Technology Assessment for Radioiodine at the Hanford Site – Paper 20240

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ABSTRACT

An evaluation was conducted to assess the practicability of technologies of sufficient promise and maturity to warrant treatability tests for remediating radioiodine (I-129) in groundwater at the 200-UP-1 operable unit at the U.S. Department of Energy Hanford Site. Technologies were evaluated based on recent updated knowledge of subsurface iodine transport behavior at Hanford, and the effectiveness, implementability, and cost of potential treatment technologies for I-129. Because more than two-thirds of the iodine at Hanford is estimated to be present in its oxidized form as iodate, treatment technologies were evaluated with respect to addressing iodate. In situ treatments were identified as having limited implementability because the plume core is located directly beneath the Environmental Restoration Disposal Facility disposal cells, a landfill used for the disposal of low-level radioactive, hazardous, and mixed wastes generated from Hanford cleanup activities. Further limitations in treatment technologies for radioiodine were due to the isotopic distribution of subsurface iodine. Data indicate that stable iodine (I-127) concentrations are up to three orders of magnitude greater than I-129, limiting the effectiveness of potential technologies because they are not isotope-specific. Ex situ approaches also lacked the effectiveness and maturity required to treat I-129. Hence, the technology evaluation did not identify any technologies that needed treatability testing, driven by site and contaminant properties that hinder effectiveness and/or implementability of the technologies.

INTRODUCTION

Radioiodine (I-129) in groundwater at the U.S. Department of Energy Hanford Site in southeastern Washington State is the result of planned and unplanned releases of process liquid wastes and wastewater to the soil via discharges to the subsurface. Radioiodine is an environmental concern due to its long half-life of nearly 16 million years, toxicity, and mobility in the environment. Health risks occur when I-129 concentrates in the thyroid gland, thereby increasing the risk of thyroid cancer. The U.S. Environmental Protection Agency has set the maximum contaminant level of I-129 in groundwater at 1 pCi/L, the lowest of all radionuclides on the federal register. These factors make I-129 a significant remediation issue at the Hanford Site.

Groundwater in the 200-UP-1 operable unit (OU) contains a large, dilute I-129 plume associated with past nuclear weapons production activities at Hanford. Because the feasibility study could not identify a cleanup technology for I-129 treatment, the 2012 Record of Decision (ROD) for the 200-UP-1 OU Interim Remedial Action specified hydraulic containment of the I-129 plume, update of the conceptual model for I-129, and further evaluation of potentially applicable I-129 treatment technologies. The 200-UP-1 OU ROD further stated that in the event a viable treatment technology is not available, the use of a Technical Impracticability (TI) waiver may need to be considered as part of the final remedy [1].

To meet ROD requirements, a thorough review of potential I-129 remediation options was carried out to identify information needs for some of the more promising technologies that could be addressed in laboratory scoping studies [2, 3]. These laboratory scoping studies provided laboratory data for these technologies to augment the available literature data.

In addition to remediation technology efforts, refinements to the conceptual site model [4-6] and information needed as part of a TI waiver have also been documented [7]. These documents provide site context to be considered in the technology evaluation process, which has been summarized in [8]. The information on this paper is based on information published in all of these documents, focusing on the summary provided in [8].

200-UP-1 IODINE PLUME

The 200-UP-1 groundwater OU consists of the groundwater beneath the southern portion of the 200 West Area within the Hanford Central Plateau, as shown in Fig. 1. Plume maps for 1993 and 2017 (Fig. 2) 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 (~3 km^2), estimated to be tens of meters thick, and its core is located beneath the Environmental Restoration Disposal Facility (ERDF). 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.

