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Assessment of Technologies for I-129 Remediation in the 200-UP-1 Operable Unit

September 2019

MJ Truex
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Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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Pacific Northwest National Laboratory
Richland, Washington 99354

Summary

Iodine-129 (I-129) generated at the U.S. Department of Energy Hanford Site during plutonium production was released to the subsurface, resulting in several large, dilute plumes in the groundwater, including the plume in the 200-UP-1 operable unit (OU). A requirement in the Record of Decision (ROD) for interim remedial action at the 200-UP-1 OU is to "evaluate potential treatment options for I-129 as part of the selected remedy through further technology evaluation. If one or more viable technologies are identified, treatability tests will be conducted for those technologies." The requirements also state: "In the event a viable treatment technology is not available, the use of a technical impracticable waiver under 40 CFR 300.430(f)(l)(ii)(c) may need to be considered as part of the final remedy." Recent work has increased the understanding of iodine behavior in the Hanford subsurface. A thorough review of potential remediation technologies has also been completed, including laboratory testing of promising technologies to evaluate their effectiveness for the iodine conditions at the 200-UP-1 OU. Using the combined information from these efforts, this report evaluates potential remediation technology options for the 200-UP-1 OU with respect to the need for conducting treatability tests and to document the technology status in support of future OU decisions for addressing the I-129 plume.

Iodine chemical speciation and isotopic distribution in the subsurface, plume size, iodine transport behavior, depth to groundwater, and infrastructure limitations affect the effectiveness, implementability, and cost of potential remedies. For instance, data indicate that stable iodine (I-127) concentrations are two- to three-orders of magnitude greater than I-129 concentrations and that over two-thirds of the iodine in groundwater is present as iodate, a quarter is present as organo-iodine, and less than 5% is present as iodide. Attenuation mechanisms have been identified, but the plume is expected to persist for more than 150 years. The plume core (above 10 pCi/L) is located directly beneath the current Environmental Restoration Disposal Facility (ERDF) disposal cells and the path of plume-core migration continues beneath the ERDF expansion zone. These factors, along with the technology information collected from review of technology sources and recent site-specific laboratory testing, were considered in the technology evaluation. The evaluation examined technologies in terms of effectiveness, implementability, and relative cost similarly to how the screening evaluation is conducted for a *Comprehensive Environmental Response, Compensation, and Liability Act* feasibility study. This evaluation also accounted for the maturity of the technologies with respect to the ability for viable consideration in a feasibility study.

The technology evaluation did not identify any technologies that need treatability testing to support remedy decisions. The practicability of all candidate technologies is low driven by site and contaminant properties that hinder effectiveness and/or implementability of the technologies. The compiled information in this report is intended to be a resource for 200-UP-1 OU decisions, including the technical assessment of whether groundwater restoration is attainable from an engineering perspective, as would be required for consideration of a technical impracticability waiver.

Acknowledgments

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Acronyms and Abbreviations

ARAR	Applicable or Relevant and Appropriate Requirements
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
DOE	U.S. Department of Energy
EIC	Effectiveness, Implementability, and Cost
EPA	U.S. Environmental Protection Agency
ERDF	Environmental Restoration Disposal Facility
MNA	monitored natural attenuation
OU	operable unit
P&T	pump and treat
PNNL	Pacific Northwest National Laboratory
PRB	permeable reactive barrier
ROD	Record of Decision
SVE	soil vapor extraction
TI	technical impracticability (waiver)

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1.0 Introduction

Isotopes of iodine were generated during plutonium production within the nine production reactors at the U.S. Department of Energy (DOE) Hanford Site. Reactor operations at the Hanford Site generated iodine-129 (I-129), an iodine isotope with a 16-million-year half-life, which was 1) stored in single-shell and double-shell tanks, 2) discharged to liquid disposal sites (e.g., cribs and trenches), 3) released to the atmosphere during fuel reprocessing operations, or 4) captured by off-gas absorbent devices (silver reactors) at chemical separations plants (PUREX, B-Plant, T-Plant, and REDOX). Releases of I-129 to the subsurface have resulted in two large dilute plumes in groundwater, including the plume in the 200-UP-1 operable unit (OU).

A requirement in the Record of Decision (ROD) for Interim Remedial Action, Hanford 200 Area Superfund Site, 200-UP-1 Operable Unit (EPA et al. 2012) is to “evaluate potential treatment options for I-129 as part of the selected remedy through further technology evaluation. If one or more viable technologies are identified, treatability tests will be conducted for those technologies.” The requirements also state: “In the event a viable treatment technology is not available, the use of a technical impracticable waiver under 40 CFR 300.430(f)(l)(ii)(c) may need to be considered as part of the final remedy.” Currently, groundwater in the 200-UP-1 OU is contaminated with carbon tetrachloride, uranium, nitrate, chromium (total and hexavalent), I-129, technetium-99 (Tc-99), and tritium. The preferred alternative in the ROD specifies 35 years active remediation using groundwater pump-and-treat, monitored natural attenuation (MNA) for portions of the contaminated groundwater, and institutional controls until cleanup levels for unrestricted use are met (EPA et al. 2012). Hydraulic containment of groundwater is being performed while a remediation technology evaluation for I-129 is performed to support a subsequent remedial decision for the I-129 plume.

The plan developed for the ROD-required I-129 remedial technology evaluation (DOE 2017) included activities to improve understanding of iodine behavior in the subsurface, compile information relevant for a decision to support a final ROD, and compile and evaluate I-129 remediation technology information in this context. The plan included a flow chart to show the relationship of these activities and the associated decisions and actions to be performed (Figure 1). Recent work has increased the understanding of iodine behavior in the Hanford subsurface as a refinement of the conceptual site model. A thorough review of potential remediation technologies has also been completed, including a literature study and laboratory testing of promising technologies to evaluate their effectiveness for the iodine conditions at the 200-UP-1 OU. Using the combined information from these efforts, this report evaluates potential remediation technology options for the 200-UP-1 OU with respect to the need for conducting treatability tests and to document the technology status in support of future operable unit decisions for addressing the I-129 plume. The scope of this report is identified by the dark blue box in Figure 1.

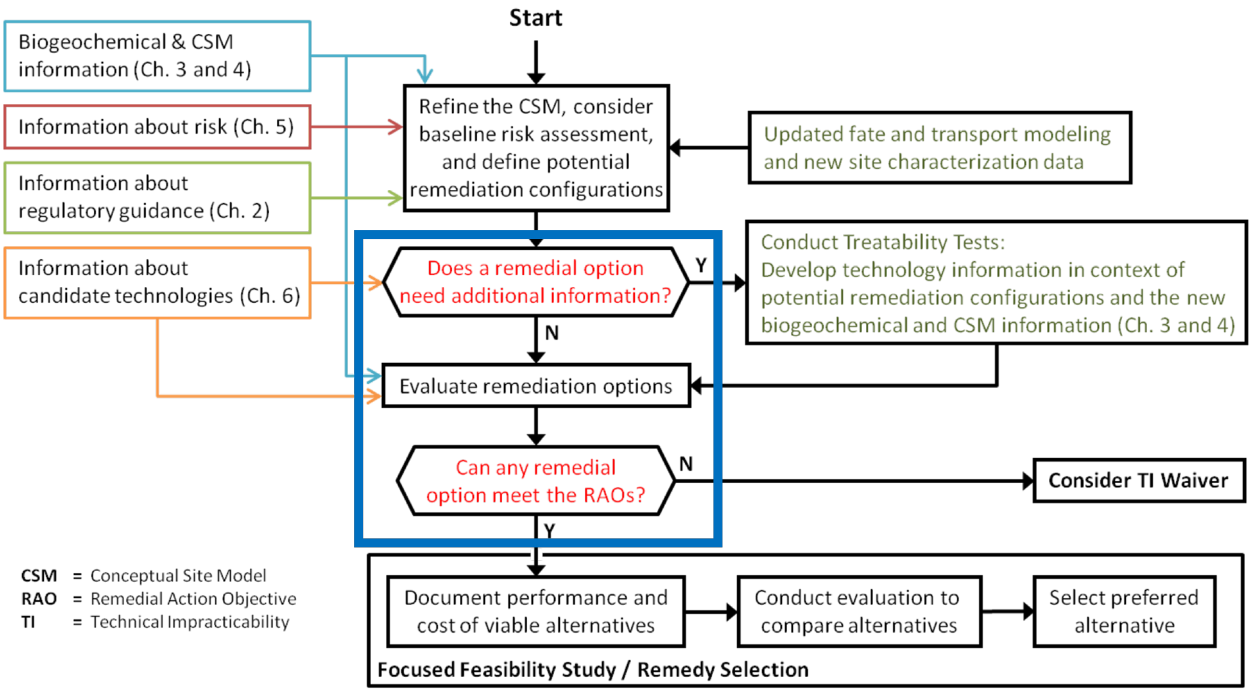

Figure 1. Flow chart for I-129 remedy evaluation (adapted from DOE 2017). The blue box highlights the elements of this process included in this report.

