
4.5E-01
4.0E-01
3.5E-01
3.0E-01
2.5E-01
2.0E-01
1.5E-01
1.0E-01
5.0E-02
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

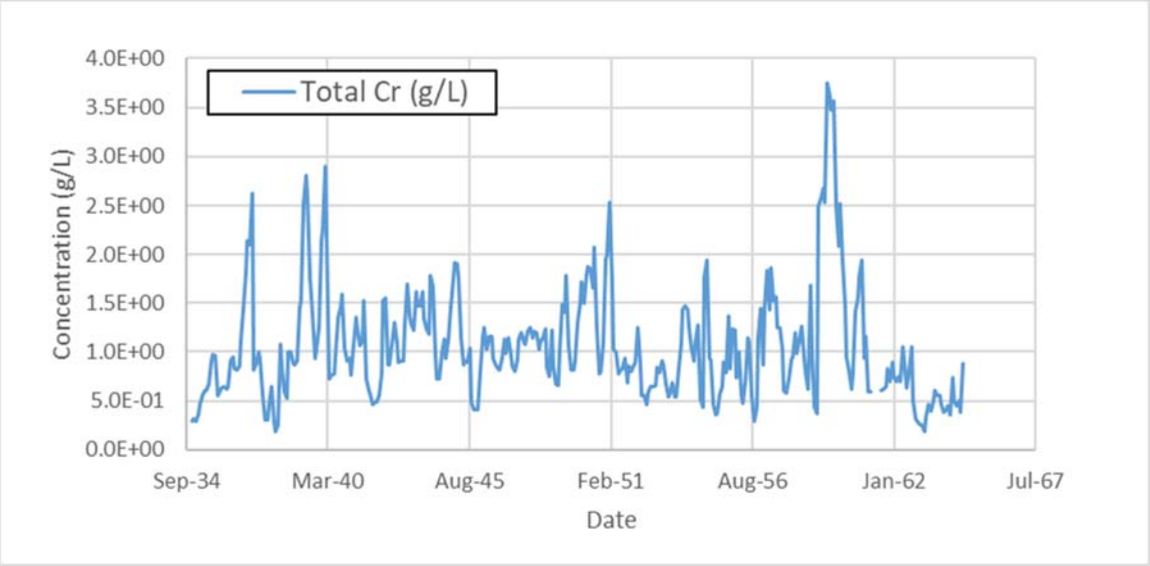

4.0E-02
3.5E-02
3.0E-02
2.5E-02
2.0E-02
1.5E-02
1.0E-02
5.0E-03
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Cd+2 (g/L)

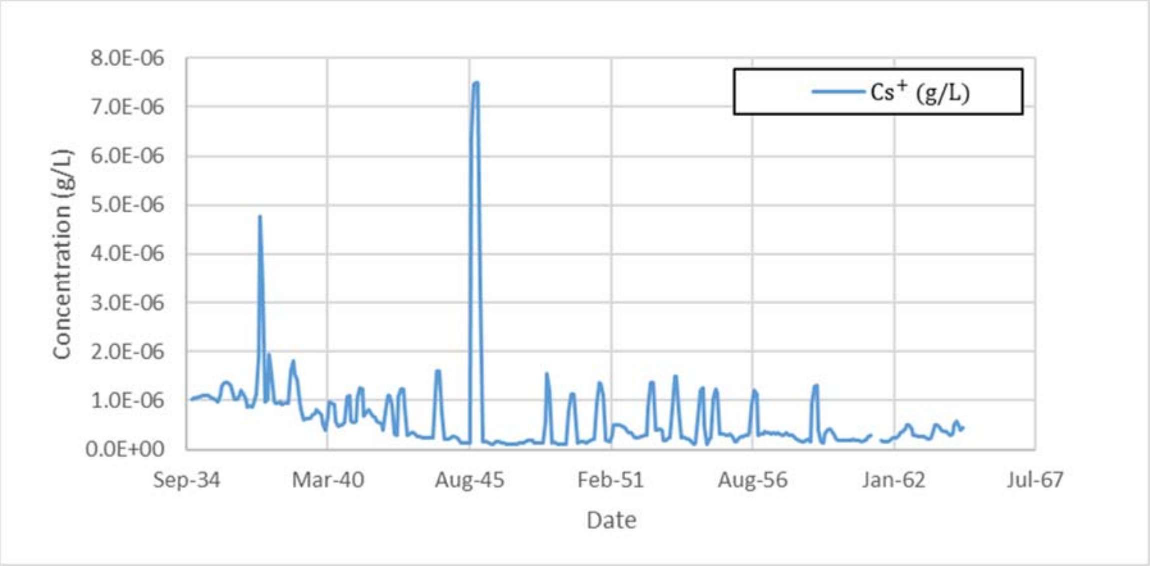

4.0E-01
3.5E-01
3.0E-01
2.5E-01
2.0E-01
1.5E-01
1.0E-01
5.0E-02
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Ce+3 (g/L)

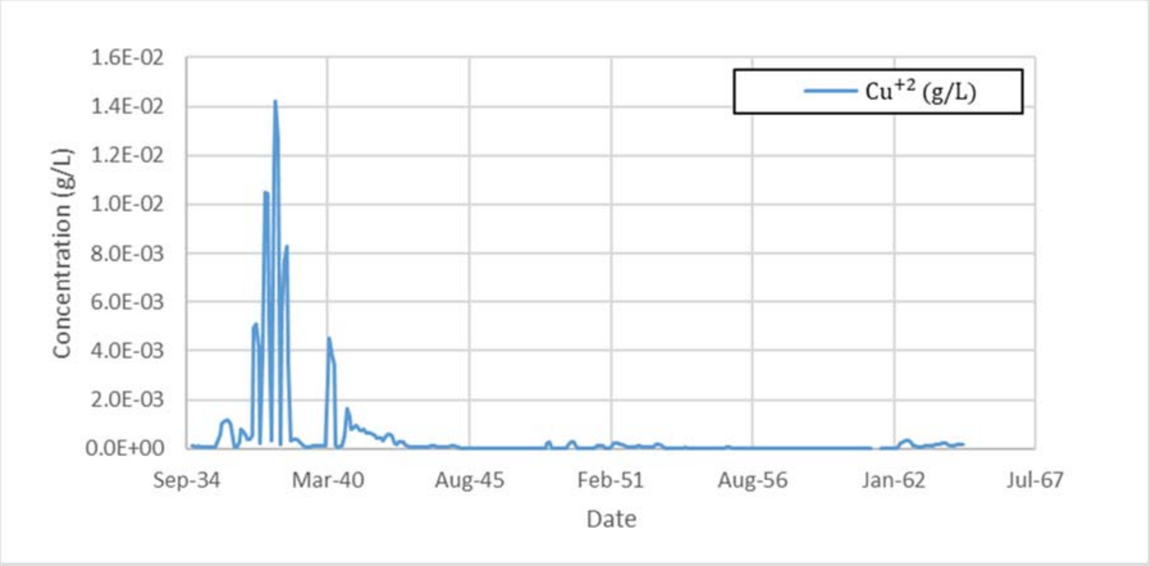

Cl⁻ (g/L)
Concentration (g/L)
4.0E+00
3.5E+00
3.0E+00
2.5E+00
2.0E+00
1.5E+00
1.0E+00
5.0E-01
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

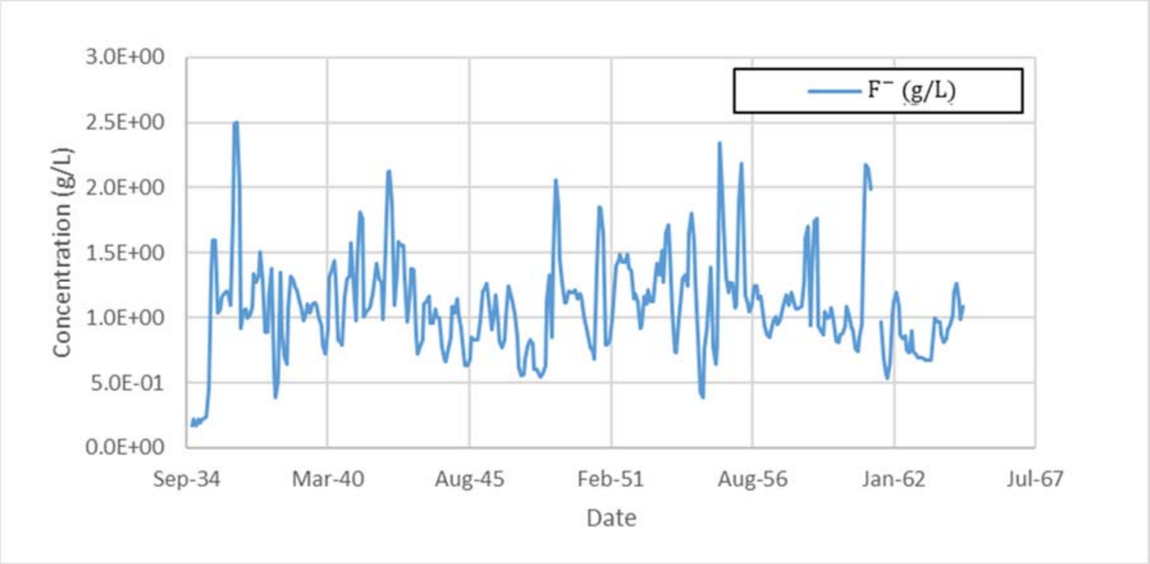

CN⁻ (g/L)
Concentration (g/L)
1.8E-02
1.6E-02
1.4E-02
1.2E-02
1.0E-02
8.0E-03
6.0E-03
4.0E-03
2.0E-03
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

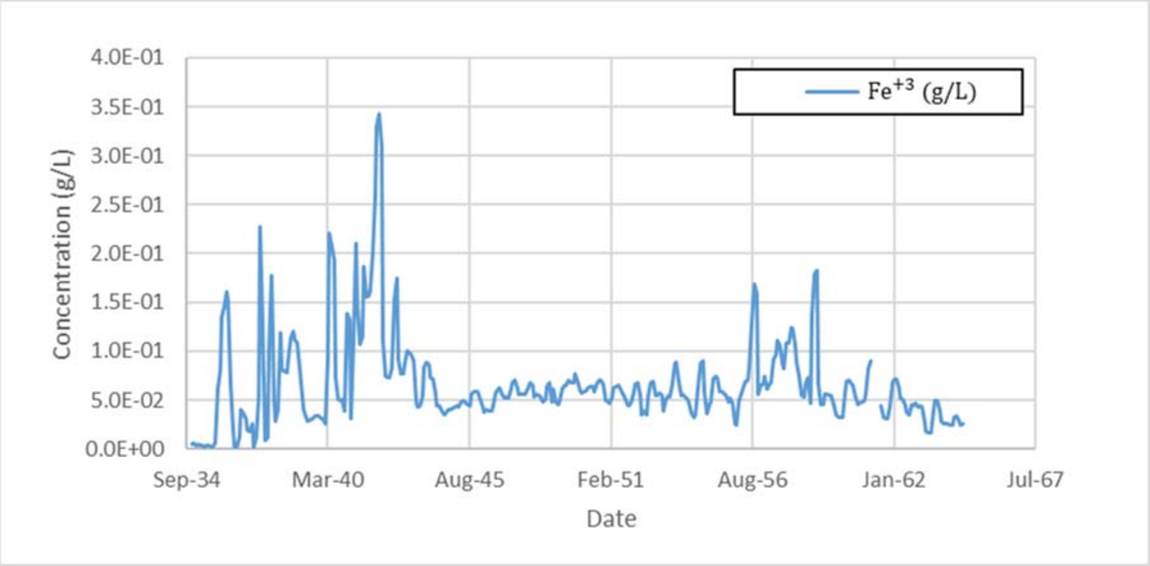

CO_3^{-2} (g/L)
Concentration (g/L)
4.0E+01
3.5E+01
3.0E+01
2.5E+01
2.0E+01
1.5E+01
1.0E+01
5.0E+00
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

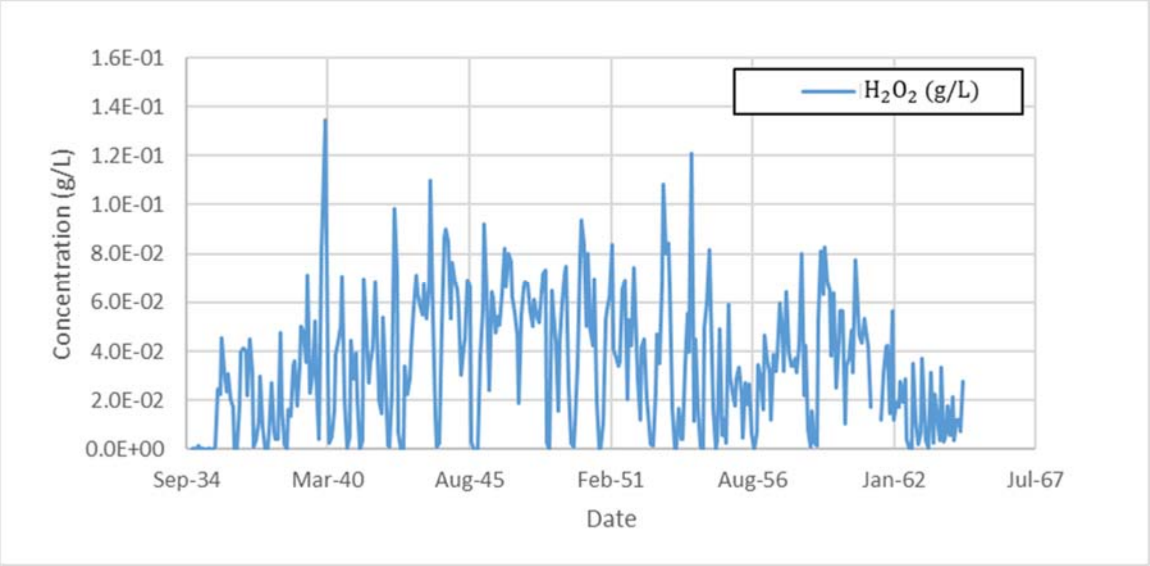

Total Cr (g/L)
Concentration (g/L)
4.0E+00
3.5E+00
3.0E+00
2.5E+00
2.0E+00
1.5E+00
1.0E+00
5.0E-01
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

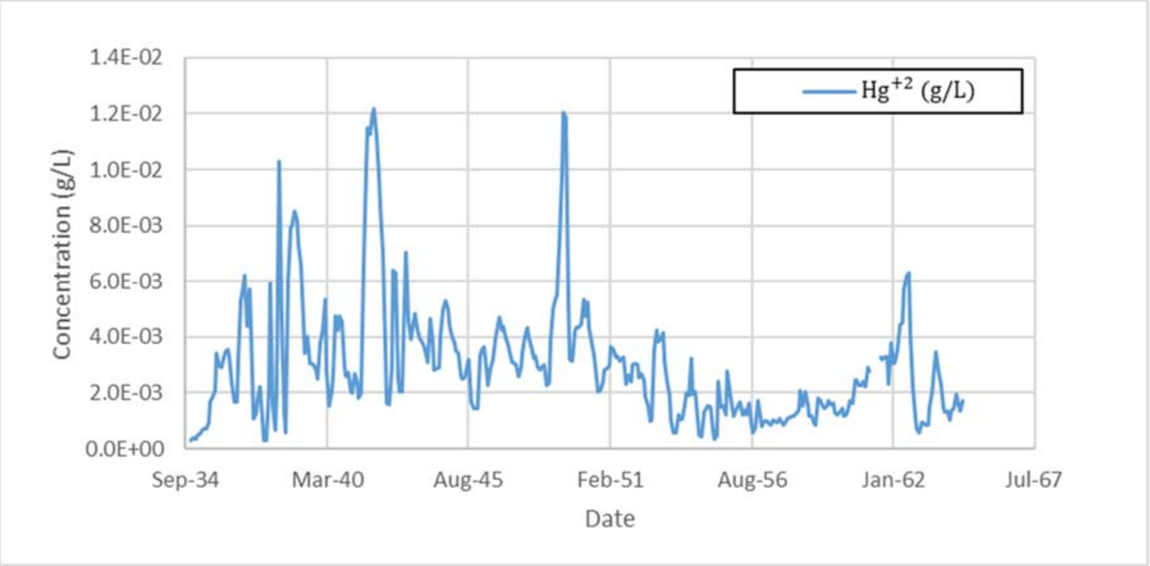

Cs$^+$ (g/L)
Concentration (g/L)
8.0E-06
7.0E-06
6.0E-06
5.0E-06
4.0E-06
3.0E-06
2.0E-06
1.0E-06
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