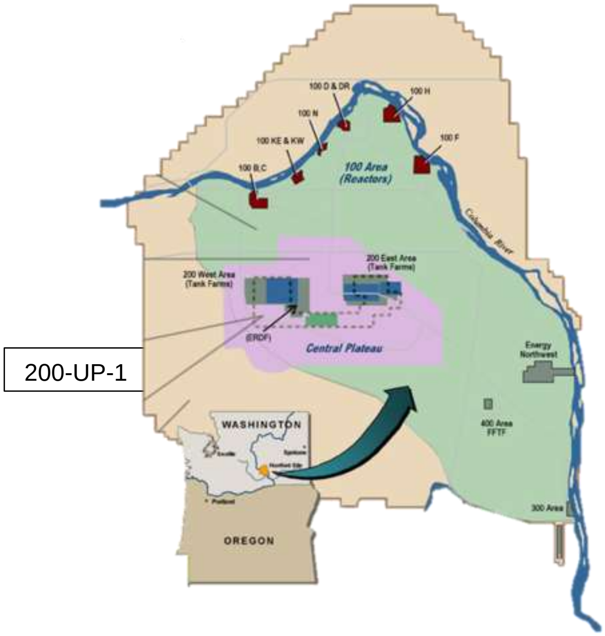

Fig. 1. Location of the 200-UP-1 operable unit at Hanford.

Both I-129 and I-129 (stable, non-radioactive iodine) are present in groundwater with a^- ratio of up to 1000:1 of I-127 to I-129 [9], 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 [4]. Therefore, the large volumes of nitric acid used during operations likely contribute to its high concentrations in groundwater. Although co-contaminants are not significant interferences for remedial approaches, the presence of high I-129 concentrations compared to I-129 concentrations must be considered because they exhibit the same geochemical behavior.

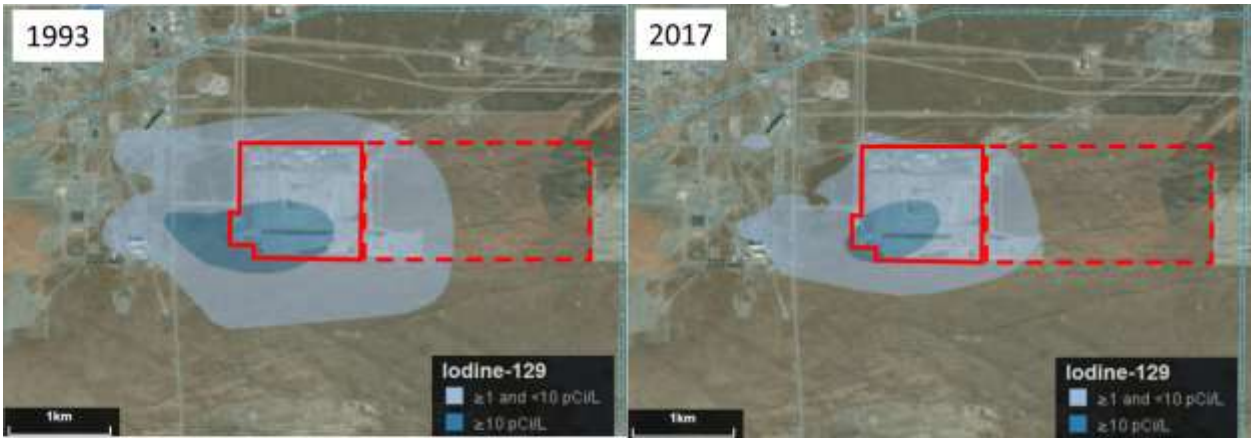

Fig. 2. Plume maps for I-129 for 1993 and 2017 from [11] as displayed by the PHOENIX web application plume animator (https://phoenix.pnnl.gov/apps/plumes/index.html). Solid red line show the current ERDF boundary. The dotted red line shows the eastward future expansion area of ERDF.