Previous work supporting elements of the I-129 remedy evaluation have been published and were used to provide context for the technology evaluation herein. A thorough review of potential I-129 remediation options was documented by Strickland et al. (2017a). That report described remediation technology options and evaluated these options to identify information needs for some of the more promising technologies that could be addressed in laboratory scoping studies. These laboratory scoping studies, documented in Strickland et al. (2017b) and Szecsody et al. (2019), provided laboratory data for these technologies to augment the available literature data. In addition to remediation technology efforts, refinements to the conceptual site model (Truex et al. 2017; Qafoku et al. 2018; Neeway et al. 2019), and information needed as part of considering a technical impracticability (TI) waiver (Rockhold et al. 2017, 2019) have been published. These documents provide site context to be considered in the technology evaluation process as identified in Figure 1.

This report includes information about I-129 remediation technologies in the context of their potential for use in the 200-UP-1 OU and meets the requirements of the I-129 remedy evaluation plan (DOE 2017) for technology evaluation. Section 2 describes the site setting for the 200-UP-1 OU. A summary of previous technology reviews is provided in Section 3. The approach for assessment of technologies is provided in Section 4, with results presented in Section 5. Section 6 contains a summary and the results of this evaluation.

2.0 200-UP-1 Operable Unit Site Setting

The 200-UP-1 groundwater OU, located on the DOE Hanford Site in southeastern Washington State, consists of the groundwater beneath the southern portion of the 200 West Area within the Central Plateau, as shown in Figure 2. Groundwater in the 200-UP-1 OU is contaminated with carbon tetrachloride, uranium, nitrate, chromium (total and hexavalent), I-129, Tc-99, and tritium. The DOE Richland Operations Office is the lead agency for remediation of the 200-UP-1 OU and the U.S. Environmental Protection Agency (EPA) is the lead regulatory agency, as identified in Section 5.6 and Appendix C of the Tri-Party Agreement.[1] In accordance with the Tri-Party Agreement, Article XIV, Paragraph 54, DOE developed and proposed remedial action for the 200-UP-1 OU through completion and approval of a remedial investigation/feasibility study (DOE 2012). The Record of Decision (ROD, EPA et al. 2012) was signed by EPA, DOE, and the Washington State Department of Ecology on September 27, 2012. The selected interim remedy was chosen in accordance with the *Comprehensive Environmental Response, Compensation, and Liability Act* (CERCLA), as amended by the *Superfund Amendments and Reauthorization Act of 1986*, the Tri-Party Agreement, and, to the extent practicable, the National Contingency Plan (40 CFR 300). This decision is based on the Administrative Record file for the 200-UP-1 OU.

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[1] Hanford Federal Facility Agreement and Consent Order by Washington State Department of Ecology, United States Environmental Protection Agency, United States Department of Energy, as Amended Through March 28, 2018, 89-10, Rev. 8.

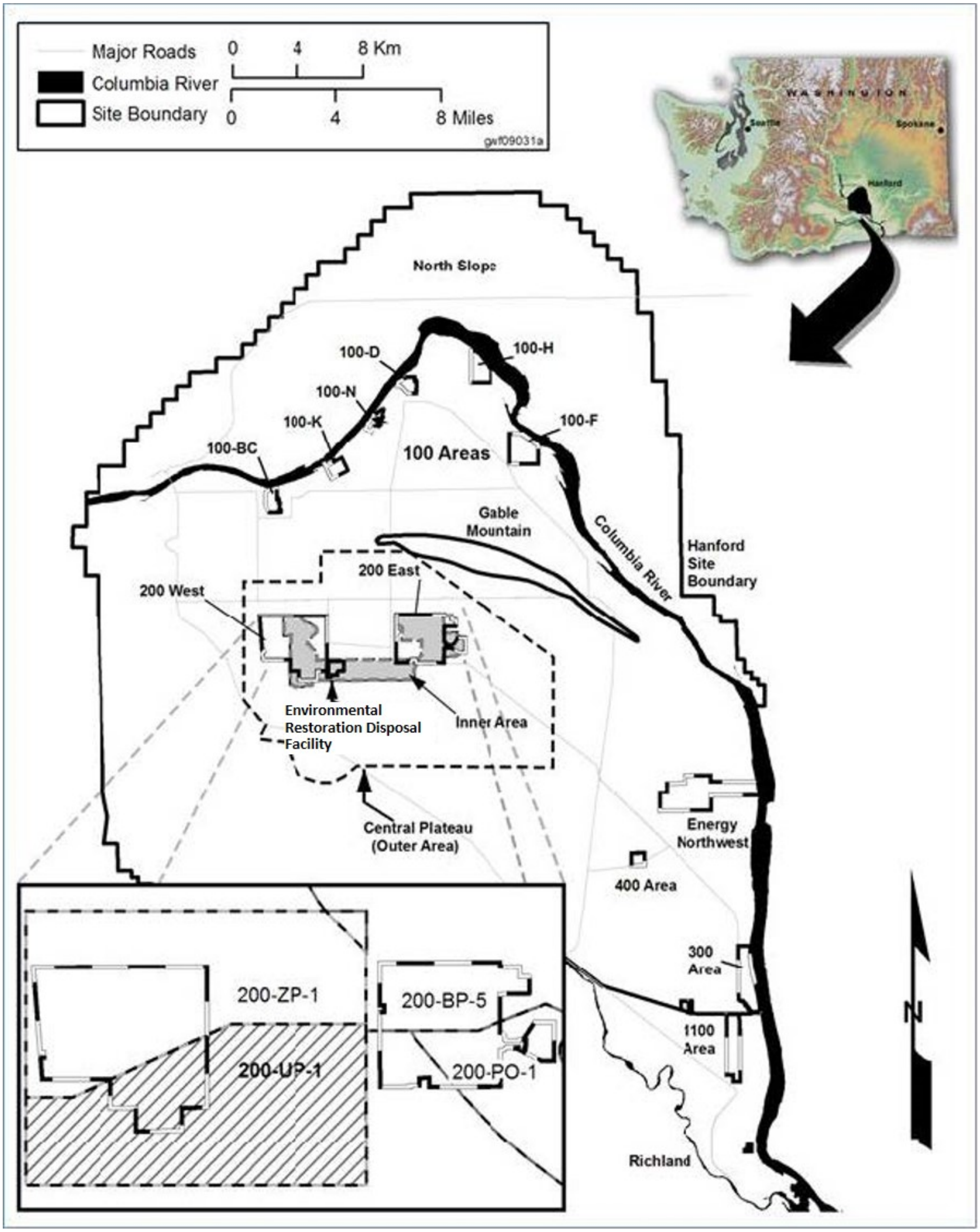


Figure 2. Location of 200-UP-1 Operable Unit on the Central Plateau at the Hanford Site (DOE 2013)

For the 200-UP-1 OU, the current I-129 plume originated from U-Plant (216-U-1 and 216-U-2 cribs) and REDOX Plant waste sites (e.g., 216-S-1&2, 216-S-7, and 216-S-9), with the REDOX Plant waste sites being the primary sources (DOE 2014). Plume maps for 1993 and 2017 (Figure 3) show that the 200-UP-1 I-129 plume has an overall areal extent (as defined by the 1 pCi/L contour) that has declined with minimal downgradient migration. The plume core area with I-129 concentrations greater than 10 pCi/L has also decreased. The overall plume extent is large and the plume thickness is up to tens of meters, although there is uncertainty in this estimate. Recent I-129 concentration results range from 1 pCi/L (i.e., at the maximum contaminant level) to about 20 pCi/L within the 200-UP-1 plume.