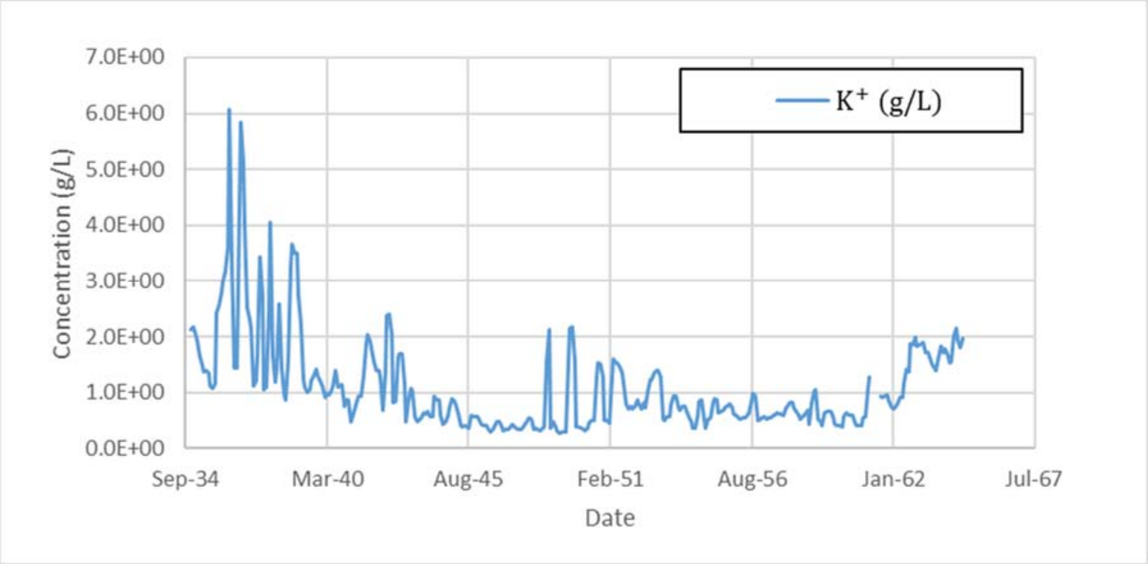

Cu^{+2} (g/L)
Concentration (g/L)
1.6E-02
1.4E-02
1.2E-02
1.0E-02
8.0E-03
6.0E-03
4.0E-03
2.0E-03
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

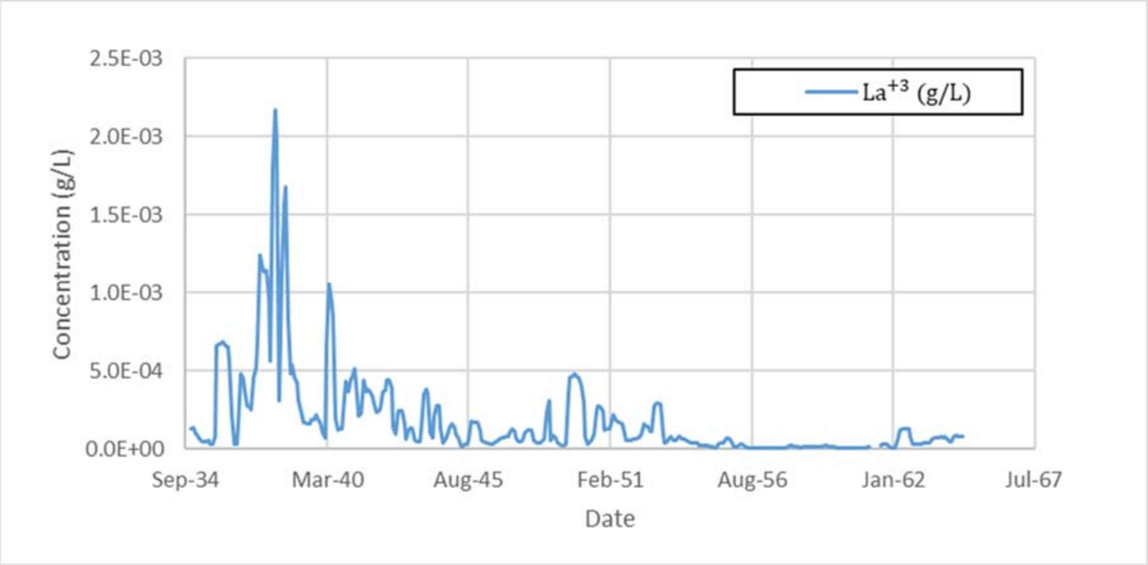

3.0E+00
2.5E+00
2.0E+00
1.5E+00
1.0E+00
5.0E-01
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
F⁻ (g/L)

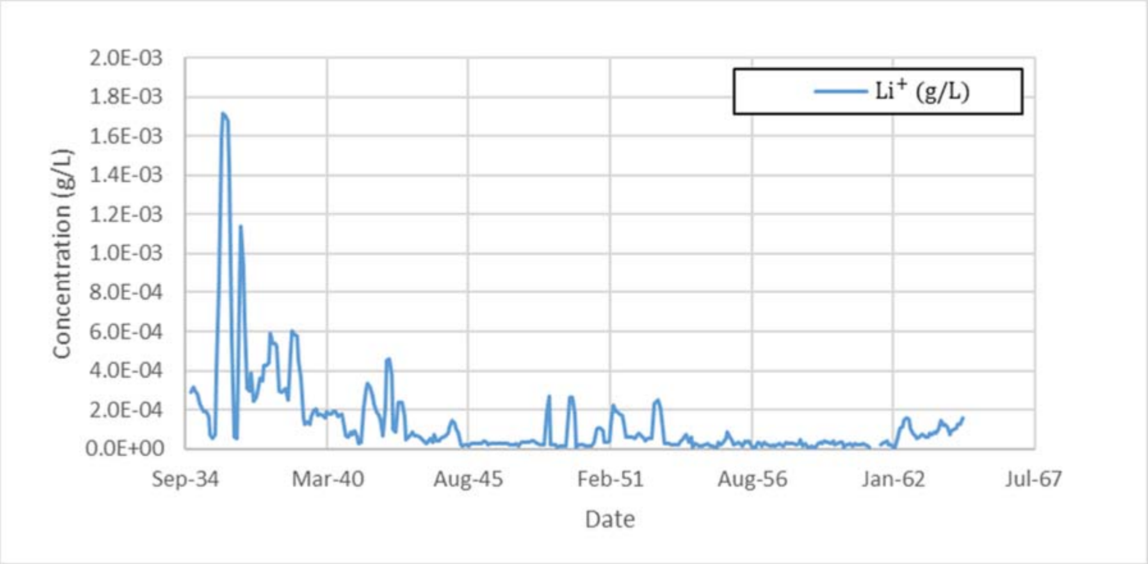

4.0E-01
3.5E-01
3.0E-01
2.5E-01
2.0E-01
1.5E-01
1.0E-01
5.0E-02
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Fe⁺³ (g/L)

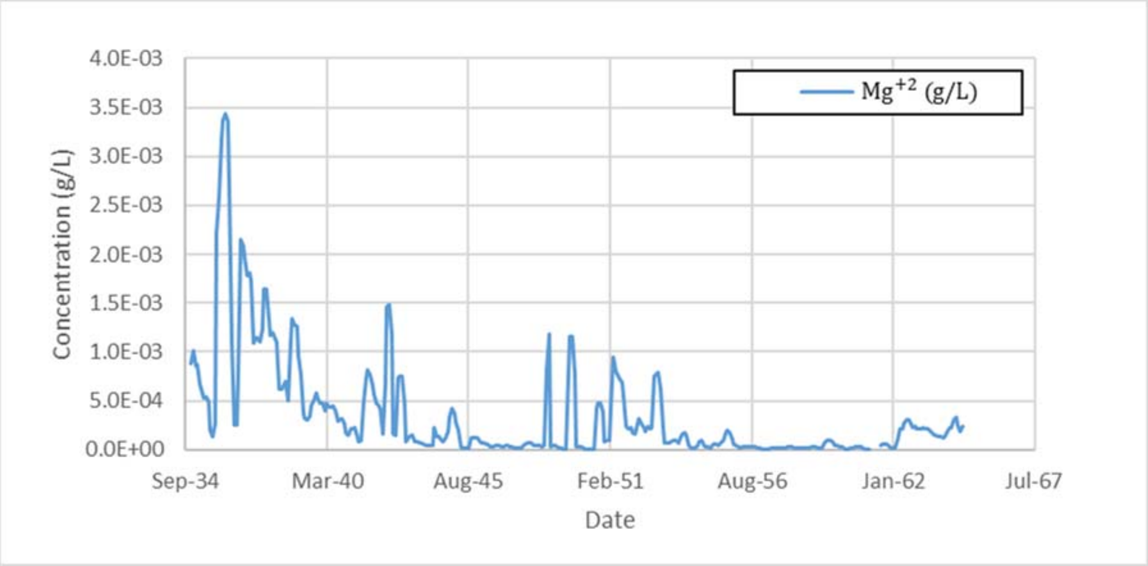

H_2O_2 (g/L)
Concentration (g/L)
1.6E-01
1.4E-01
1.2E-01
1.0E-01
8.0E-02
6.0E-02
4.0E-02
2.0E-02
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

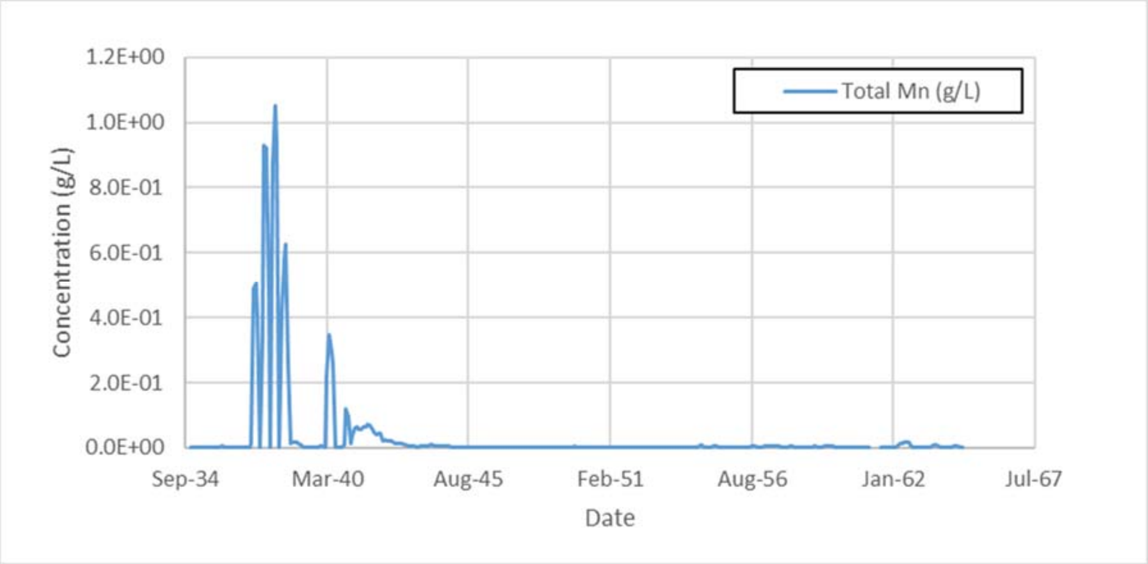

Hg^{+2} (g/L)
Concentration (g/L)
1.4E-02
1.2E-02
1.0E-02
8.0E-03
6.0E-03
4.0E-03
2.0E-03
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

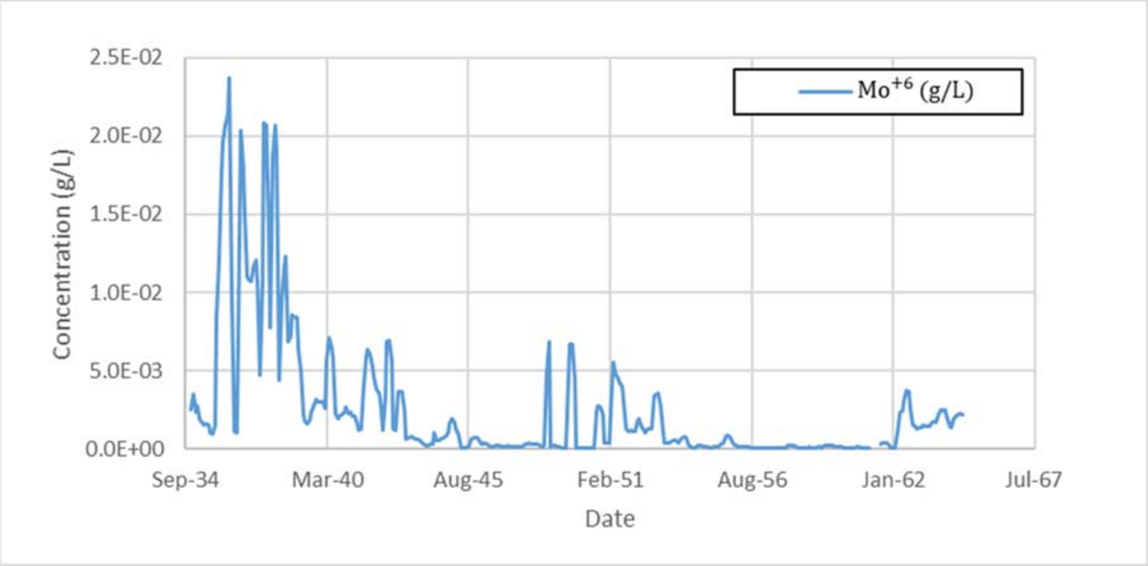

K⁺ (g/L)
Concentration (g/L)
7.0E+00
6.0E+00
5.0E+00
4.0E+00
3.0E+00
2.0E+00
1.0E+00
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

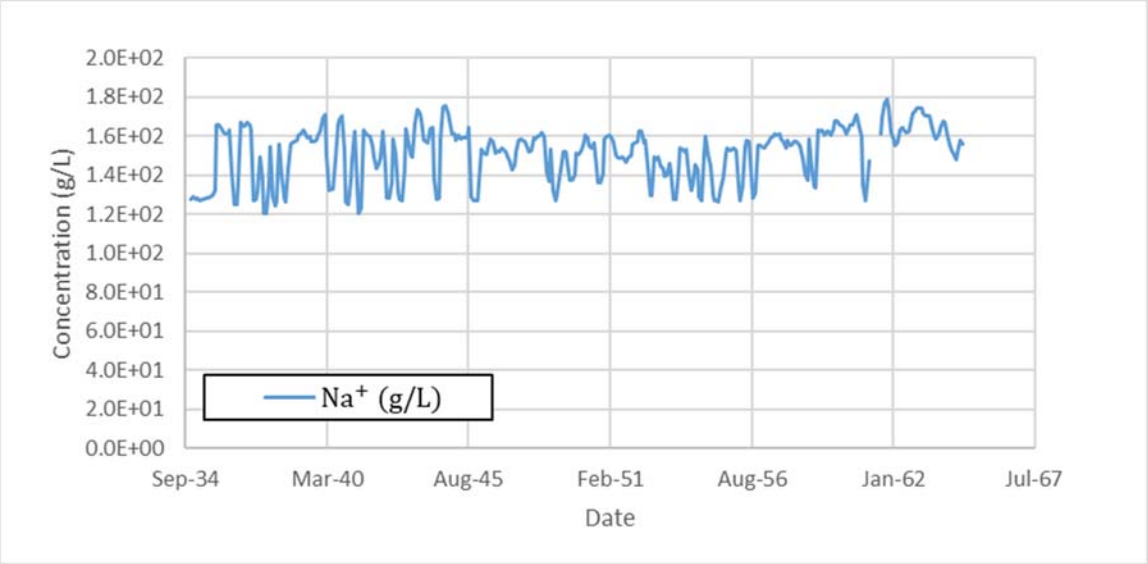

La⁺³ (g/L)
Concentration (g/L)
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