IODINE CONCEPTUAL MODEL

Biogeochemical processes that influence iodine subsurface behavior are described in [5] and are represented in Fig. 3 with three main blocks of grouped processes: GAS, AQUEOUS, and SOLID. This process model is based on iodine work in [4] and [10]. The AQUEOUS block shows the predominant species, which have been measured at Hanford as iodide (~5%), iodate (~70%), and organic iodine complexes (~25%) [4-6]. Although organic iodine complexes exist in Hanford groundwater, many of these complexes are soluble and easily transported in groundwater [4].

The SOLID block describes co-precipitation and adsorption processes that naturally attenuate I-129, with rates and extent of these reactions 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. Iodate, the predominant species in Hanford groundwater, is less mobile because it is co-precipitated with calcite and iron oxides and adsorbs to iron oxides in sediments.

TECHNOLOGY INFORMATION REVIEW

A detailed literature review of potential I-129 remediation technologies was conducted as a first step in identifying the most promising approaches for laboratory testing. The information obtained from screening was then be used to support the need for further testing or to provide technology information for consideration of a TI waiver [7]. The technology maturity and applicability were defined using criteria that ranged from a conceptual remediation technology to a remediation technology that had already been demonstrated for I-129 [2]. A total of 21 in situ technologies and 17 ex situ technologies were initially identified. Each technology was reviewed by an expert panel for its potential application to Hanford, narrowing the list of potential technologies to 11 as shown in TABLE I.

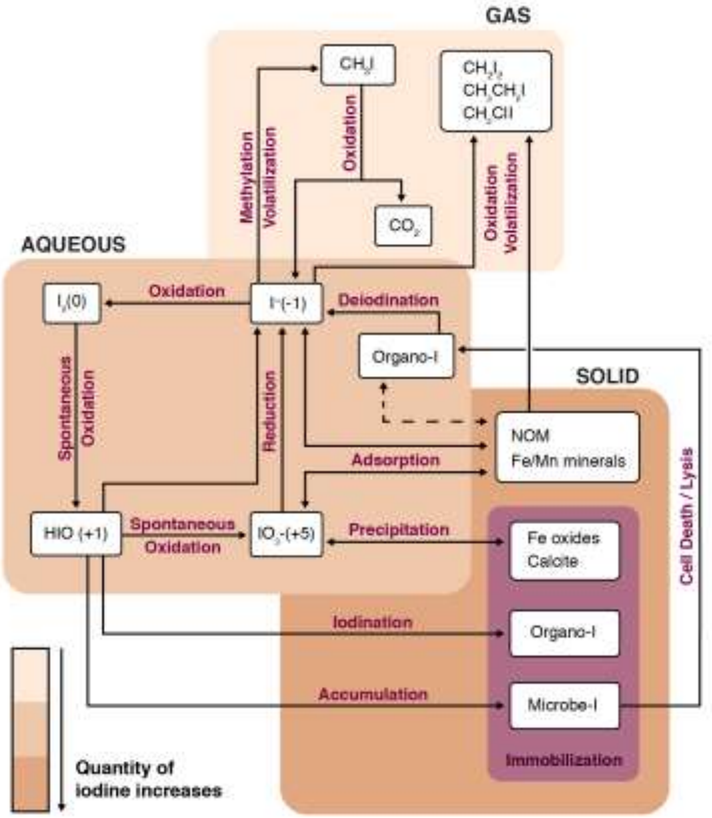

Fig. 3. Hanford site biogeochemical process for iodine (adapted from [5,6]).

LABORATORY SCOPING TESTS

Laboratory testing focused primarily on identifying the effectiveness of potential remedies listed in TABLE I. However, four technologies did not require laboratory evaluation:

1. **Monitored natural attenuation (MNA)** did not require additional laboratory testing because it was evaluated as part of the iodine conceptual site model at Hanford [4-6].
2. **In situ bioaccumulation** of iodine was not tested in the laboratory because literature information was sufficient and additional information was not needed on uptake and release [5].
3. **Microbial facilitated volatilization** did not require additional laboratory testing because it was evaluated as part of the iodine conceptual site model at Hanford [4-6].
4. **Microbial enhanced ex situ ion exchange** was not tested because operational difficulties of biological treatment that resulted in biofouling at the injection wells [11].