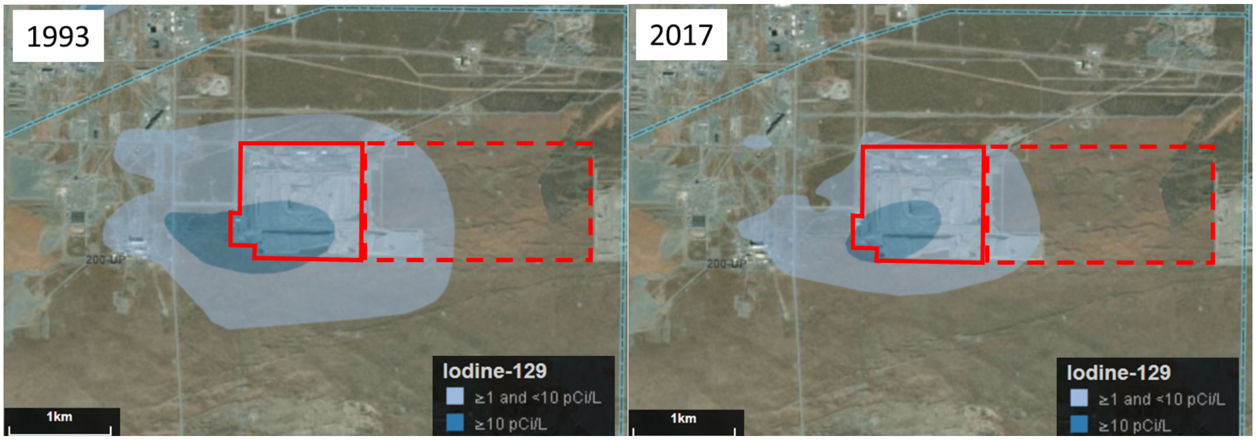
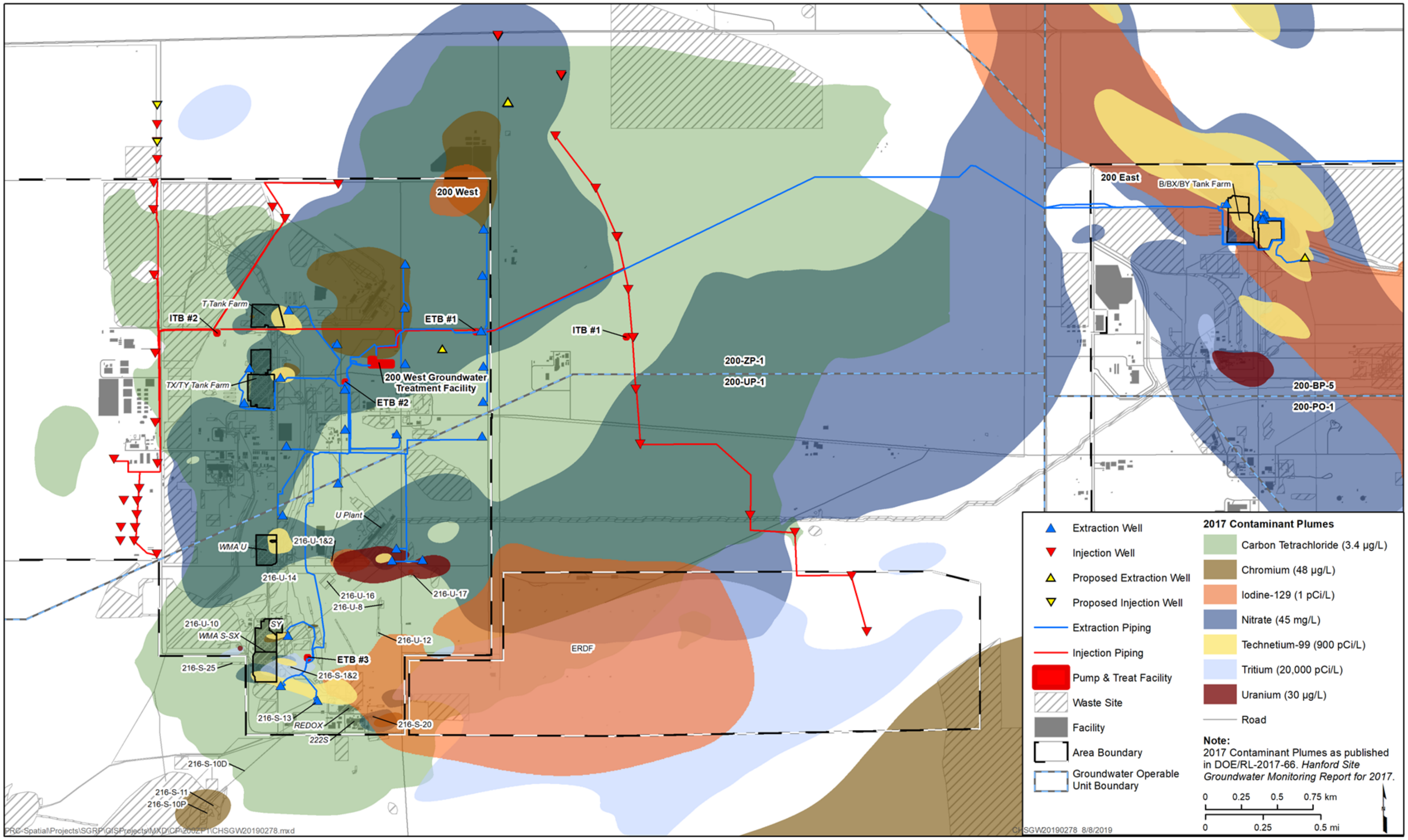


Figure 3. Plume maps for I-129 for 1993 and 2017 from DOE (2018a) as displayed by the PHOENIX web application plume animator (https://phoenix.pnnl.gov/apps/plumes/index.html). Solid red line show the current Environmental Restoration Disposal Facility (ERDF). The dotted red line shows the eastward future expansion area of ERDF.

Several co-contaminants are present within the I-129 plumes, including chromium, nitrate, Tc-99, uranium, and tritium. However, a large portion of the I-129 plume does not intersect with other contaminant plumes above the drinking water standards (Figure 4) other than tritium. Both I-129 and I-127 (stable, non-radioactive iodine) are present within the I-129 with an $^{127}IO_3^-/^{129}IO_3^-$ ratio of up to 1000 (Levitskaia et al. 2017), indicating much higher I-127 concentrations in the groundwater. The source of I-127 is not known, but iodine commonly exists as a trace constituent of nitric acid (Truex et al. 2017). Therefore, the enormous volumes of nitric acid used during operations are a likely contributing source for the groundwater I-127. For assessment of potential remediation approaches, co-contaminants are not significant interferences. However, the presence of high I-127 concentrations compared to I-129 concentrations must be considered because they exhibit the same geochemical behavior.



Note:
2017 Contaminant Plumes as published in DOE/RL-2017-66. *Hanford Site Groundwater Monitoring Report for 2017.*

Figure 4. 200 West Area contaminant plumes and pump-and-treat system wells (adapted from DOE 2018b).

Future plume behavior depends on fate and transport factors, including natural attenuation processes, and the duration of the current hydraulic control injection element of the ROD (Truex et al. 2017). Analysis of groundwater samples from the 200 West Area (Zhang et al. 2013) show a mix of iodine species present with, on average, about 70% of the iodine present as IO_3^-, about 26% as organo-iodine, and a small amount (about 4%) as I^-. In addition, sequential extraction of Hanford sediment samples (Xu et al. 2015) showed a significant fraction of iodine in sediment-associated phases in addition to aqueous and adsorbed phases.

Neeway et al. (2019) described a network of biogeochemical processes that control the fate and transport behavior of iodine in the subsurface (Figure 5). These processes are important in regard to natural attenuation of I-129. The rate and extent of these reactions are dependent on iodine concentration, redox potential, pH, the presence of organic matter (NOM), redox-sensitive elements in minerals (e.g., iron and manganese), and microbial enzymatic activity. This process model is based on iodine work by Chang et al. (2014) and Truex et al. (2017).