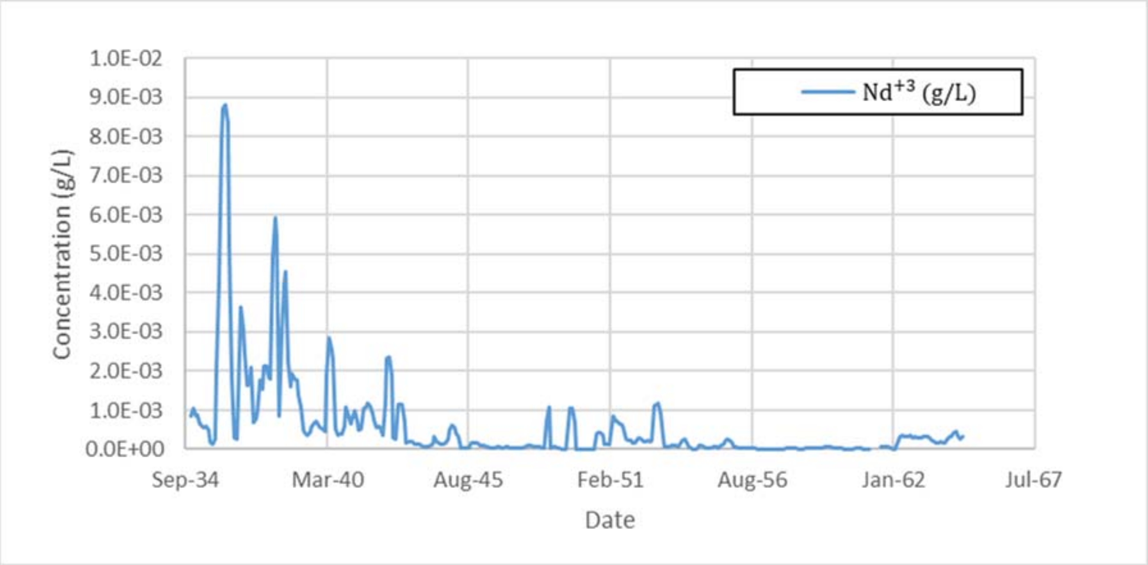

2.0E-03
1.8E-03
1.6E-03
1.4E-03
1.2E-03
1.0E-03
8.0E-04
6.0E-04
4.0E-04
2.0E-04
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Li$^+$ (g/L)

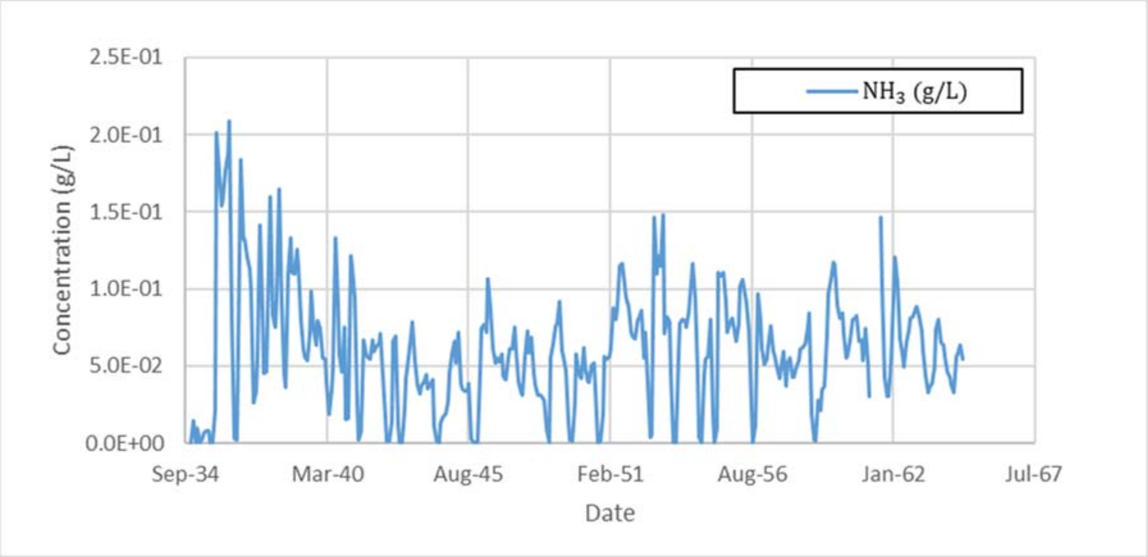

4.0E-03
3.5E-03
3.0E-03
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Mg^{+2} (g/L)

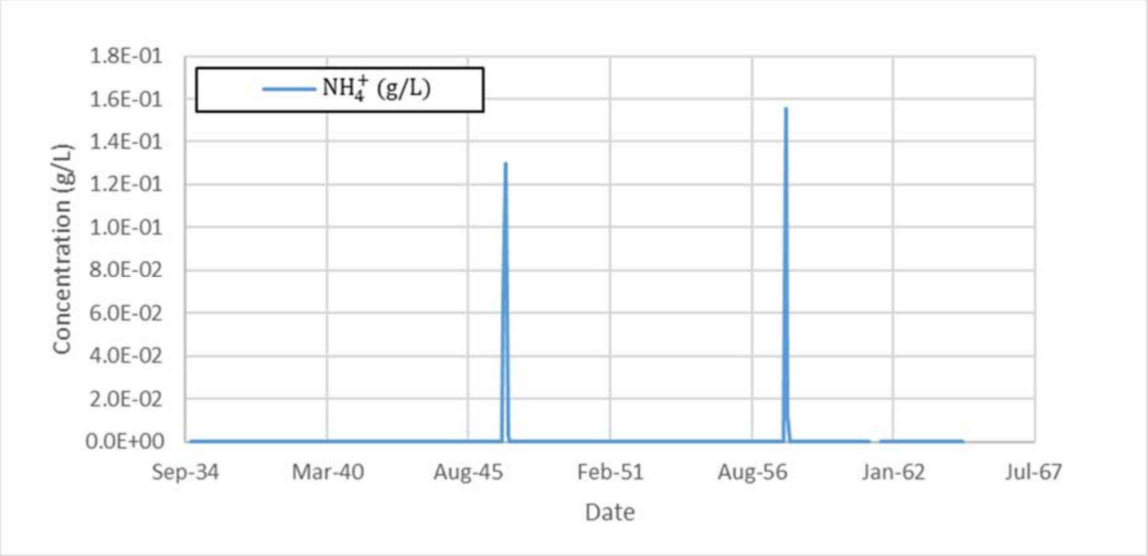

1.2E+00
1.0E+00
8.0E-01
6.0E-01
4.0E-01
2.0E-01
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Total Mn (g/L)

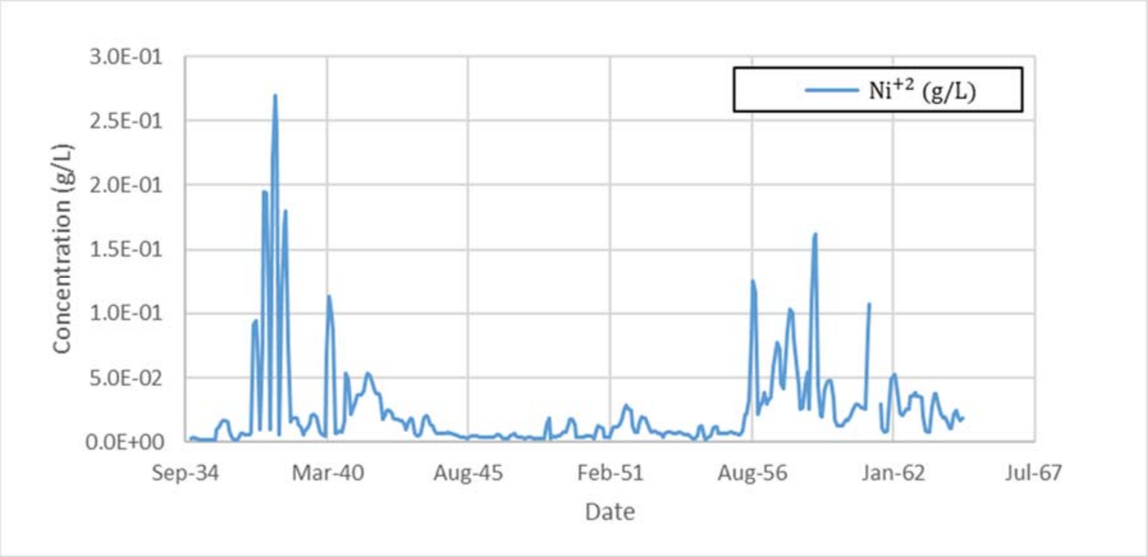

2.5E-02
2.0E-02
1.5E-02
1.0E-02
5.0E-03
0.0E+00
Concentration (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Mo+6 (g/L)


Concentration (g/L)
2.0E+02
1.8E+02
1.6E+02
1.4E+02
1.2E+02
1.0E+02
8.0E+01
6.0E+01
4.0E+01
2.0E+01
0.0E+00
Na+ (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

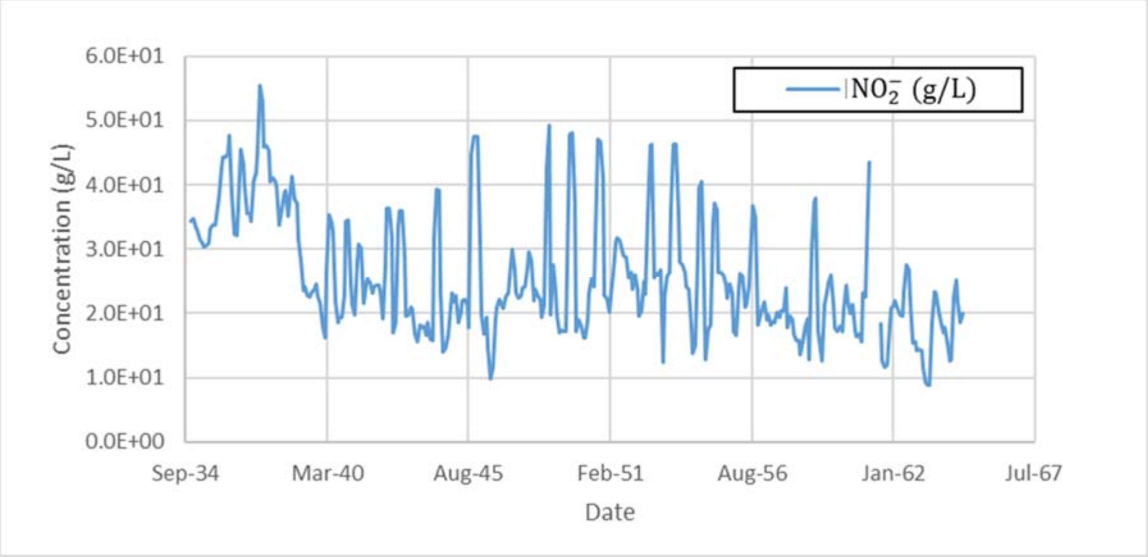

Concentration (g/L)
1.0E-02
9.0E-03
8.0E-03
7.0E-03
6.0E-03
5.0E-03
4.0E-03
3.0E-03
2.0E-03
1.0E-03
0.0E+00
Nd+3 (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date


NH$_3$ (g/L)
Concentration (g/L)
2.5E-01
2.0E-01
1.5E-01
1.0E-01
5.0E-02
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

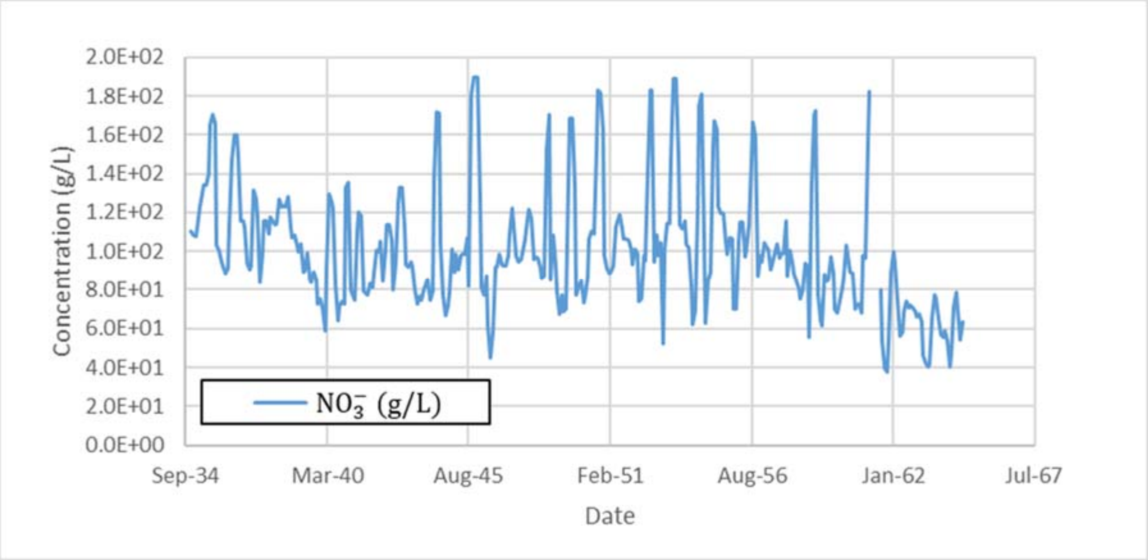

NH$_4^+$ (g/L)
Concentration (g/L)
1.8E-01
1.6E-01
1.4E-01
1.2E-01
1.0E-01
8.0E-02
6.0E-02
4.0E-02
2.0E-02
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