Testing of the remaining seven technologies was organized into three 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 pump-and-treat (P&T) operations.

TABLE I. Promising remediation technologies (from [2]).

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 P&T	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.

IN SITU SEQUESTRATION

Four in situ sequestration technologies were included in the initial laboratory scoping tests:

1. **In Situ Sequestration by Calcite (iodate).** This technology was investigated using three methods [3]. 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 approaches 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 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.
2. **In Situ Sequestration by Apatite (iodate)**. 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 [3]. 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.
3. **In Situ Sequestration by Iron Oxides (iodate and iodide).** Laboratory results indicated that iodate and iodide were effectively removed from Hanford groundwater by iron oxides, especially hydrous ferric oxides (HFO), either through sorption or co-precipitation processes [3].

The high sorption capacity at neutral pH conditions and potential to precipitate HFO in situ were positive factors for this approach. However, co-precipitation requires direct contact of the precipitating solution and the iodine. Sorption for iodide is relatively strong, but reversible. These factors relate to a relatively low effectiveness for this technology and relate to low implementability and was therefore not recommended.

4. **In Situ Sequestration by Organic Carbon.** 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 [3]. 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 distribution coefficient value was 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.

IN SITU MOBILIZATION FOR ENHANCED P&T

To accelerate the removal of I-129 in the subsurface, where sorption limits extraction efficiency, one candidate technology was tested for enhancing P&T operations:

1. **Dithionite for Mobilizing Subsurface Iodine**. Laboratory results indicated that dithionite-treated sediments enabled up to four times more and up to three orders of magnitude faster iodine leaching from the sediment relative to leaching untreated sediment [3]. The accelerated mobilization of iodine indicated that dithionite was a candidate technology for iodine treatment, provided that a candidate technology for above ground treatment could be identified. However, concentrations of dithionite required for releasing iodate also released other unwanted constituents from the sediments and increased the sulfate concentration in the groundwater. These secondary effects are significant, limit the ability to apply this approach, and was therefore not recommended.

EX SITU TREATMENTS

A wide range of materials for above ground treatment were also tested for their capacity to remove iodate from groundwater [3]. Initial batch screening of ferrihydrite, bismuth oxy(hydroxide), and bismuth-cobalt-aluminum demonstrated a removal efficiency sufficient to reduce concentrations of I-129 from 30 pCi/L to 1 pCi/L, even when concentrations of stable iodine (I-127) were 1000 times higher than I-129. Column tests were then executed, monitoring for breakthrough of iodate where a threshold of 3% of the influent concentration met this same performance metric with the following outcomes:

1. **Resin-bead forms of** polyacrylonitrile **(PAN)-bismuth oxyhydroxide, PAN-bismuth subnitrate, and PAN-ferrihydrite**. Batch testing with the PAN encapsulation material (commercially available from Global Phosphate Solutions) demonstrated strong iodate retention, with distribution coefficients of 475,000, 199,000, and 2790 mL/g, respectively, for each material. However, all PAN materials performed poorly in column tests with Hanford groundwater; therefore, further consideration of this technology was not recommended.
2. **CHM-20 (cerium-based) and ASM-10-HP (ferrihydrite-based) resins**. These commercial ResinTech resins were initially manufactured to treat arsenate. No engineering adjustments were made to the resins for the batch and column testing for iodine capture [3]. Batch testing demonstrated reasonable iodate retention, with distribution coefficients of 10,200 and 5800 mL/g, respectively.

However, column testing showed the resins had a low capacity for iodate retention and negative interactions with groundwater constituents. Hence, the performance of these resins in a flow-through system was considered marginal and were not recommended.