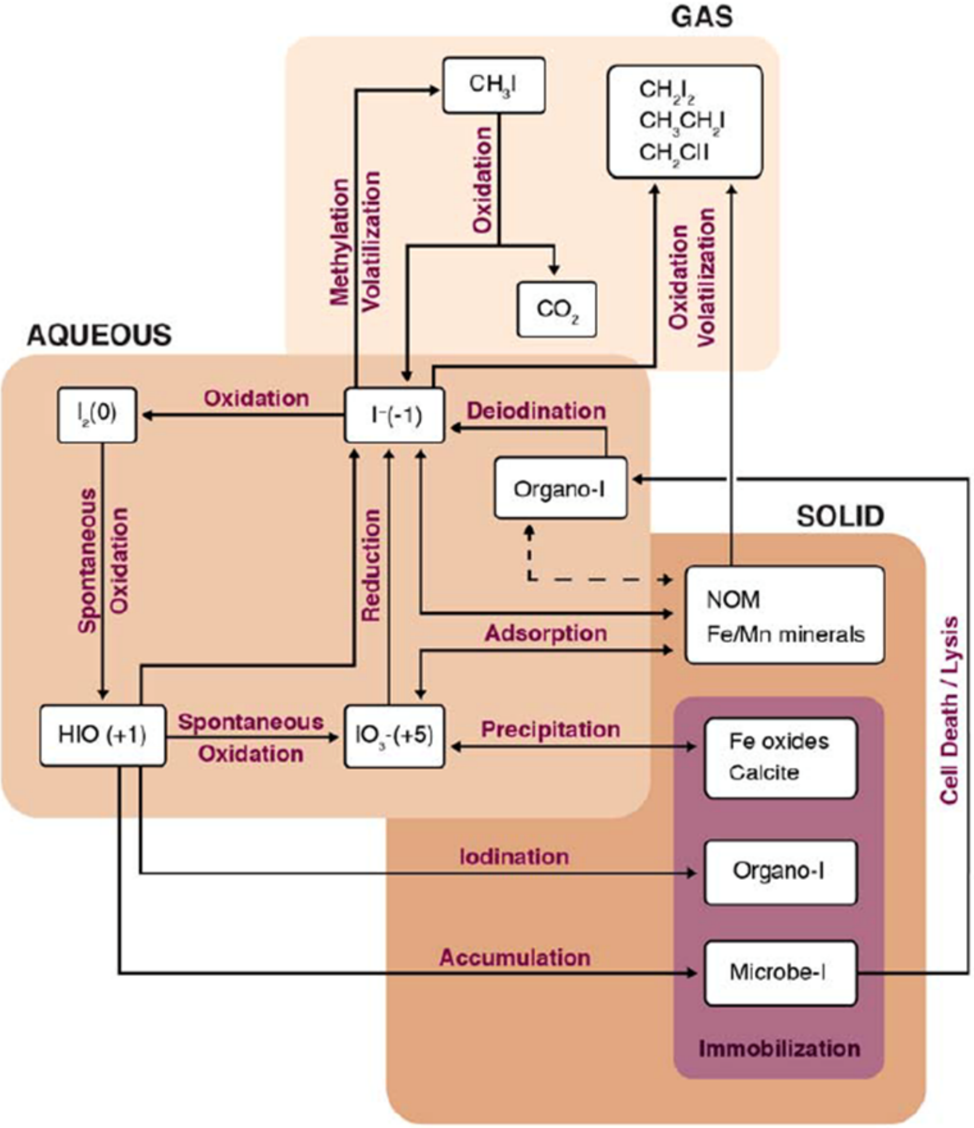


Figure 5. Hanford site biogeochemical process for iodine (adapted from Neeway et al. 2019)

Figure 5 contains three main blocks of grouped processes: The GAS, AQUEOUS, and SOLID blocks. The gas phase (GAS block) includes iodine methylation reactions. The iodine from CH_3I can be redeposited in the aqueous phase through oxidation of the methyl group, where the products of this reaction are iodide and CO_2.

The AQUEOUS block is where much of the iodine cycling important to groundwater occurs. The predominant iodine species are iodide, iodate, and organic iodine complexes (organo-I). All three of these species typically exist simultaneously in groundwater with relative concentration depending on the groundwater biogeochemical conditions. Oxidation processes cycle iodide cycles through I_2 and HIO to iodate. I_2 and HIO are less thermodynamically stable species and are thus less prevalent in the aqueous phase. HIO can be oxidized to iodate or reduced to iodide. Iodate can also be reduced to iodide. Organo-I complexes may also exist in solution. Iodide can also be produced by deiodination of aqueous organo-I.

Solid-phase interactions and species (SOLID block) are also important for iodine in the subsurface. Sorption of iodide, iodate, and organo-I compounds to organic matter and minerals is a baseline process in the subsurface that retards iodine movement relative to water movement. Iodate, in particular, associates with iron oxides and carbonates. Iodate in these solid phases exchanges with the aqueous phase through sorption and dissolution/precipitation interactions. This set of interactions represents an attenuation mechanism that slows the flux of iodate toward the groundwater. Iodine can complex with organic compounds that are associated with the solid rather than aqueous phase. Microbial interactions with iodine are also important. Microbial reactions are associated with many of the iodine aqueous-phase transformations and microbes in biofilms associated with the sediments can be a zone of accumulation for iodine. Iodine accumulated with microbes is cycled back to the aqueous phase through the cycle of cell death and lysis.

3.0 Summary of Previous Technology Information Compilations

A broad set of potentially applicable iodine remediation methods were identified in the *UP-1 Evaluation Plan for Iodine* (DOE 2017) based on a literature search and prior reports. The evaluation plan grouped potential remediation technologies into categories reflecting the location of the treatment, which related to the function of the treatment. The categories included ex situ treatment (for media extracted from the subsurface or secondary waste streams), surface source removal, control of contaminant flux through the vadose zone, and groundwater plume remediation. The evaluation plan provided a brief description of each technology, notes on technology maturity, and a qualitative categorization of technology cost (i.e., low, moderate, or high). As part of implementing the remedy evaluation, additional technology information review and laboratory scoping studies were conducted as described in the sections below.

3.1 Technology Information Review

A subsequent, more detailed review of potential I-129 remediation technologies was conducted to augment the list of potential technologies in the *UP-1 Evaluation Plan for Iodine* (DOE 2017) and identify those for which additional information was needed in the form of scoping laboratory studies to support a determination of the need for further treatability testing or to provide technology information for consideration of a TI waiver (Strickland et al. 2017a). The development status was defined using the codes shown in Table 1, which ranged from a conceptual remediation technology to a remediation technology that has been demonstrated for I-129. Multiple development status codes were allowed per technology. The remediation technologies were grouped as in situ technologies (Table 2) and ex situ technologies (Table 3).

Table 1. Codes used to describe the development status of I-129 remediation technologies (Strickland et al. 2017a).

Code	Description of Technology Status
1	Demonstrated for iodine (one or more species) in the field
2	Demonstrated for another contaminant in the field
3	Demonstrated in the laboratory for iodine (one or more species)
4	Demonstrated for another contaminant in the laboratory
5	Literature (journal articles or technical report) is available showing a basis for the technology
6	Conceptual remediation technology supported by literature/paper study/modeling

Table 2. In situ groundwater remediation technologies (adapted from Strickland et al. 2017a).