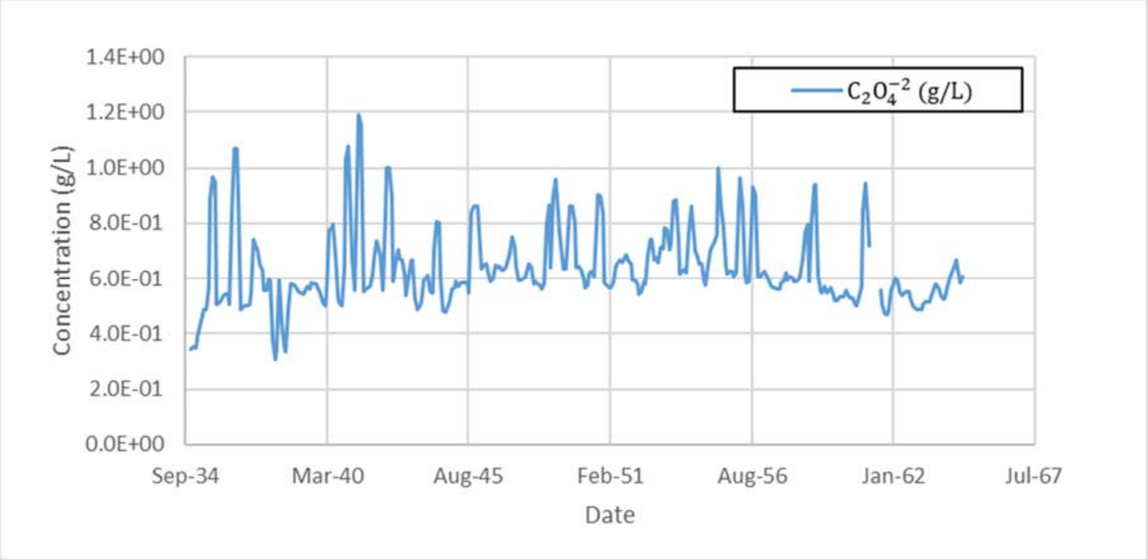


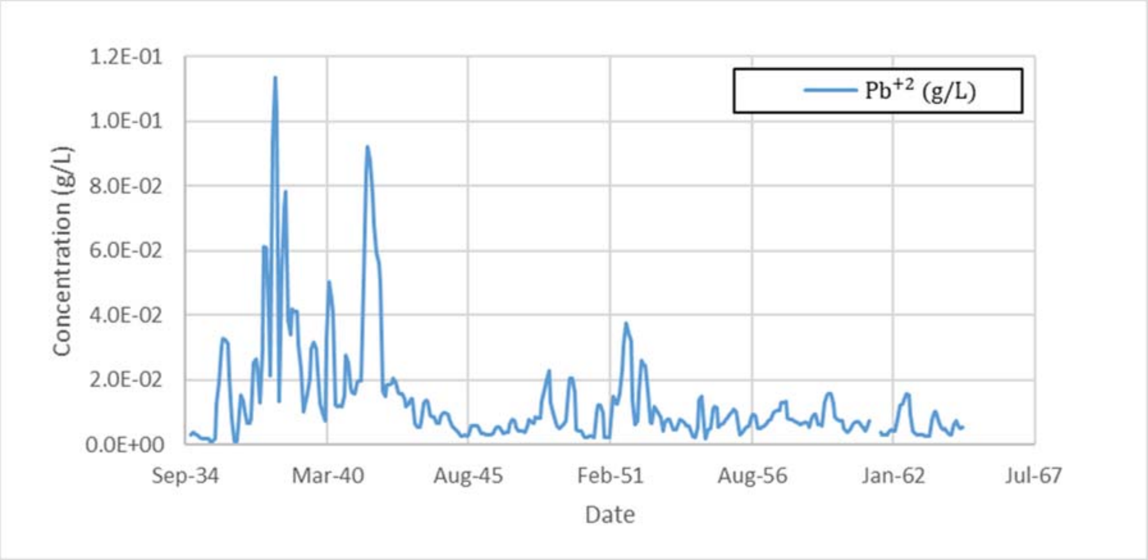
Ni^{+2} (g/L)
Concentration (g/L)
3.0E-01
2.5E-01
2.0E-01
1.5E-01
1.0E-01
5.0E-02
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

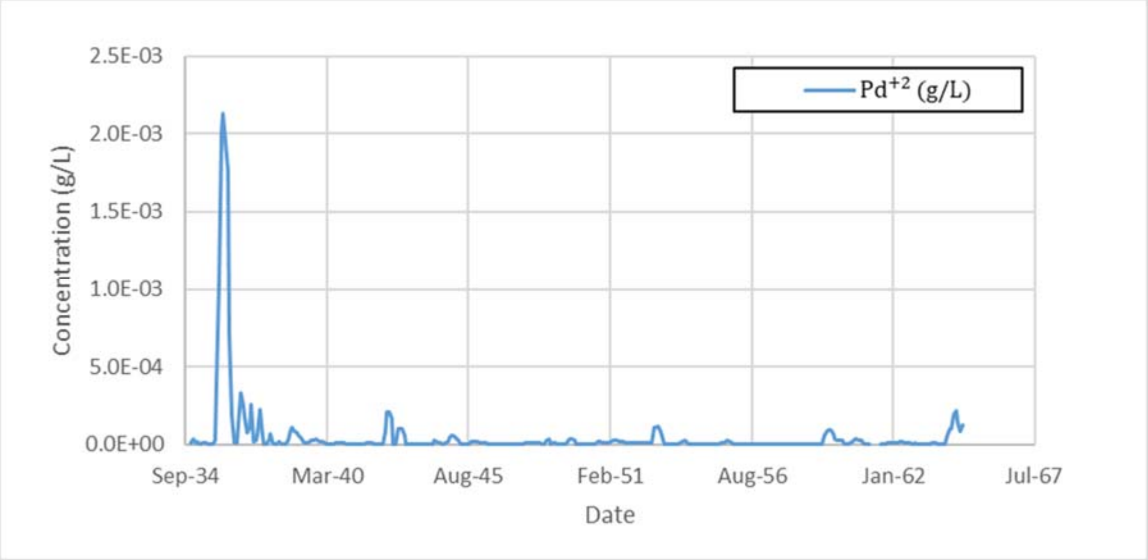


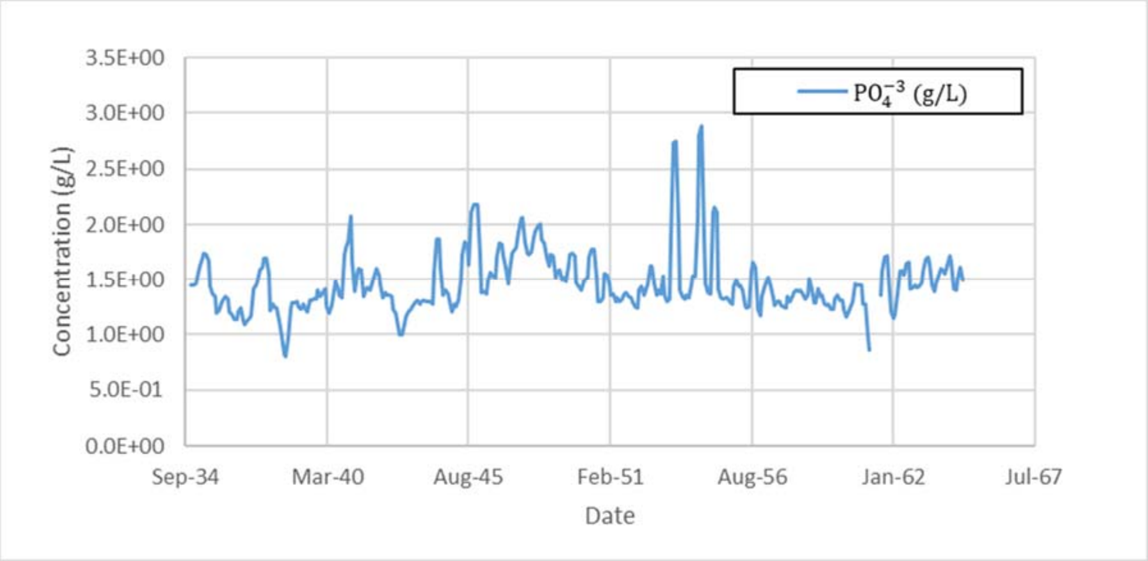
NO_2^- (g/L)
Concentration (g/L)
6.0E+01
5.0E+01
4.0E+01
3.0E+01
2.0E+01
1.0E+01
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

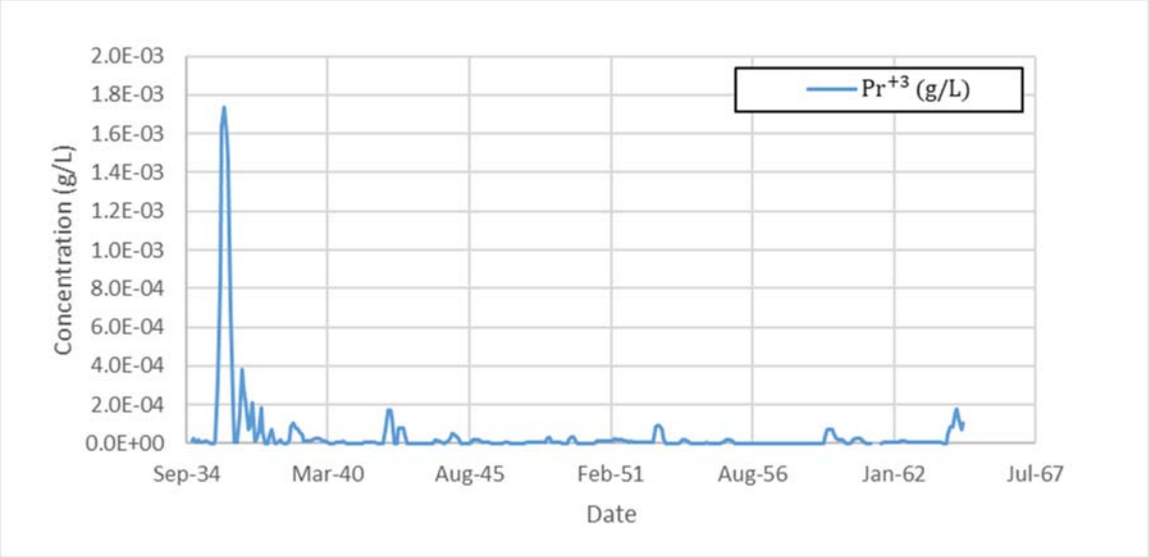

2.0E+02
1.8E+02
1.6E+02
1.4E+02
1.2E+02
1.0E+02
8.0E+01
6.0E+01
4.0E+01
2.0E+01
0.0E+00
Concentration (g/L)
NO_3^- (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

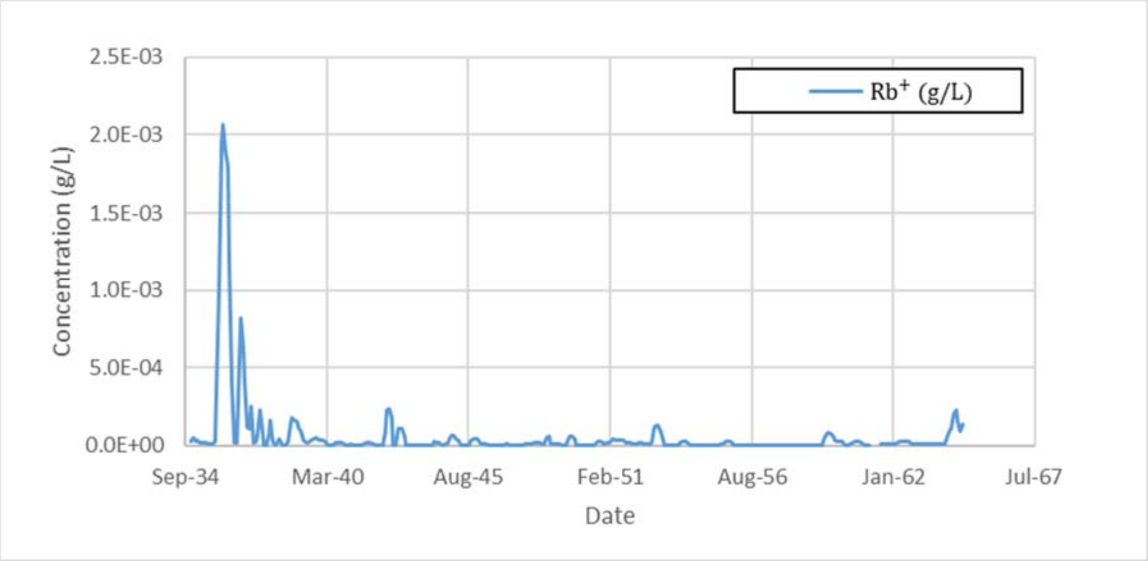

1.4E+00
1.2E+00
1.0E+00
8.0E-01
6.0E-01
4.0E-01
2.0E-01
0.0E+00
Concentration (g/L)
$C_2O_4^{-2}$ (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

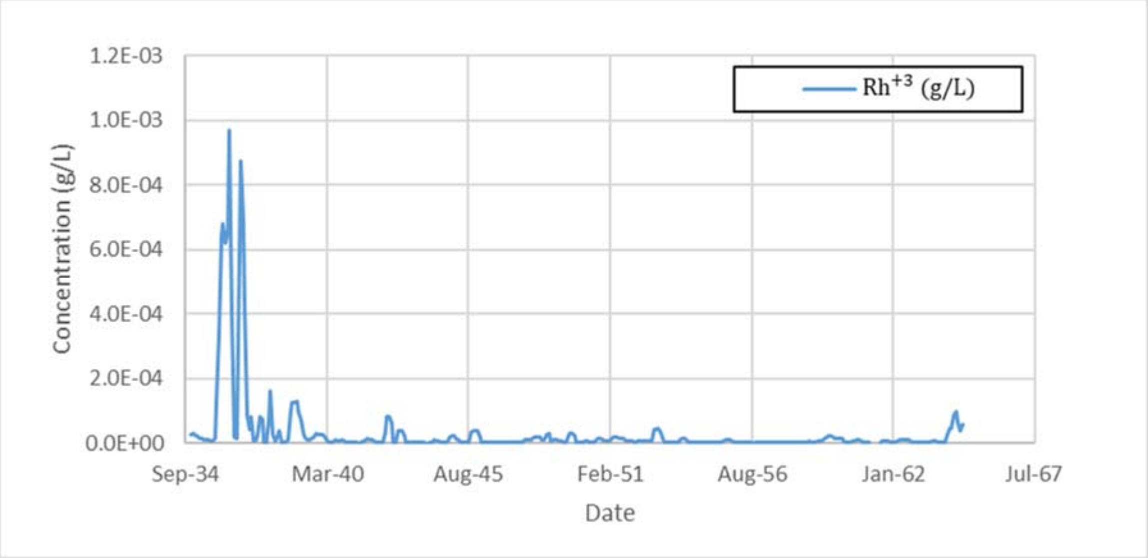

Pb^{+2} (g/L)
Concentration (g/L)
1.2E-01
1.0E-01
8.0E-02
6.0E-02
4.0E-02
2.0E-02
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

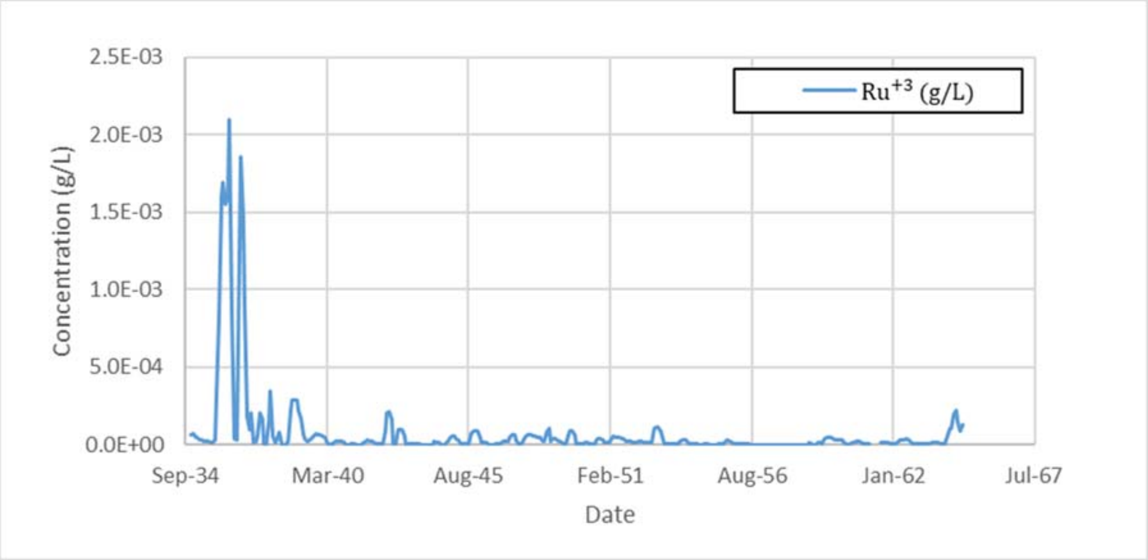

Pd^{+2} (g/L)
Concentration (g/L)
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