REMEDIATION TECHNOLOGY ASSESSMENT

The data collected in the literature review were used to identify technologies for testing in the laboratory, as described above under in situ, enhanced P&T, and ex situ categories. Scale-up tests were performed on three technologies: 1) in situ sequestration by iron oxides, 2) dithionite for mobilizing subsurface iodine, and 3) ex situ resins (both PAN-based and commercial resins). However, logistical factors, and site-specific I-129 chemistry factors were used to evaluate the technology applicability for the promising candidates in TABLE I with respect to the effectiveness, implementability, relative cost, and maturity similar to the screening approach for a Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) feasibility study. Key factors for consideration included the following:

1. **Depth to plume and its areal extent and thickness**. The I-129 plume in the aquifer is located about 75 m below ground surface. The areal extent of the plume above 1 pCi/L is about 3.4 km^2 with a thickness of ~40 m. For all technology categories, implementability and cost are affected by the plume size. For example, 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 water extracted amendments needed for injection.
2. **Spatial relationship of ERDF and plume core**. 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 installing wells outside of the ERDF footprint.
3. **I-127 and I-129 concentrations are present at a ratio of 1000:1**. Because biogeochemical processes are the same for either isotope, treatment processes need to address the total iodine concentration yet have a high effectiveness to meet concentration standards that are based only on I-129 concentrations.
4. **Iodine chemical speciation**. Based on measurements of I-127, groundwater data show that most of the iodine is present as the iodate species [4], which is assumed to have the same chemical speciation as I-129. Hence, technologies need to be able to treat iodate.

The results of the overall assessment are summarized in TABLE II, based on the technology effectiveness, implementability, relative cost, and maturity. Technologies screened out after laboratory scoping tests were not evaluated for effectiveness, implementability, and cost (EIC) in TABLE II. 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. No technologies were identified for I-129 treatment at 200-UP-1.

TABLE II. I-129 remediation technology assessment for the 200-UP-1 operable unit (from [8]).

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 [4-6]. The plume will attenuate over a long time period (>100 years) while it migrates downgradient [7].	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 [4].	Screened out prior to EIC evaluation			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 [4].	Screened out prior to EIC evaluation			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 [4].	Screened out prior to EIC evaluation			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 [4].	Medium: Direct treatment is needed for co-precipitation to sequester iodate. Capacity is also consumed by I-129. 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 [4].	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 shows potential for microbial reactions to produce volatile iodine species [4-7].	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.

Technology	Evaluation Summary	Effectiveness	Implementability	Relative Cost	Maturity and Conclusion
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 other problematic constituents (e.g., Fe, As, Mn) and sulfate is produced from amendment degradation [4].	Low: Secondary affects from release of Fe, As, Mn, and sulfate affect water quality and can hinder aboveground treatment processes.	Low: Plume core is beneath ERDF, which precludes direct treatment for releasing iodate. Radius of influence for mobilization treatment is small relative to the plume size. Secondary effects are complicated and would need to be mitigated.	High Cost: Relative cost would be high due to the numerous boreholes needed for direct mobilization treatment. Adds cost to P&T with some benefits but several negative consequences.	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.
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 [11]. This approach is unnecessary because resins for treatment of iodate directly are available [10].	Screened out prior to EIC evaluation			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 [4].	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.
EIC – effectiveness, implementability, and cost					

CONCLUSIONS

The technology evaluation for I-129 at the 200-UP-1 OU at Hanford needed to consider key site factors such as iodine chemical speciation and isotopic distribution in the subsurface, plume size, iodine transport behavior, depth to groundwater, and infrastructure limitations. The laboratory screening provided additional information for conducting the technology evaluation with respect to effectiveness, implementability, and relative cost similar to the screening approach for a CERCLA feasibility study. Technology maturity was also considered in this evaluation.

The technology evaluation did not identify any viable technologies for treatability testing. The practicability of all candidate technologies was low, driven by site and contaminant properties that hinder effectiveness and/or implementability of the technologies. This information can be used to support consideration of a TI waiver.

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