Technology	Brief Description	Status
Monitored Natural Attenuation (MNA)	Natural processes reduce the mass or concentration of the contaminant plume.	2, 3, 4, 5
In Situ Sequestration by Calcite (iodate)	Volumetric or permeable reactive barrier (PRB) treatment to co-precipitate iodate with calcite mineral(s).	3
In Situ Sequestration by Apatite (iodate)	Volumetric or PRB treatment to incorporate iodate into apatite mineral(s).	2, 3, 4, 5
In Situ Sequestration by Bioaccumulation	Uptake and sequestration of iodine compounds by microbes.	5, 6
Microbial Facilitated Volatilization	Microbial iodine methylation + soil vapor extraction (SVE) capture of methyl iodide.	5, 6
Enhanced Pump and Treat	P&T with injection of compounds to increase iodine mobility.	2, 3, 6
In Situ Sequestration by Iron Oxides (iodate)	Volumetric treatment to co-precipitate iodate with iron oxide compounds.	3, 5, 6
In Situ Sequestration by Organic Carbon	Volumetric or PRB treatment to accumulate iodate or iodide into stable, low-solubility organic matter.	5, 6
In Situ Sequestration by Alkaline Treatment	Use alkaline treatment followed by buffering to dissolve aluminosilicate to promote “armoring” compounds that coat/incorporate iodine and decrease long-term iodine mobility.	4
In Situ Sequestration by Metals (iodide)	Volumetric or PRB treatment to create Ag-, Hg-, or Cu-iodide compounds.	1, 3
In Situ Sequestration by Iron Sulfide	Volumetric treatment to precipitate iodine with iron sulfide compounds.	5, 6
Zero Valent Iron	Nano- or micro-sized zero valent iron particles to promote hydrogen generation and redox conditions that facilitate related sequestration reactions.	2, 4, 6
Microbial oxidation process	Microbial oxidation of iodide to promote cycling of iodine to forms suitable for sequestration or reduced mobility.	5, 6
Physical Groundwater Flow Barrier	Use of a physical barrier (e.g., grout curtain, freeze wall) for plume containment or to direct groundwater flow.	2
Air Sparging	Injection of air into the aquifer to facilitate mass transfer from aqueous to gas phases + SVE capture.	2, 6
Acid Induced Volatilization	Acidify to pH 3 or lower to form iodine gas + SVE capture.	6
Vacuum Induced Volatilization	Simple vacuum to induce volatilization from groundwater + SVE capture.	2, 6
Electrokinetic Remediation (enhanced)	Subsurface electrodes promote iron precipitation, an acid front, contaminant desorption, and ion migration.	2, 3,4, 5
Extraction Wells (P&T)	Groundwater and dissolved contamination are extracted via wells, with aboveground treatment of the water.	1
Horizontal Wells	Variant of standard vertical extraction wells.	2
Targeted P&T + MNA for lower concentrations	Combination of P&T and MNA.	1, 2

Table 3. Aboveground treatment technologies for aqueous-, gas-, and solid-phase media (adapted from Strickland et al. 2017a).

Technology	Brief Description	Status
Adsorption from Water	Capture of iodine from water onto solid-phase sorbent media (e.g., activated carbon, silver-impregnated carbon, zeolite, layered double hydroxides, organoclay, argentite, aerogels, SAMMS (self-assembled mercaptan on mesoporous silica), carbon nanotubes, photocatalytic adsorption).	2, 3, 4, 5
Ion Exchange	Mass transfer process where ions from solid phase media [e.g., commercial or ABEC (aqueous biphasic extraction chromatographic) resins] are exchanged for ions in solution.	2, 3, 4, 5
Microbial Enhanced Ion Exchange	Microbial facilitated iodine reduction to facilitate/enhance ion exchange.	2, 4
Gas Phase Adsorption	Capture of iodine from gas on solid phase adsorbents (e.g., activated carbon, macroreticular resins, silver-impregnated alumina, silver exchanged mordenite, silver-loaded zeolite, silver impregnated silica-gel, bismuth-based sorbents, or silver-functionalized silica aerogels).	1, 2, 3, 4, 5
Membrane Separation – Reverse osmosis	Selective transmittal of water across a membrane due to hydrostatic pressure.	2, 3, 4, 5
Membrane Separation – Electrodialysis	Separation of ions from water by applying an electric current across a membrane.	2, 3, 4, 5
Electrochemical Separation – Capacitive Deionization	Current applied across porous activated or aerogel carbon electrodes captures ions in the electrodes.	2, 3, 4, 5
Electrochemical Separation – Electrochemically Switched Ion Exchange	A switched electrode potential is used to capture ions by adsorption on an electroactive film at the electrode.	2, 3, 4, 5
Electrochemical Separation – Electrolysis	Uses electrolysis to oxidize halides.	4, 5
Co-precipitation and Coagulation	Aqueous precipitation of iodine with an Ag, Cu, Hg, or Pb compound.	2, 4, 5
Gas Phase Absorber (scrubber)	Preferential partitioning of iodine from gas phase to a liquid phase (e.g., caustic, mercuric nitrate-nitric acid, hyperazeotropic nitric acid, fluorocarbon, polymethylsiloxane, molten hydroxide).	1, 2, 3, 4, 5
Soil Washing	Ex situ scrubbing of soils using water-based solution, possibly with surfactants/additives.	2, 4, 5
Immobilization/ Encapsulation and Solidif./Stabilization, cementitious waste forms	Encapsulation of iodine in a solid monolith with requisite leaching characteristics; cast stone/cementitious encapsulation.	2, 3, 4, 5
Glass Waste Form	Vitrification of solid phase to a glass waste form.	2, 3, 4, 5
Adsorption/ Vitrification	Capture iodine from gas onto nanoporous alumina, precipitate with Ag, Cu, or Na_4SiO_4 then vitrify the alumina/iodine + glass formers to encapsulate the iodine.	3, 5
Low-Temperature Vitrification	Iodine is captured on an adsorbent and is subsequently sequestered in low-temperature sintering glasses (e.g., bismuth-phosphate-zinc oxide, lead-boron-zinc).	3, 5
Encapsulation in Tin	Fusing iodide sorbed on silver-loaded sorbents with tin in a hot-pressing process to produce a consolidated mass.	3, 5

Strickland et al. (2017a) used the listing of in situ remediation technologies (Table 2) and ex situ remediation technologies (Table 3) to identify technologies with promise for application under Hanford Site conditions (Table 4).

Table 4. Promising remediation technologies (adapted from Strickland et al. 2017a).

Technology	Basis for selection
Monitored Natural Attenuation (MNA)	Baseline approach, natural attenuation mechanisms have been identified.
In Situ Sequestration by Calcite (iodate)	Scientific literature and site-specific experiments have shown iodate incorporation with calcite.
In Situ Sequestration by Apatite (iodate)	Scientific literature describes iodate incorporation with apatite.
In Situ Sequestration by Iron Oxides (iodate)	Experiments and literature have shown iodate interaction with iron oxides.
In Situ Sequestration by Organic Carbon	Scientific literature describes iodate interaction with organic carbon.
In Situ Sequestration by Bioaccumulation	Scientific literature describes iodine accumulation by microorganisms.
Microbial Facilitated Volatilization	Scientific literature and site-specific experiments have shown methylation of iodide to create a volatile compound.
Enhanced Pump and Treat	Scientific literature has shown that dithionite will reduce and dissolve iron oxide precipitates from sediment which will release associated constituents.
Ex Situ Aqueous Adsorption	Scientific literature describes iodine adsorption processes.
Ex Situ Ion Exchange	Scientific literature and groundwater treatment experience indicates the potential for iodine ion exchange processes.
Microbial Enhanced Ex Situ Ion Exchange	Scientific literature describes iodate reduction to iodide, which is more readily captured on ion exchange media.

3.2 Laboratory Scoping Study Summary

Laboratory scoping studies were conducted for most of the technologies listed in Table 4 as a means to provide site-specific information for assessment of effectiveness, implementability, and relative cost. Testing focused primarily on effectiveness for addressing iodate, the most prevalent iodine species in the Hanford groundwater. Four of the technologies listed in Table 4 were evaluated and it was determined that laboratory scoping tests were not needed for these candidates. MNA did not require additional laboratory testing because it was evaluated as part of studies to refine the conceptual site model for iodine at the Hanford Site (Truex et al. 2017; Qafoku et al. 2018; Neeway et al. 2019). In situ bioaccumulation of iodine was not tested in the laboratory because literature information was sufficient to describe uptake and release characteristics for bioaccumulation (Neeway et al. 2019). Microbial facilitated volatilization was evaluated in laboratory testing that was incorporated into the refined conceptual site model for iodine at the Hanford Site (Truex et al. 2017; Qafoku et al. 2018; Neeway et al. 2019). Microbial enhanced ex situ ion exchange was not tested because other iodate-specific ion exchange resins were tested and Hanford Site experience has shown operational difficulties of biological treatment with respect to reinjection of treated water to the aquifer (DOE 2018b).

Testing of in situ technologies was organized into three broad categories: 1) in situ sequestration or removal of iodine from groundwater, 2) in situ mobilization of iodine to enhance extraction efficiency, and 3) ex situ removal of iodine from groundwater to support P&T operations.