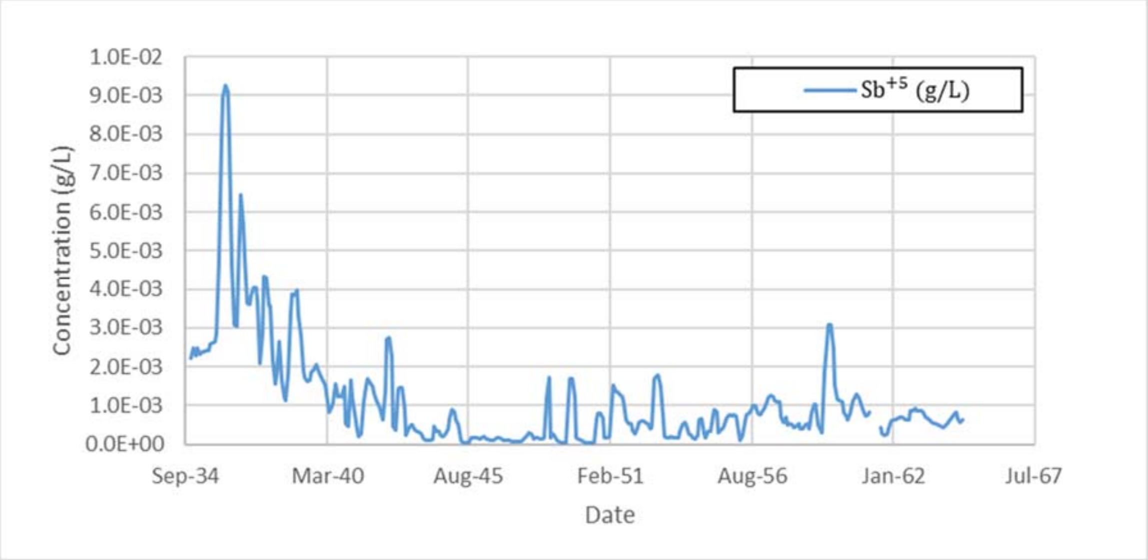

PO_4^{-3} (g/L)
Concentration (g/L)
3.5E+00
3.0E+00
2.5E+00
2.0E+00
1.5E+00
1.0E+00
5.0E-01
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

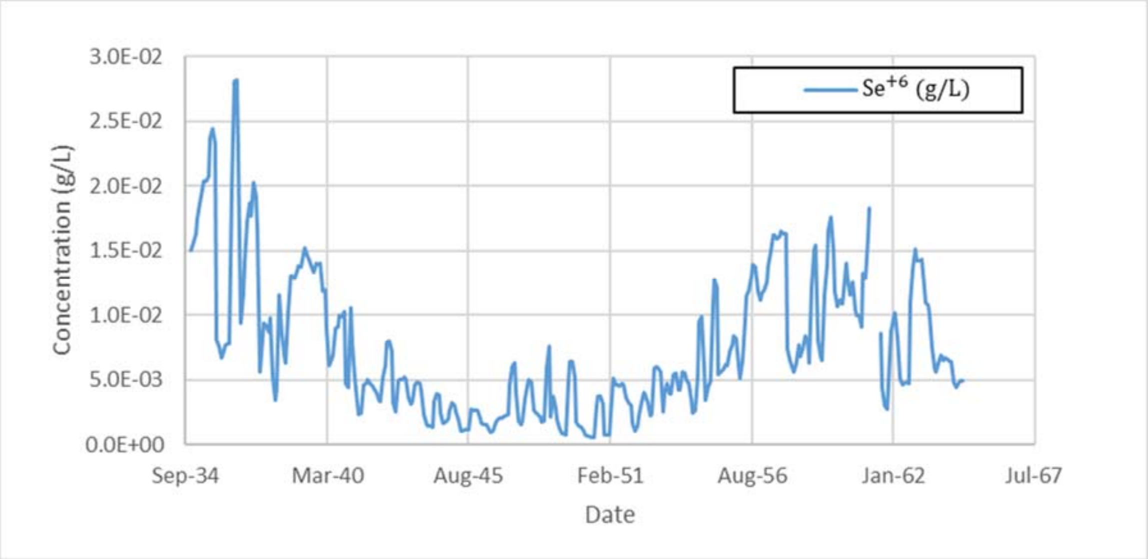

Pr^{+3} (g/L)
Concentration (g/L)
2.0E-03
1.8E-03
1.6E-03
1.4E-03
1.2E-03
1.0E-03
8.0E-04
6.0E-04
4.0E-04
2.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

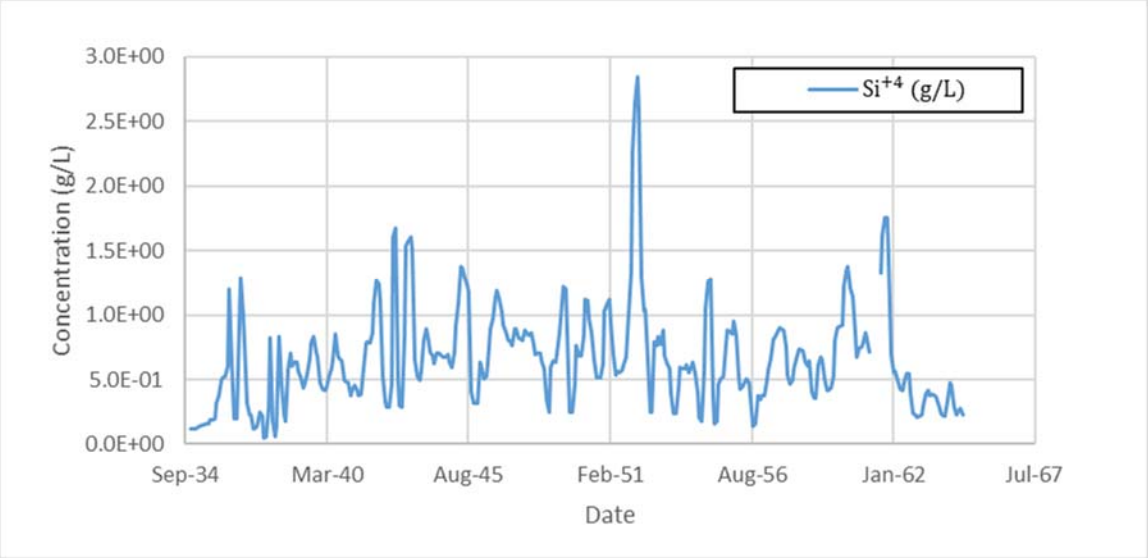

2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Concentration (g/L)
Rb⁺ (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

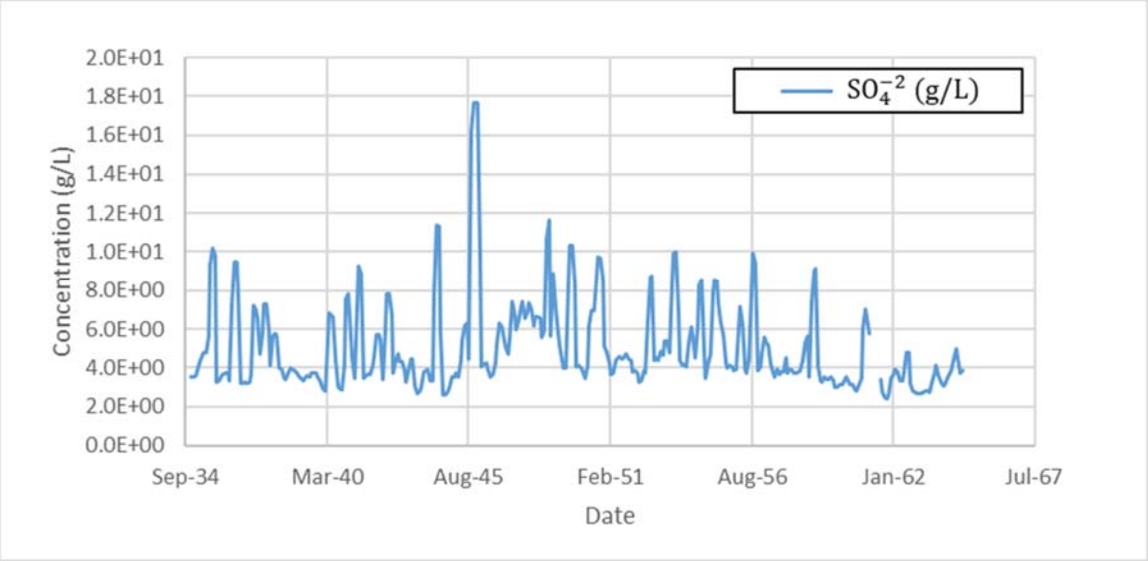

1.2E-03
1.0E-03
8.0E-04
6.0E-04
4.0E-04
2.0E-04
0.0E+00
Concentration (g/L)
Rh⁺³ (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date


Concentration (g/L)
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Ru+3 (g/L)

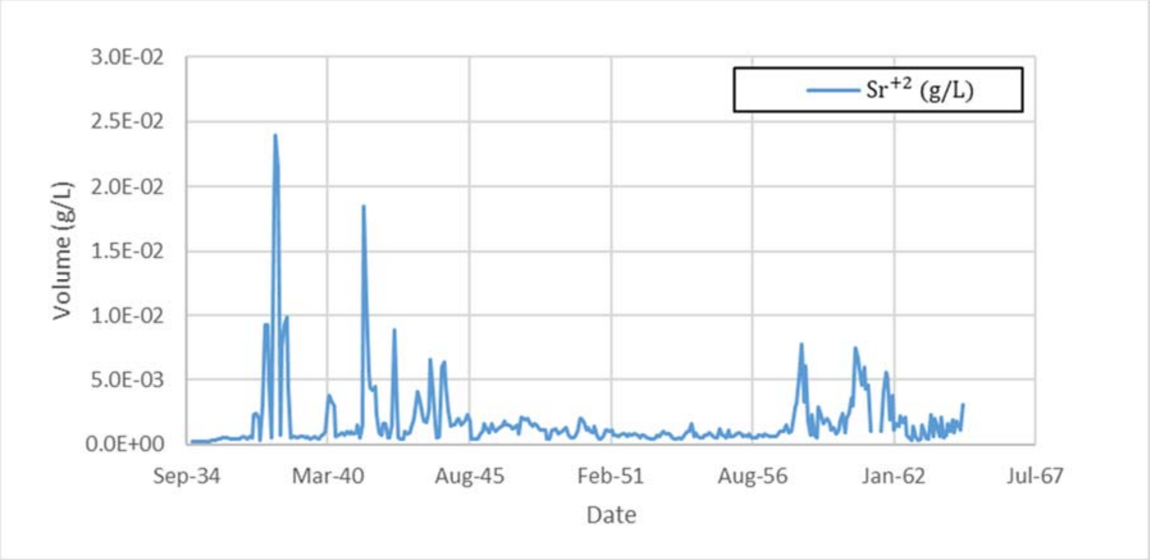

Concentration (g/L)
1.0E-02
9.0E-03
8.0E-03
7.0E-03
6.0E-03
5.0E-03
4.0E-03
3.0E-03
2.0E-03
1.0E-03
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Sb+5 (g/L)


3.0E-02
2.5E-02
2.0E-02
1.5E-02
1.0E-02
5.0E-03
0.0E+00
Concentration (g/L)
Se^{+6} (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

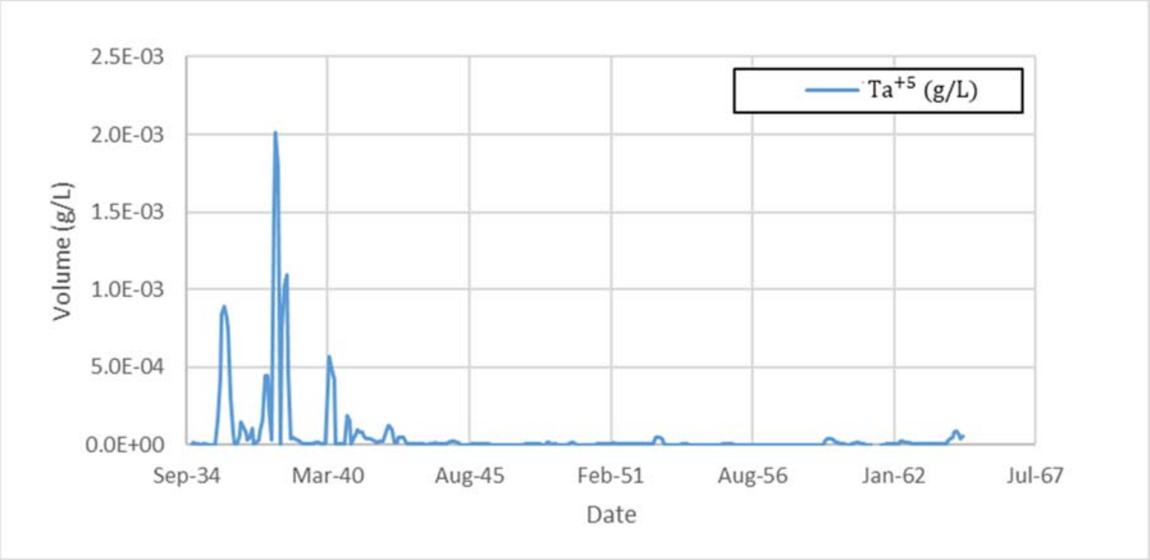

3.0E+00
2.5E+00
2.0E+00
1.5E+00
1.0E+00
5.0E-01
0.0E+00
Concentration (g/L)
Si^{+4} (g/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

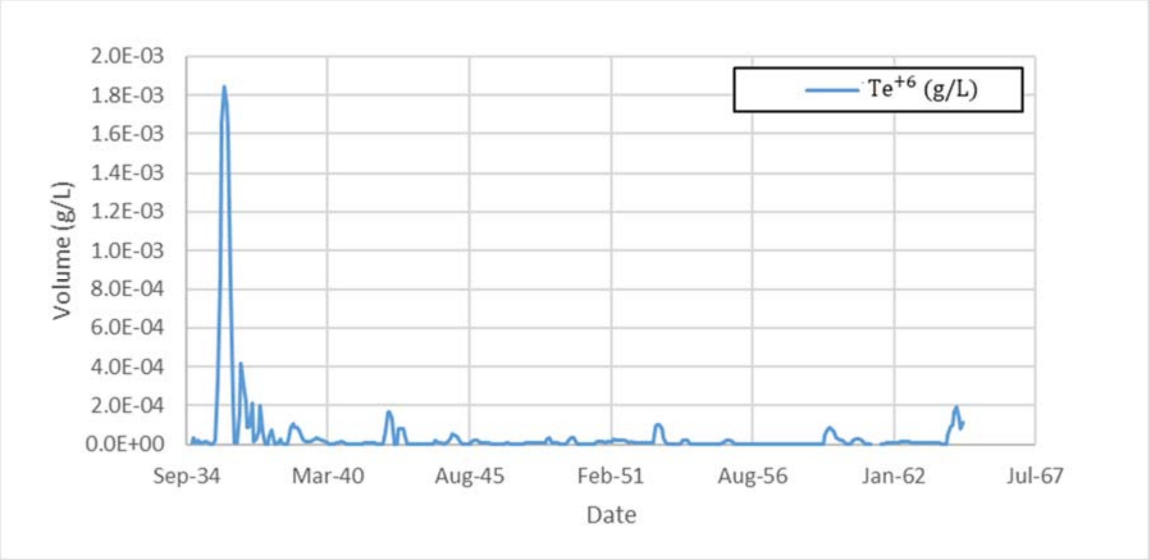


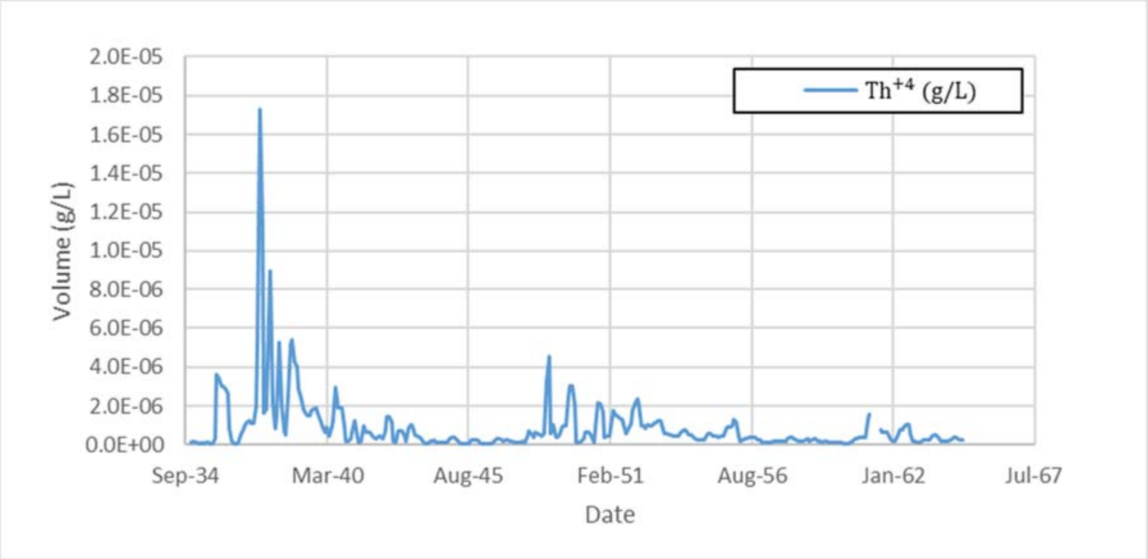
SO_4^{-2} (g/L)
Concentration (g/L)
2.0E+01
1.8E+01
1.6E+01
1.4E+01
1.2E+01
1.0E+01
8.0E+00
6.0E+00
4.0E+00
2.0E+00
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