3.2.1 In Situ Sequestration

For in situ sequestration, co-precipitation of iodate with calcium carbonate was investigated using three methods (Szecsody et al. 2019) with additional related investigations reported by Kerisit et al. (2018) and Lawter et al. (2018). The first method evaluated iodate uptake as a function of calcite precipitation rate. The second method explored the impact of solution chemistry on iodate uptake by calcite. The final method examined the impact of surface area on iodate uptake. While all three investigations demonstrated the ability to remove iodate from Hanford-representative solutions at relevant total iodine solution concentrations, none of the approaches were effective at removing more than 70% iodate from solution. This presents a serious shortcoming for in situ application; therefore, further consideration of in situ formation of calcite for remediation of I-129 is not recommended.

Precipitation of initially amorphous calcium-phosphate (which slowly crystallizes to apatite), another potential in situ treatment approach, inconsistently removed a small amount of iodate from solution at pH 11 and above, and none at pH 9.0 and 7.5 (Szecsody et al. 2019). In the 40 experiments performed as part of this study, many showed greater uptake during initial amorphous calcium phosphate precipitation, and less iodate uptake as the precipitate crystallized to hydroxyapatite. Iodate removal from solution via either sorption onto or incorporation into apatite was insufficient to meet the maximum contaminant level of 1 pCi/L in groundwater; therefore, further consideration of this technology for remediation of I-129 was not recommended.

A series of batch adsorption/desorption experiments was also conducted to determine the effectiveness of several organic materials for sequestering iodate and iodide from Hanford groundwater (Szecsody et al. 2019). The organic materials that were evaluated in this study were chitin, lignin, and humic acid sorbed to a representative Hanford sediment. Of the three organic carbon materials tested, only chitin showed potential as an in situ remediation technology for iodide (average K_d value of 74.9 ± 4.3 mL/g). However, iodine within the groundwater at 200-UP-1 is primarily in the form of iodate, which limits the effectiveness of chitin as a removal technology. As such, further consideration of this technology was not recommended.

Laboratory results indicated that iodate and iodide are effectively removed from Hanford groundwater by iron oxides, especially HFO, either through sorption or co-precipitation processes (Szecsody et al. 2019). The high sorption capacity at neutral pH conditions, low cost, and likely ability to precipitate HFO in situ indicate that this approach is a candidate for iodate and iodide remediation in the 200-UP-1 OU. This technology is considered further in this report.

3.2.2 In Situ Mobilization for Enhanced Pump-and-Treat

The use of dithionite was identified as a potential remedial approach for enhancing P&T extraction. Results demonstrated that dithionite treated sediments enabled much greater (4x or more) and rapid (one to three orders of magnitude) leaching of iodine from the sediment compared to leaching of untreated sediment (Szecsody et al. 2019). This technology may accelerate removal of iodine from the surface by P&T in areas where sorption limits extraction efficiency. This technology is considered further in this report.

3.2.3 Ex Situ Treatment

A wide range of materials for above ground treatment were also tested for their capacity to remove iodate from groundwater (Szecsody et al. 2019). Synthetic groundwater was used in the experiments to evaluate iodate removal in the presence of the competing anions. Ferrihydrite, bismuth oxy(hydroxide), and bismuth-cobalt-aluminum are the most promising materials, and the observed batch-test removal efficiency is sufficient to reduce concentrations of I-129 from 30 pCi/L to 1 pCi/L, even in the presence of a total iodine concentration loading due to the presence of stable iodine (I-127) in the groundwater at concentrations 1000 times higher than I-129. Based on promising results for effectiveness of these materials, additional testing was conducted to evaluate resin-bead forms of these materials (PAN-bismuth oxyhydroxide, PAN-bismuth subnitrate, and PAN-ferrihydrite), and similar ion-exchange resins identified from a vendor.

Resin beads selected for testing included use of a resin to encapsulate favorable materials from earlier testing and resins obtained from a resin vendor. Batch testing results showed resin sorption K_d values were in a range similar to expectations from previous material testing (Table 5). Of the resins tested, two are considered to be commercially available, CHM-20 and ASM-10-HP from ResinTech (West Berlin, NJ). The PAN-bismuth oxyhydroxide and PAN-bismuth subnitrate resins show possibility for enhanced performance using the bismuth-based materials but are not readily available for detailed consideration with these active materials. (The PAN encapsulation material is commercially available from Global Phosphate Solutions, Idaho Falls, ID.) The ASM-10-HP resin demonstrated that the commercially available resins have good effectiveness comparable to the results for the resin using the laboratory-based material. Based on these results, CHM-20 and ASM-10-HP resins were carried forward and their evaluation is documented in this report.

Table 5. Batch testing results with iodate for materials and resins with the potential for use in an aboveground treatment system for iodate in groundwater (Szecsody et al. 2019)

Material/Resin Bead Tested	Measured K_d (mL/g)
Initial test: 530E resin (P&T system)	78
Initial test: Ferrihydrite material	3,230
Initial test: Bismuth (oxy)hydroxide material	200,000
Initial test: Bi-Co-Al material	13,000
Material for use in PAN beads: Freeze-dried ferrihydrite material	83,900
Material for use in PAN beads: Bismuth oxyhydroxide material	21,400,000
Material for use in PAN beads: Bismuth subnitrate material	21,400,000
Laboratory resin beads: PAN-ferrihydrite[(a)]	2,780
Laboratory resin beads: PAN-bismuth oxyhydroxide[(a)]	475,000
Laboratory resin beads: PAN-bismuth subnitrate[(a)]	199,000
Commercial resin beads: CHM-20, Cerium material	10,200
Commercial resin beads: ASM-10-HP, Ferrihydrite material	5,890

[*] Based on 0.1 g of active sorbent.

Column tests were conducted to better assess performance of the CHM-20 and ASM-10-HP resins (Szecsody et al. 2019). These tests allowed evaluation of resin capacity and kinetics in a flow-through system. Columns were packed with the resins and 200-Area groundwater spiked with 100 µg/L iodate was injected at the resin bed velocity for the Purolite resin used in the current 200 West P&T system for treating technetium-99. The effluent was monitored for breakthrough of iodate where a threshold of 3% of the influent concentration was used to signify a breakthrough equivalent to meeting resin performance needs for I-129 in the presence of high I-127 concentrations (1000 times the I-129 concentration). Tests were conducted with I-127 and interpreted in terms of I-129 performance.

Iodate breakthrough for the tested resins is shown in Figure 6. The 3% concentration breakthrough occurred after about 125 pore volumes for both resins. These data were used to calculate a resin capacity on the order of 10 µg-iodine/g-resin for the CHM-20 and ASM-10-HP resins at (Szecsody et al. 2019) and on the order of 35 µg-iodine/g-resin at full breakthrough. As indicated by Szecsody et al. (2019), engineering of the resin column approach may render a resin capacity closer to the capacity observed in batch tests (i.e., about 200 µg-iodine/g-resin). Analysis of the current Purolite A530E resin used for Tc-99 in the 200 West P&T system showed the spent resin (spent because of Tc-99 breakthrough) contained about 50 µg-iodine/g-resin (as I-127) (Levitskaia et al. 2017). Analysis of spent uranium resin (DOWEX 21K) by Campbell et al. (2018) showed the spent resin (spent because of uranium breakthrough) contained about 35 µg-iodine/g-resin (as I-127). Note that these current resins used in the 200 West P&T system would accumulate iodine in the iodide form. Thus, the column data show the tested resins have a similar capacity to these other resins, but have the ability to retain iodate. If the resin capacity were able to approach the capacity observed in the batch tests, the capacity of the tested resins would be an order of magnitude higher (about 200 µg-iodine/g-resin).

Resin capacity is important to consider in terms of contaminant loading (i.e., the influent concentration). For the Purolite resin, the observed Tc-99 loading on spent resin is only about 20 µg-Tc-99/g-resin (Levitskaia et al. 2017), but influent concentrations are on the order of tens of ng/L (nanogram/L) compared to the µg/L range of influent concentration of iodine and uranium contaminants. For the DOWEX 21K resin, the observed uranium loading on spent resin is about 18,500 µg-U/g-resin (Campbell et al. 2018), with influent concentrations in the 100 µg/L range. For comparison, the batch capacity for the tested resins of about 200 µg-iodine/g-resin (based on the batch resin capacity) would need to address iodine concentrations on the order of tens to hundreds of µg/L. Thus, the capacity in comparison to the contaminant loading (i.e., the number of liters needed to reach contaminant capacity for a gram of resin) is much lower for the tested resins (order-of-magnitude range of 1 L/g-resin) than this ratio for uranium (order-of-magnitude range of 100s L/g-resin) or Tc-99 (order-of-magnitude range of 100s L/g-resin) on the current resins used to treat these contaminants in the 200 West P&T system.

The characteristic of each iodate breakthrough curve shown in Figure 6 includes a time when the effluent concentration is greater than the influent concentration ($C/C_o > 1$). This characteristic is attributed to interactions between the resin and the groundwater constituents. It appears that a portion of the initially sorbed iodate was released as the resin material was altered through exchange of ions between the resin and the groundwater. The likely exchange is nitrate or carbonate in the groundwater for chloride in the resin. This type of resin alteration from groundwater chemistry is unfavorable for resin performance, though preconditioning of the resins to the groundwater chemistry may alleviate some of this issue.

Based on the observed results of low capacity and the negative interactions with groundwater constituents, the performance of these resins in a flow-through system is considered marginal.

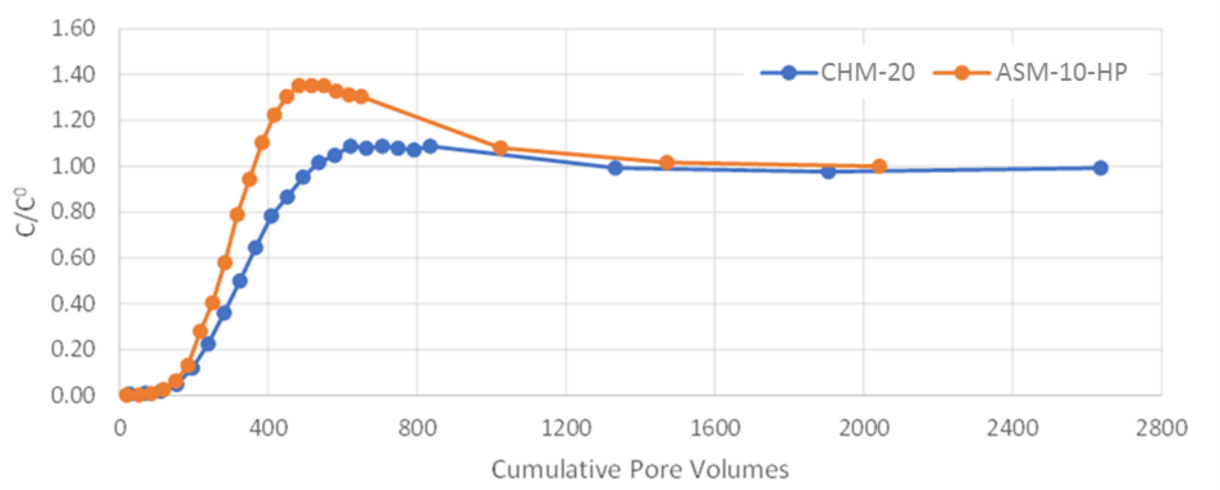


Figure 6. Column effluent concentration of iodate expressed as a ratio to the influent concentration for the cerium-based resin (CHM-20) and the ferrihydrite-based resin (ASM-10-HP) (adapted from Szecsody et al. 2019).

4.0 Technology Assessment Approach

Building on the technology reviews and recommendations in DOE (2017) and Strickland et al. (2017a), technology information, the data collected in laboratory scoping studies, logistical factors, and site-specific I-129 chemistry factors were used to evaluate the technology applicability. Technologies were evaluated with respect to the need for conducting treatability tests and to document the technology status in support of future OU decisions for addressing the I-129 plume. This evaluation used the categories of effectiveness, implementability, relative cost, and maturity in assessing each technology, similarly to how technologies would be considered in the screening element of a CERCLA feasibility study. However, this evaluation is not intended to supplant the screening process if a feasibility study were to be conducted. The approach was selected so that the results include the categories of evaluation and the site-specific factors that are consistent with consideration of technologies for a feasibility study.

The logistical factors and site-specific I-129 chemistry factors for consideration in the evaluation were identified from OU and Hanford Site information (DOE 2013, 2018a,b) and from refined conceptual site model reports (Truex et al. 2017; Qafoku et al. 2018; Neeway et al. 2019). Key factors are described in the list below.

- Depth to plume and its areal extent and thickness
  - Figure 3 shows the I-129 plume for 2017. The I-129 plume in the 200-UP-1 OU is located in the aquifer with the water table about 75 m below ground surface. The areal extent of the plume above 1 pCi/L is about 3.4 km^2. The plume thickness is on the order of 40 m.
  - Both in situ and ex situ remediation effectiveness, implementability, and cost are affected by the plume size. Plume areal extent affects the number of wells needed for extraction or for amendment distribution based on the radius of influence for the extraction or in situ treatment process. Plume thickness affects the amount of extracted water or injected amendments needed for a given area of lateral plume extent. Depth to water table and plume thickness affect the well depth needed and associated implementation factors and costs.
- Spatial relationship of ERDF and plume core
  - Figure 3 shows the location of ERDF in relation to the I-129 plume.
  - Because the current disposal cell and the planned expansion zone for ERDF are directly above the I-129 plume core, installation of wells for groundwater extraction or addition of in situ remediation amendments would be restricted to remain outside of this area.
- Presence of I-127 comingled with I-129
  - Evaluation of groundwater and ion exchange resins (Truex et al. 2017; Levitskaia et al. 2017; Campbell et al. 2018) indicates that I-127 is present in the groundwater at a concentration ranging up to 1000 times higher than the concentration of I-129.
  - Chemical species of iodine would incorporate either I-127 and I-129 and biogeochemical processes are essentially the same for either isotope. Therefore, treatment processes would need to address the total I-127 and I-129 concentration, yet have a high effectiveness to meet concentration standards that are based only on the I-129 concentration.
- Iodine chemical speciation
  - For the 200-UP-1 OU, groundwater data show that most of the iodine is present as the iodate species (Truex et al. 2017), based on measurements of I-127, which is assumed to have the same chemical speciation as I-129. Other species present in groundwater at much lower concentrations are iodide and organo-I complexes.

  - Remediation technology effectiveness is affected by the chemical form of I-129 such that treatment of iodate is important for the 200-UP-1 OU groundwater.
- Hanford subsurface geochemical conditions
  - The Hanford Site subsurface conditions control the iodine speciation and type of relevant remediation processes as described by Neeway et al. (2019). The Hanford Site conditions and associated iodine behavior is significantly different than conditions at the DOE Savannah River Site, where I-129 contamination is also a groundwater concern.
  - Because the iodine behavior at the Hanford Site is different than at the Savannah River Site, remediation approaches being considered and implemented at the Savannah River Site cannot be directly applied for the Hanford Site.

The effectiveness, implementability, relative cost, and maturity evaluation used the approach for technology screening identified in the CERCLA feasibility study guidelines (EPA 1988) for consideration of potential technologies. Strickland et al. (2017a) conducted a general screening to identify a list of potentially promising technologies, and these technologies, as amended by the subsequent laboratory scoping test efforts (Strickland et al. 2017b; Szecsody et al. 2019), were the focus of the evaluation documented in this reported.

5.0 Remediation Technology Assessment

The technologies remaining for consideration after screening in Strickland et al. (2017a) were assessed to provide information as a resource for 200-UP-1 OU I-129 remedy decisions, including the technical assessment of whether groundwater restoration is attainable from an engineering perspective, as would be required for consideration of a TI waiver. The assessment is presented in Table 6. Each technology was assessed in terms of effectiveness, implementability, relative cost, and maturity. The table includes a summary of the evaluation and supporting documentation and additional information for the effectiveness, implementability, relative cost, and maturity elements of the assessment. Conclusions are included in terms of the need for conducting treatability testing and the technical practicability of the technology for application to the 200-UP-1 I-129 plume. This table augments the technology review and assessment documented in Strickland et al. (2017a) so that, collectively, these assessments provide the technology evaluation required by the 200-UP-1 I-129 evaluation plan (DOE 2017). Note that conclusions from Strickland et al. (2017a) identified a list of promising technologies (summarized in Table 4 of this document) and all other technologies reviewed were considered impracticable.