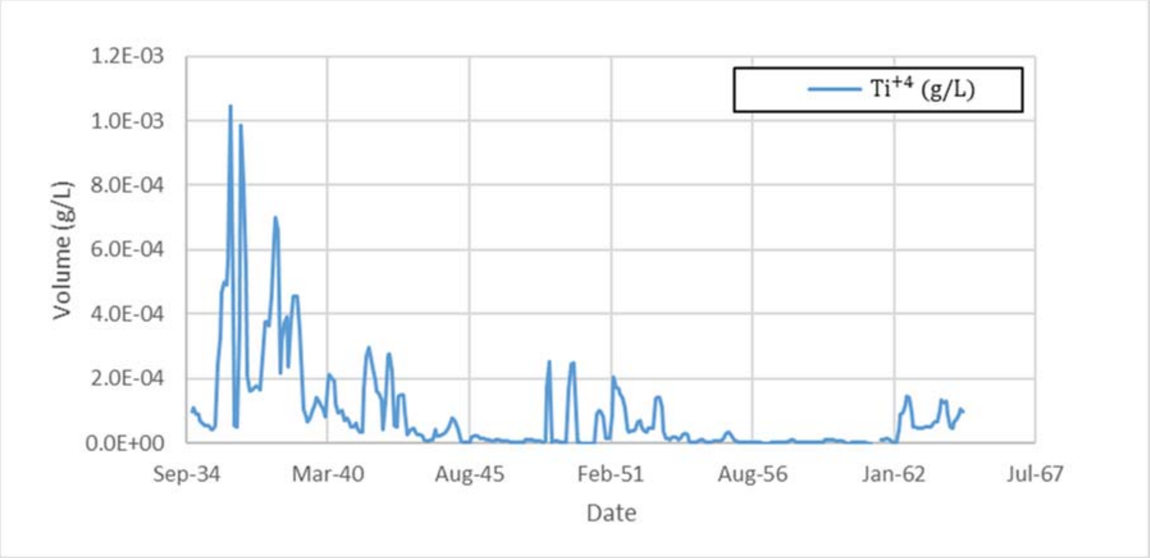


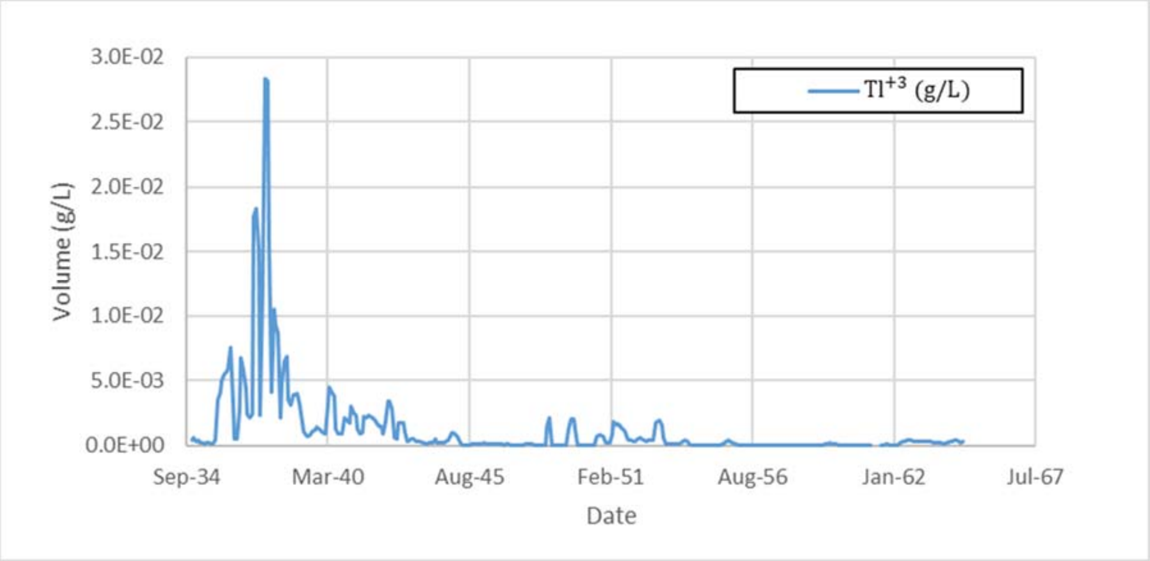
Sr^{+2} (g/L)
Volume (g/L)
3.0E-02
2.5E-02
2.0E-02
1.5E-02
1.0E-02
5.0E-03
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

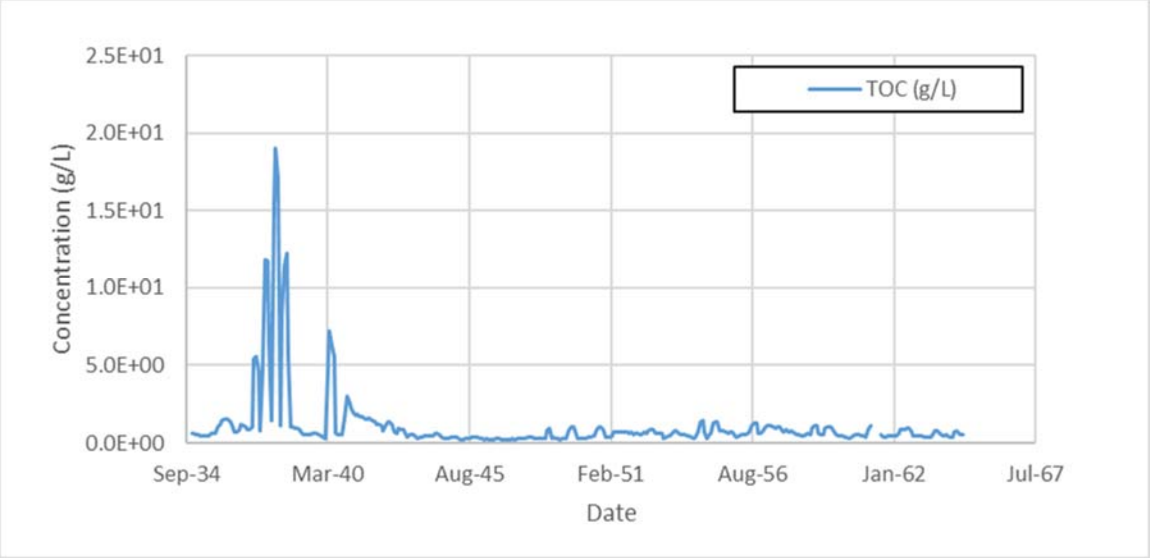

Ta^{+5} (g/L)
Volume (g/L)
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

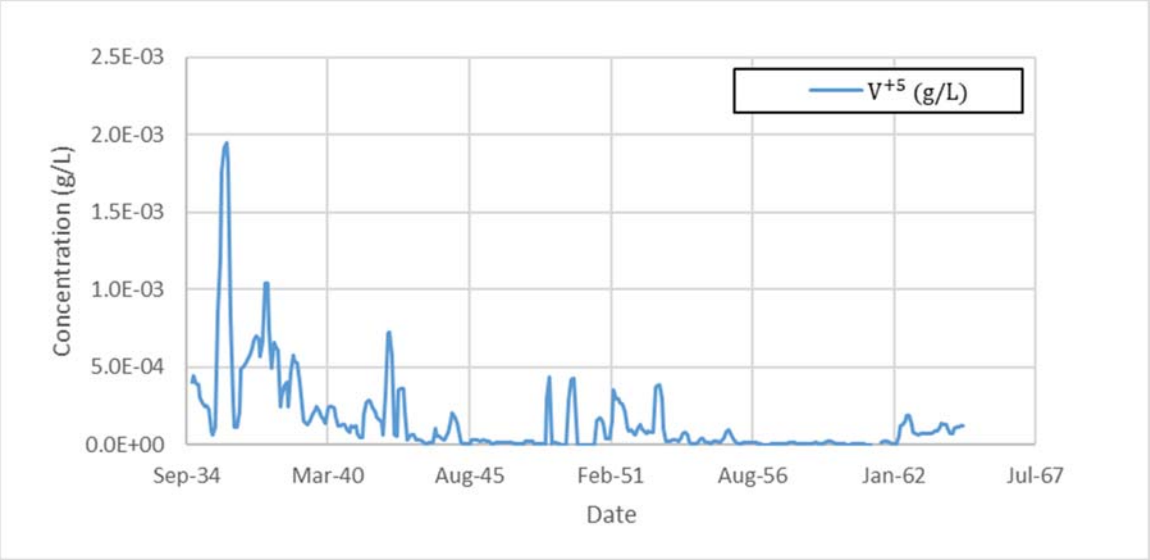

Te^{+6} (g/L)
Volume (g/L)
2.0E-03
1.8E-03
1.6E-03
1.4E-03
1.2E-03
1.0E-03
8.0E-04
6.0E-04
4.0E-04
2.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

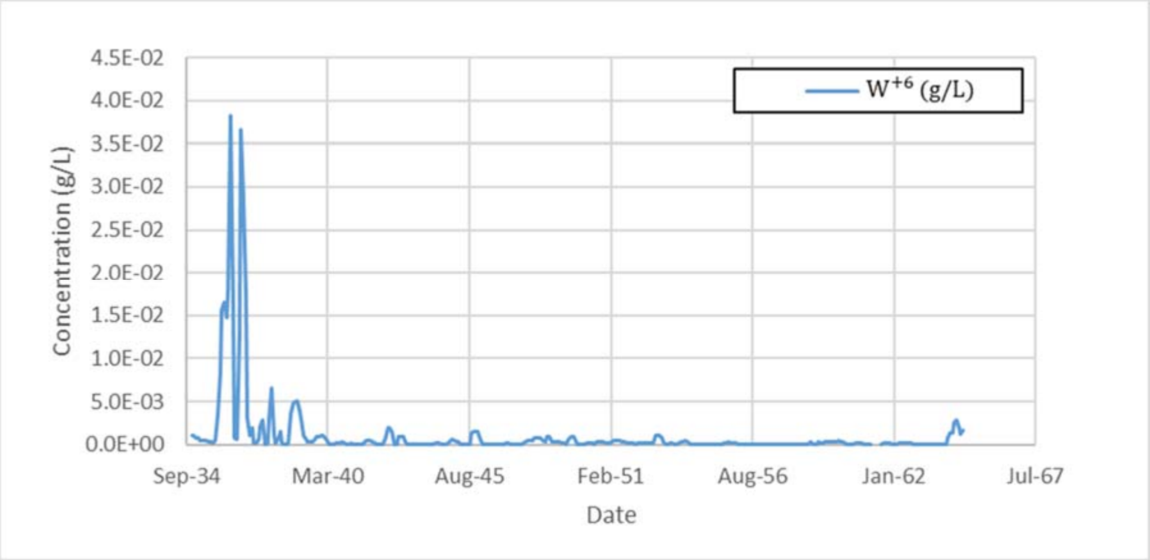

Th^{+4} (g/L)
Volume (g/L)
2.0E-05
1.8E-05
1.6E-05
1.4E-05
1.2E-05
1.0E-05
8.0E-06
6.0E-06
4.0E-06
2.0E-06
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

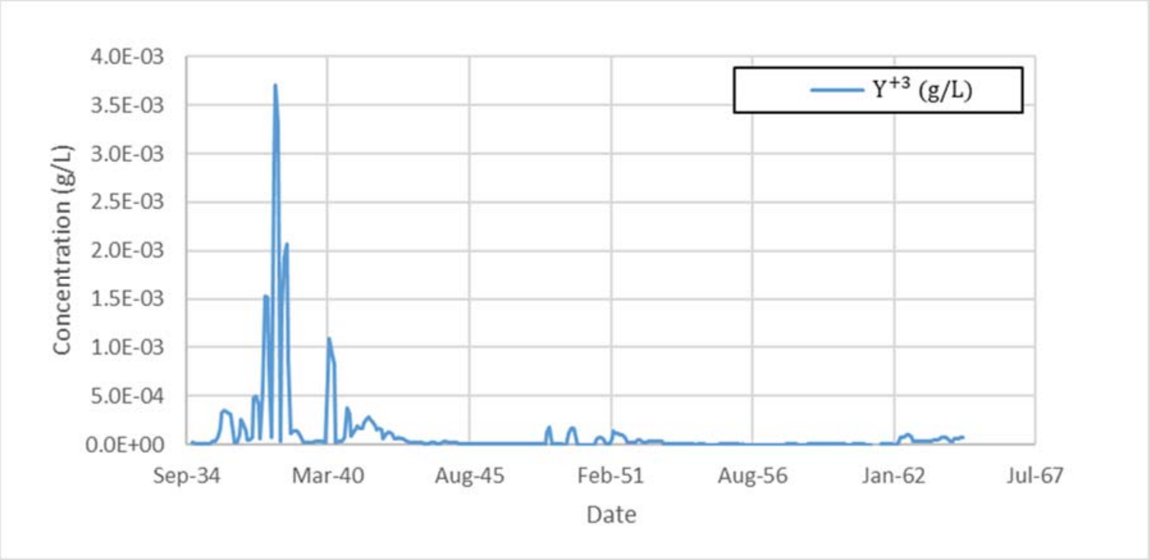

Ti^{+4} (g/L)
Volume (g/L)
1.2E-03
1.0E-03
8.0E-04
6.0E-04
4.0E-04
2.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

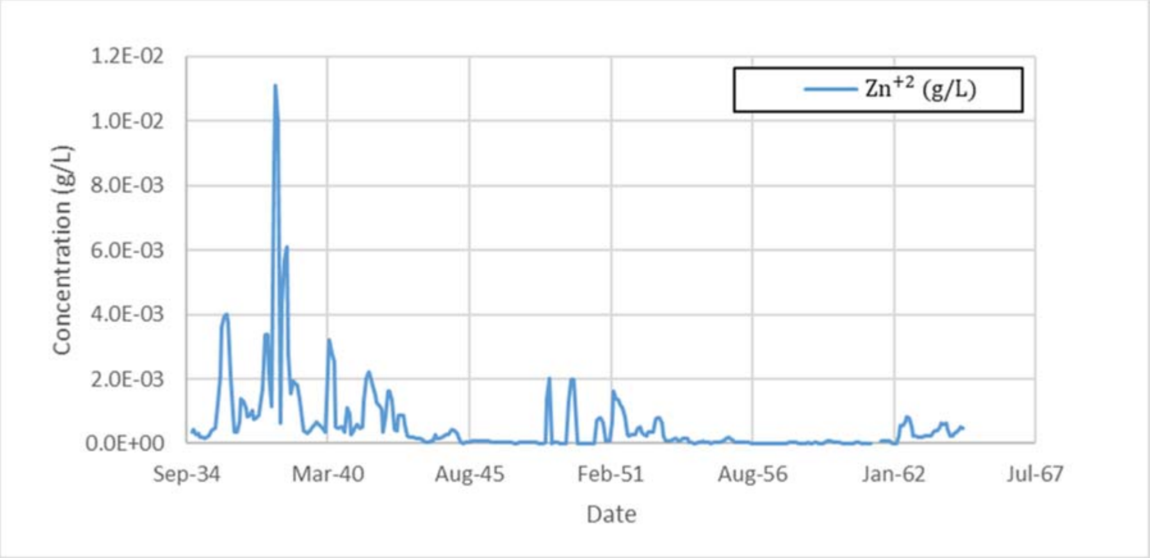

Volume (g/L)
3.0E-02
2.5E-02
2.0E-02
1.5E-02
1.0E-02
5.0E-03
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
Tl+3 (g/L)

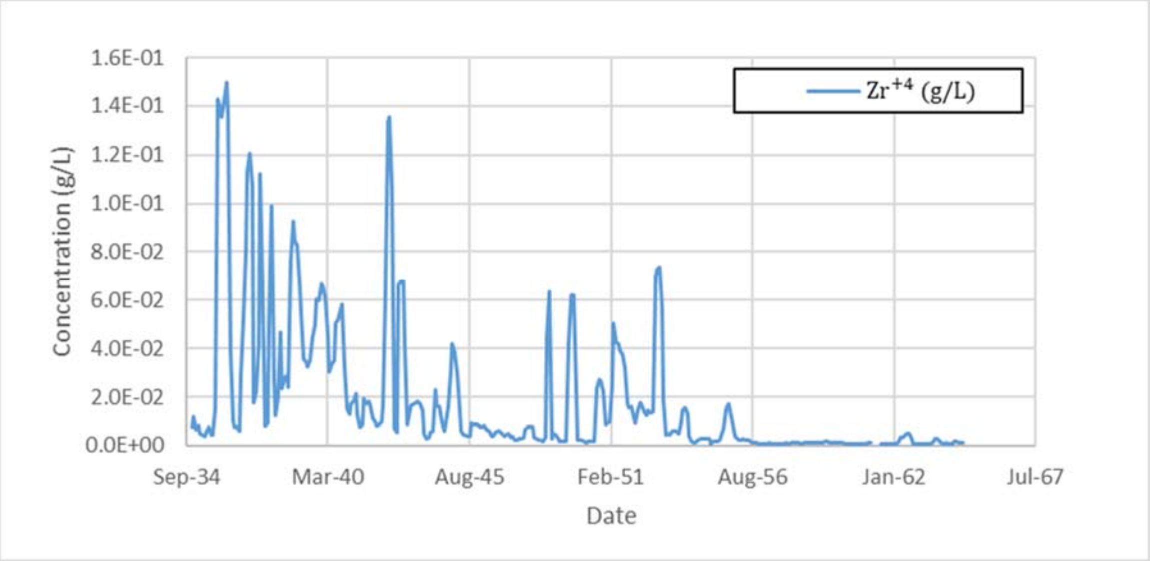

Concentration (g/L)
2.5E+01
2.0E+01
1.5E+01
1.0E+01
5.0E+00
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date
TOC (g/L)


V+5 (g/L)
Concentration (g/L)
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

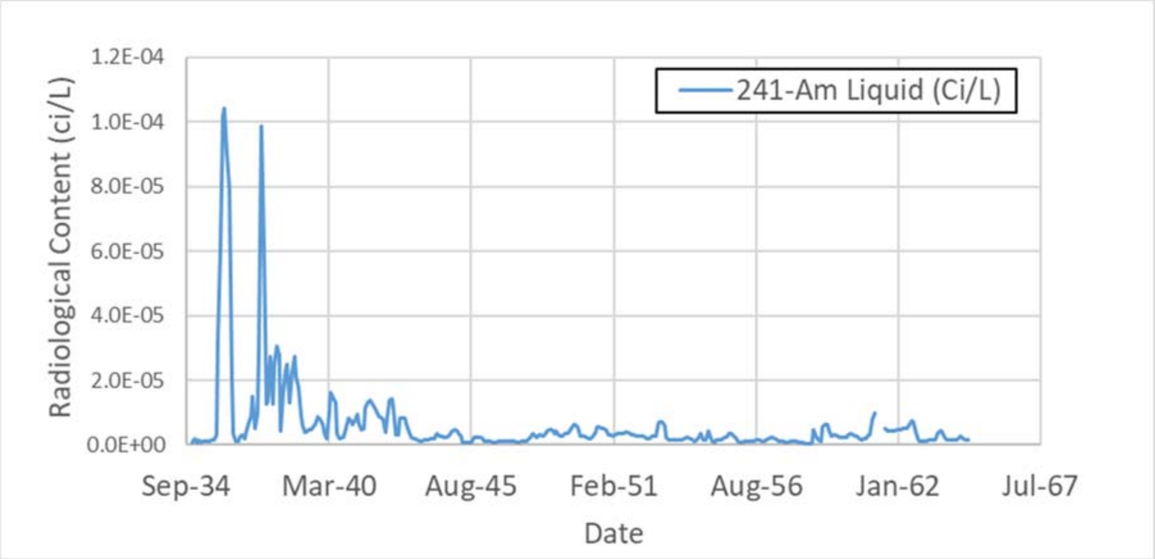

W+6 (g/L)
Concentration (g/L)
4.5E-02
4.0E-02
3.5E-02
3.0E-02
2.5E-02
2.0E-02
1.5E-02
1.0E-02
5.0E-03
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

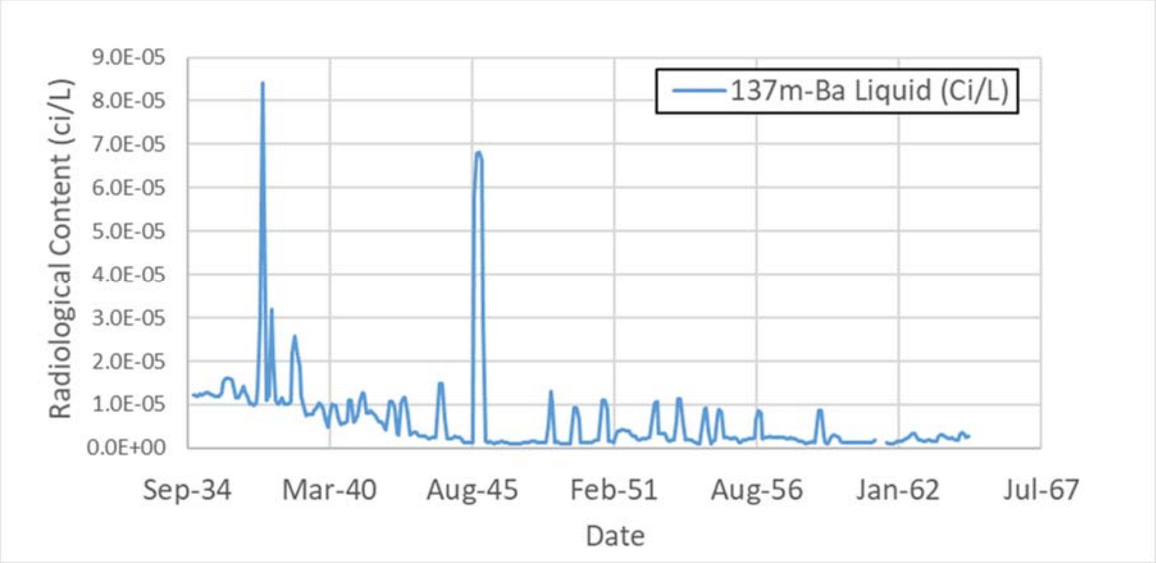

Y+3 (g/L)
Concentration (g/L)
4.0E-03
3.5E-03
3.0E-03
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

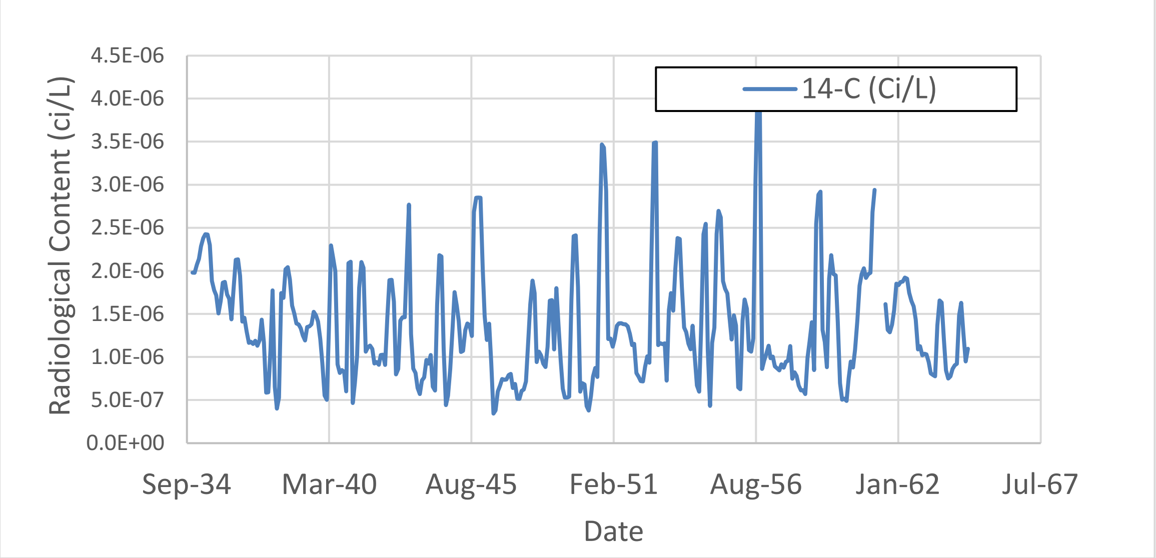

Zn+2 (g/L)
Concentration (g/L)
1.2E-02
1.0E-02
8.0E-03
6.0E-03
4.0E-03
2.0E-03
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

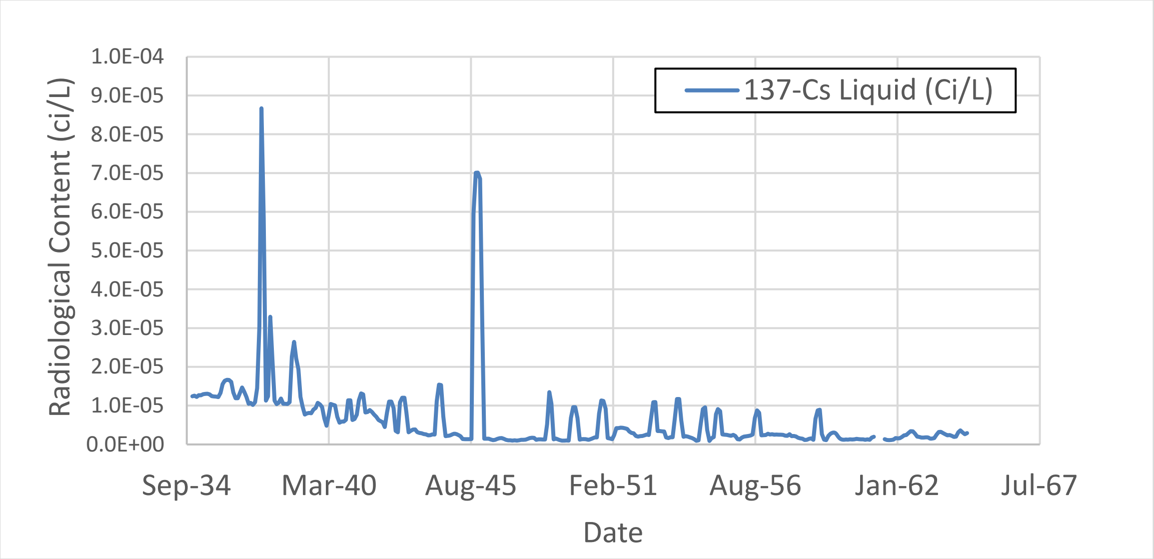

Zr^{+4} (g/L)
Concentration (g/L)
1.6E-01
1.4E-01
1.2E-01
1.0E-01
8.0E-02
6.0E-02
4.0E-02
2.0E-02
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

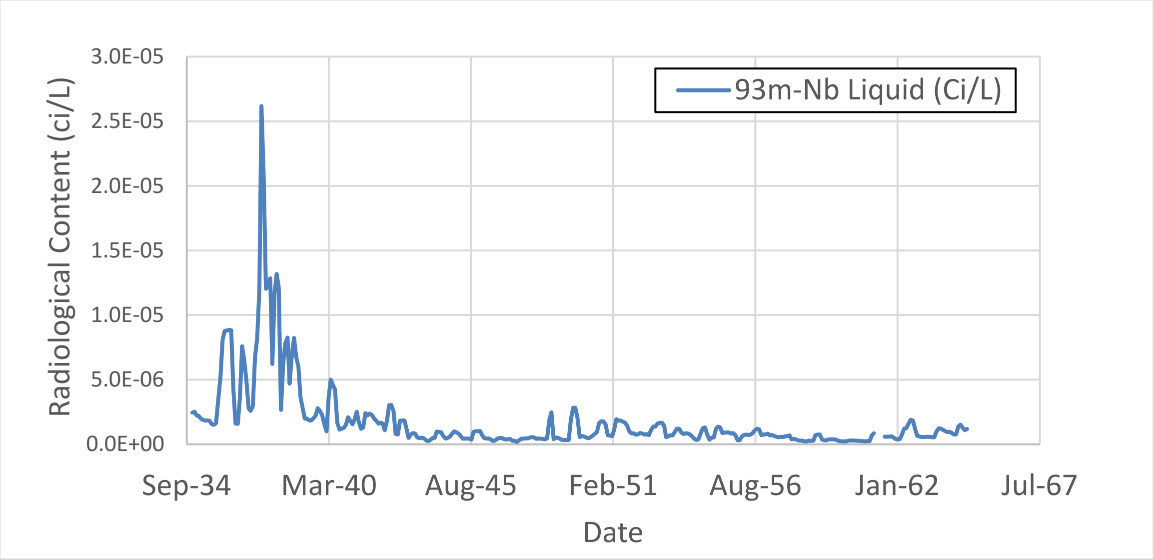
APPENDIX C
FEED VECTORS FOR SELECT RADIOLOGICAL CONSTITUENT

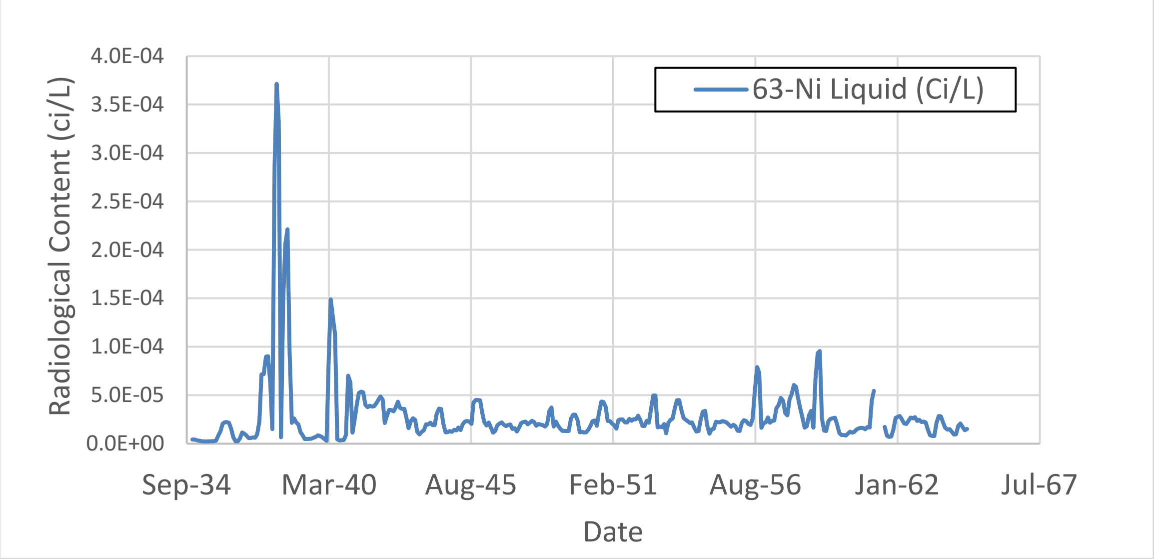

241-Am Liquid (Ci/L)
Radiological Content (ci/L)
1.2E-04
1.0E-04
8.0E-05
6.0E-05
4.0E-05
2.0E-05
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