Table 6. I-129 Remediation Technology Assessment for the 200-UP-1 Operable Unit

Technology	Evaluation Summary	Effectiveness	Implementability	Relative Cost	Maturity and Conclusion
Monitored Natural Attenuation	Attenuation processes relevant to the 200-UP-1 plume have been identified.(a) The plume will attenuate over a long time period (>100 years) while it migrates downgradient.(b)	High: The plume is attenuating.	Medium: Will need to consider long timeframe in monitoring design.	Low Cost: With appropriate monitoring design, costs can be low compared to other options.	Mature: Approach is viable if a long remediation duration is acceptable while the plume migrates downgradient.
In Situ Sequestration by Calcite (iodate)	Hanford-specific laboratory scoping tests showed poor sequestration performance not suitable for additional consideration.(c)	N/A	N/A	N/A	Low Maturity: Laboratory testing only. No treatability test is needed. Existing laboratory data are sufficient to demonstrate poor technical practicability.
In Situ Sequestration by Apatite (iodate)	Hanford-specific laboratory scoping tests showed poor sequestration performance not suitable for additional consideration.(c)	N/A	N/A	N/A	Low Maturity: Laboratory testing only. No treatability test is needed. Existing laboratory data are sufficient to demonstrate poor technical practicability.
In Situ Sequestration by Organic Carbon	Hanford-specific laboratory scoping tests showed poor sequestration performance not suitable for additional consideration.(c)	N/A	N/A	N/A	Low Maturity: Laboratory testing only. No treatability test is needed. Existing laboratory data are sufficient to demonstrate poor technical practicability.

Technology	Evaluation Summary	Effectiveness	Implementability	Relative Cost	Maturity and Conclusion
In Situ Sequestration by Iron Oxides (iodate)	Hanford-specific laboratory scoping tests showed good sequestration under co-precipitation conditions and good sorption, although sorption is reversible.(c)	Medium: Direct treatment is needed for co-precipitation to sequester iodate. Capacity is also consumed by I-127. Sorption is good, but reversible (not sequestration).	Low: Plume core is beneath ERDF, which precludes direct treatment. Radius of influence for treatment is small relative to the plume size.	High Cost: Relative cost would be high due to the numerous boreholes needed for direct treatment.	Low Maturity: Laboratory testing only. No treatability test is needed. Existing technology information, laboratory data, and site setting information are sufficient to evaluate EIC and demonstrate poor technical practicability.
In Situ Sequestration by Bioaccumulation	Scientific literature describes the potential for accumulation of iodine in biomass.(c)	Low: Reversible sequestration with cell death and lysis.	Low: Plume core is beneath ERDF, which precludes direct treatment. Radius of influence for treatment is small relative to the plume size.	High Cost: Relative cost would be high due to the numerous boreholes needed for direct treatment.	Low Maturity: Laboratory testing only. No treatability test is needed. Existing technology information, laboratory data, and site setting information are sufficient to evaluate EIC and demonstrate poor technical practicability.
In Situ Microbial Facilitated Volatilization	Scientific literature and Hanford-specific laboratory testing show potential for microbial reactions to produce volatile iodine species.(a),(c)	Low: Large, thick plume causes a fate and transport barrier for removal of volatile species. During transport, volatile species can be converted back to non-volatile species.	Low: Plume core is beneath ERDF. which precludes direct treatment and installation of gas-extraction network. Radius of influence for treatment is small relative to the plume size.	High Cost: Relative cost would be high due to the numerous boreholes needed for direct treatment.	Low Maturity: Laboratory testing only. No treatability test is needed. Existing technology information, laboratory data, and site setting information are sufficient to evaluate EIC and demonstrate poor technical practicability.
In Situ Mobilization for Enhance Pump-and-Treat	Hanford-specific laboratory testing showed that the release of iodate occurs but is non-specific and releases	Low: Secondary affects from release of Fe, As, Mn, and sulfate affect water quality and can	Low: Plume core is beneath ERDF, which precludes direct treatment for releasing iodate. Radius of	High Cost: Relative cost would be high due to the numerous boreholes needed for direct mobilization treatment.	Low Maturity: Laboratory testing only. No treatability test is needed. Existing technology information,

Technology	Evaluation Summary	Effectiveness	Implementability	Relative Cost	Maturity and Conclusion
	other problematic constituents (e.g., Fe, As, Mn) and sulfate is produced from amendment degradation.(c)	hinder aboveground treatment processes.	influence for mobilization treatment is small relative to the plume size. Secondary effects are complicated and would need to be mitigated.	Adds cost to P&T with some benefits but several negative consequences.	laboratory data, and site setting information are sufficient to evaluate EIC and demonstrate poor technical practicability.
Ex Situ Microbial Enhanced Ion Exchange	Use of microbial reduction of iodate to iodide to enhance ion exchange is problematic based on Hanford Site experience with biological treatment in a P&T system.(d) This approach is unnecessary because resins for treatment of iodate directly are available.(c)	N/A	N/A	N/A	Low Maturity: Laboratory testing only. Not considered because direct iodate treatment is available.
Ex Situ Adsorption/Ion Exchange	Hanford-specific laboratory testing showed that two commercial resins can remove iodate from groundwater at relevant concentration reduction factors, but have a marginal resin capacity.(c)	Low: Laboratory results indicate a resin system has marginal capacity in relation to the expected iodine loading.	Low: Plume core is beneath ERDF, which precludes efficient contaminant extraction well network for vertical wells. Horizontal extraction wells are inefficient due to the plume thickness.	High Cost: Inefficient well network would cause a long extraction timeframe at increased cost compared to other Hanford P&T systems.	Medium Maturity: Engineering scale-up needed. No treatability test is needed. Existing technology information, laboratory data, and site setting information are sufficient to evaluate EIC and demonstrate poor technical practicability.

(a) Truex et al 2017; Qafoku et al. 2018; Neeway et al. 2019
(b) Rockhold et al. 2019
(c) Szecsody et al. 2019
(d) DOE 2018b
EIC – Effectiveness, Implementability, and Cost

6.0 Summary and Conclusions

The 200-UP-1 OU includes a large I-129 plume. A requirement in the Record of Decision (ROD) for interim remedial action at the 200-UP-1 OU is to "evaluate potential treatment options for I-129 as part of the selected remedy through further technology evaluation. If one or more viable technologies are identified, treatability tests will be conducted for those technologies." The requirements also state: "In the event a viable treatment technology is not available, the use of a technical impracticable waiver under 40 CFR 300.430(f)(l)(ii)(c) may need to be considered as part of the final remedy." This report evaluates potential remediation technology options for the 200-UP-1 OU with respect to the need for conducting treatability tests and to document the technology status in support of future operable unit decisions for addressing the I-129 plume.

The evaluation needed to consider the site-specific conditions that affect technology effectiveness, implementability, and relative cost. Key site factors included iodine chemical speciation and isotopic distribution in the subsurface, plume size, iodine transport behavior, depth to groundwater, and infrastructure limitations. These factors, along with the technology information collected from review of technology sources and recent site-specific laboratory testing, were considered in the technology evaluation. The evaluation examined technologies in terms of effectiveness, implementability, and relative cost similarly to how the screening evaluation is conducted for a CERCLA feasibility study. This evaluation also accounted for the maturity of the technologies with respect to the ability for viable consideration in a feasibility study.

The technology evaluation, which builds on and uses the results of the technology evaluation of Strickland et al. (2017a) and site-specific laboratory scoping tests for selected technologies (Szecsody et al. 2019), did not identify any technologies that need treatability testing to support remedy decisions. The practicability of all candidate technologies is low, driven by site and contaminant properties that hinder effectiveness and/or implementability of the technologies. The compiled information in this report is intended to be a resource for 200-UP-1 OU decisions, including the technical assessment of whether groundwater restoration is attainable from an engineering perspective, as would be required for consideration of a TI waiver.

7.0 Quality Assurance

This work was performed in accordance with the Pacific Northwest National Laboratory (PNNL) Nuclear Quality Assurance Program (NQAP). The NQAP complies with the United States Department of Energy Order 414.1D, *Quality Assurance,* and 10 CFR 830 Subpart A, *Quality Assurance Requirements*. The NQAP uses NQA-1-2012, *Quality Assurance Requirements for Nuclear Facility Application* as its consensus standard and NQA-1-2012 Subpart 4.2.1 as the basis for its graded approach to quality.

QA reviews were conducted for this work in accordance with the QA program. There were no reportable QA issues with the information included in this report. The QA controls applied to the references cited are described in the cited PNNL reports.

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