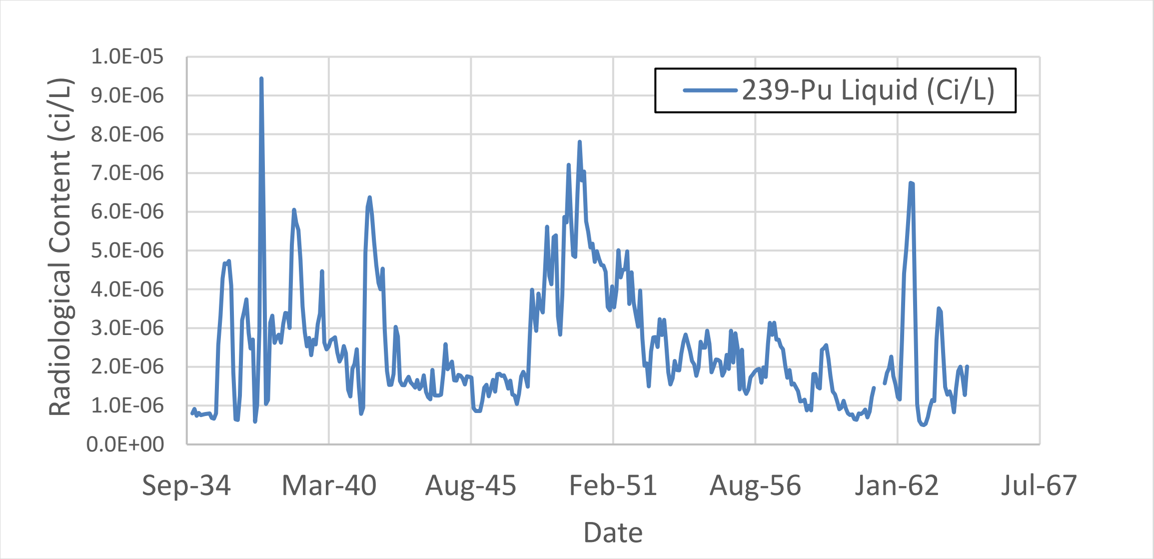

137m-Ba Liquid (Ci/L)
Radiological Content (ci/L)
9.0E-05
8.0E-05
7.0E-05
6.0E-05
5.0E-05
4.0E-05
3.0E-05
2.0E-05
1.0E-05
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

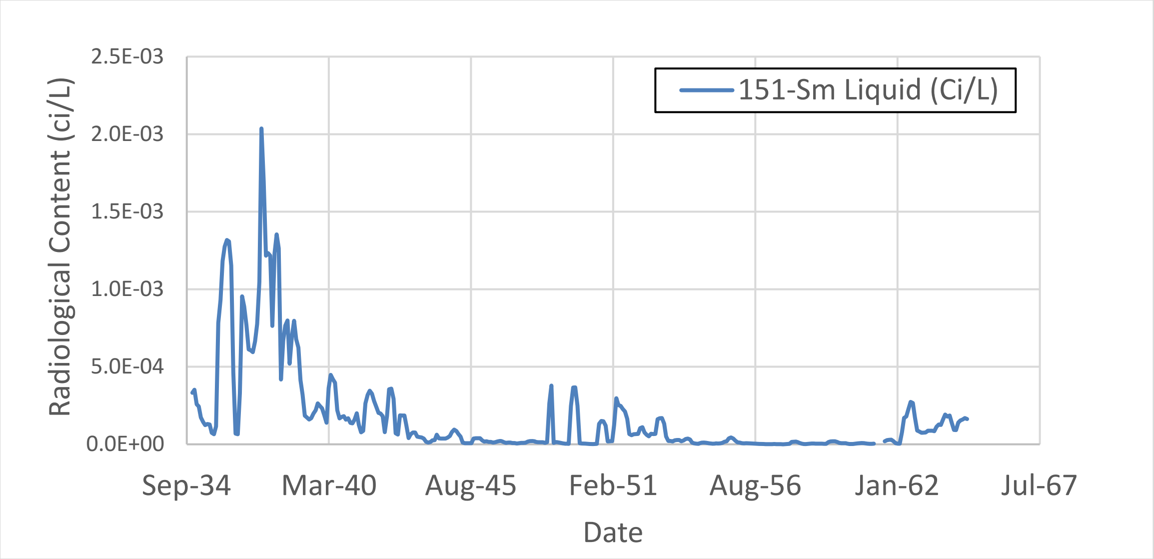

14-C (Ci/L)
Radiological Content (ci/L)
4.5E-06
4.0E-06
3.5E-06
3.0E-06
2.5E-06
2.0E-06
1.5E-06
1.0E-06
5.0E-07
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

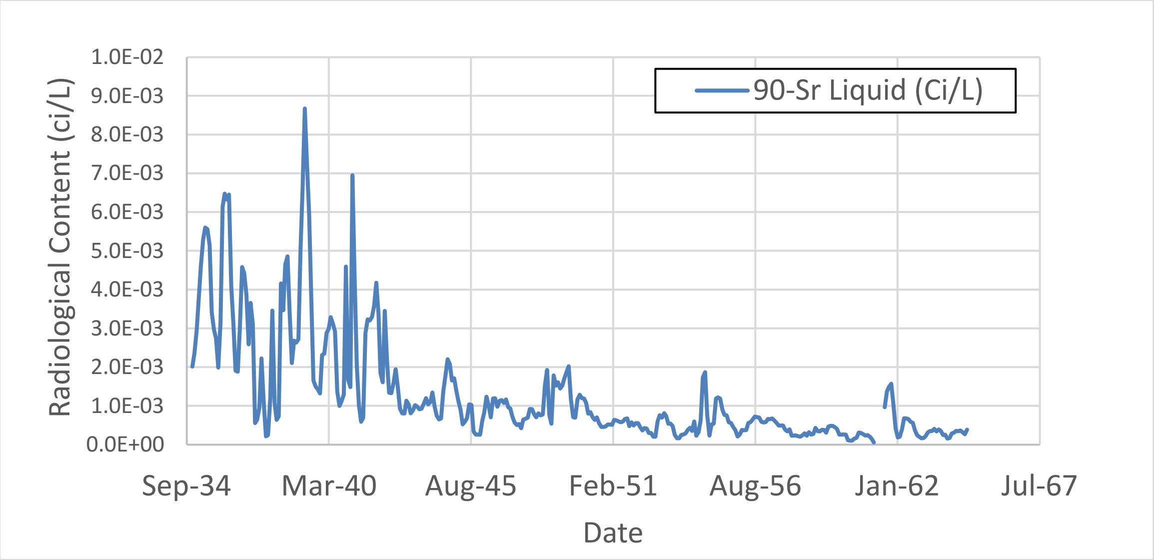

137-Cs Liquid (Ci/L)
Radiological Content (ci/L)
1.0E-04
9.0E-05
8.0E-05
7.0E-05
6.0E-05
5.0E-05
4.0E-05
3.0E-05
2.0E-05
1.0E-05
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

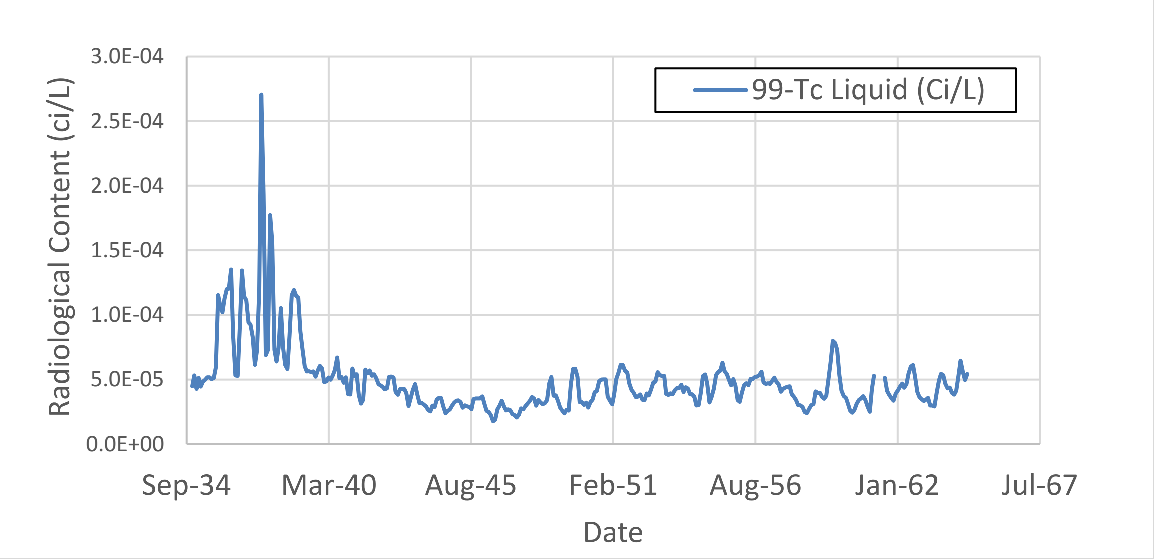

93m-Nb Liquid (Ci/L)
Radiological Content (ci/L)
3.0E-05
2.5E-05
2.0E-05
1.5E-05
1.0E-05
5.0E-06
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

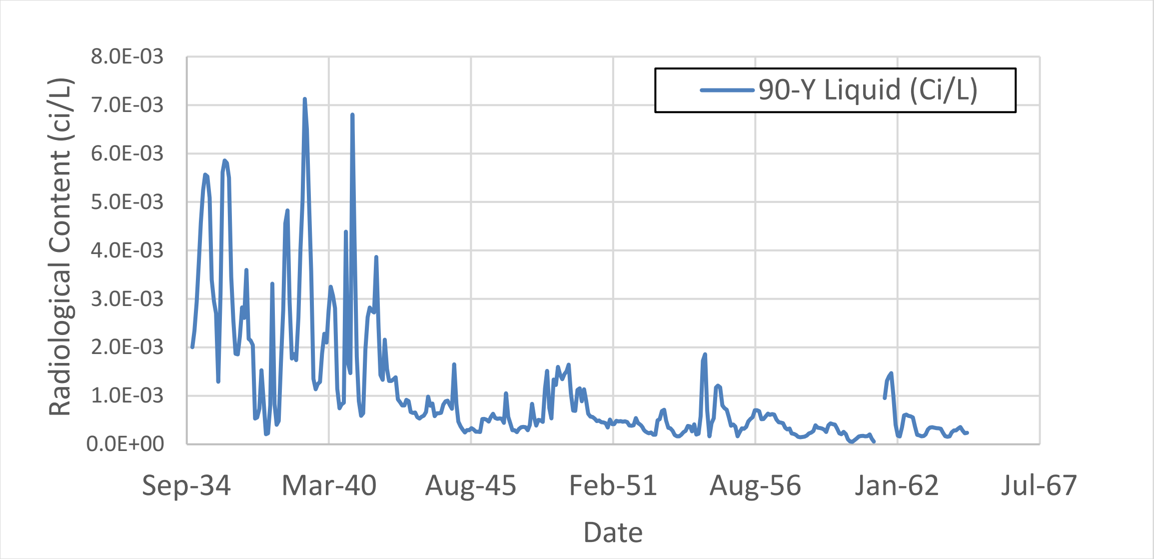

63-Ni Liquid (Ci/L)
Radiological Content (ci/L)
4.0E-04
3.5E-04
3.0E-04
2.5E-04
2.0E-04
1.5E-04
1.0E-04
5.0E-05
0.0E+00
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date

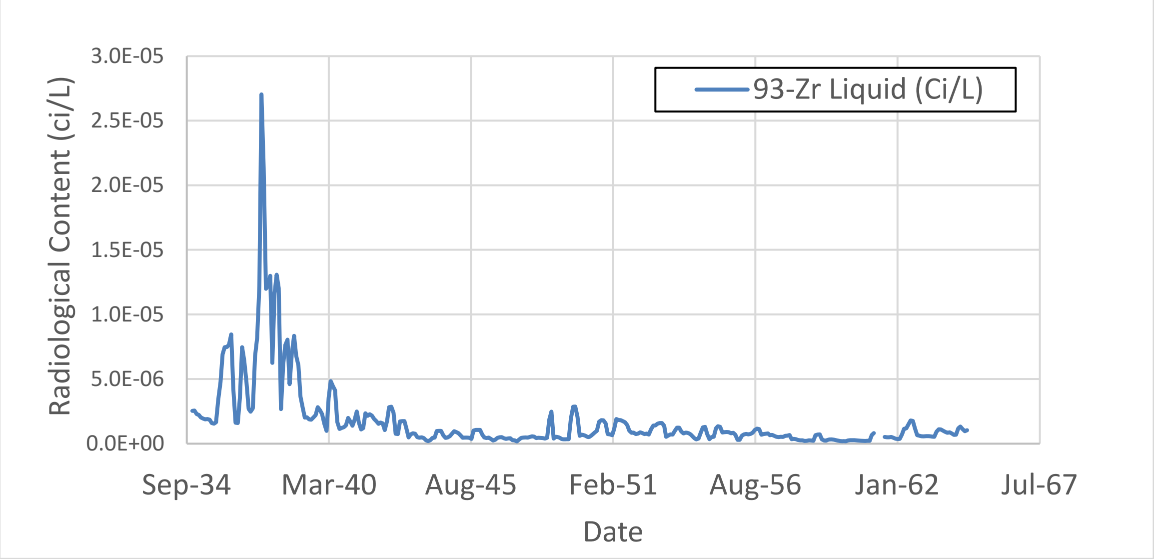

Radiological Content (ci/L)
1.0E-05
9.0E-06
8.0E-06
7.0E-06
6.0E-06
5.0E-06
4.0E-06
3.0E-06
2.0E-06
1.0E-06
0.0E+00
239-Pu Liquid (Ci/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date


Radiological Content (ci/L)
2.5E-03
2.0E-03
1.5E-03
1.0E-03
5.0E-04
0.0E+00
151-Sm Liquid (Ci/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date


Radiological Content (ci/L)
1.0E-02
9.0E-03
8.0E-03
7.0E-03
6.0E-03
5.0E-03
4.0E-03
3.0E-03
2.0E-03
1.0E-03
0.0E+00
90-Sr Liquid (Ci/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date


3.0E-04
2.5E-04
2.0E-04
1.5E-04
1.0E-04
5.0E-05
0.0E+00
Radiological Content (ci/L)
99-Tc Liquid (Ci/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date


8.0E-03
7.0E-03
6.0E-03
5.0E-03
4.0E-03
3.0E-03
2.0E-03
1.0E-03
0.0E+00
Radiological Content (ci/L)
90-Y Liquid (Ci/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date


3.0E-05
2.5E-05
2.0E-05
1.5E-05
1.0E-05
5.0E-06
0.0E+00
Radiological Content (ci/L)
93-Zr Liquid (Ci/L)
Sep-34
Mar-40
Aug-45
Feb-51
Aug-56
Jan-62
Jul-67
Date