Initial Sitewide Groundwater remediation Strategy of the Hanford Site, WA: Its Application, Lessons Learned and Future Path forward

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ABSTRACT

In 1989, the Washington State Department of Ecology (Ecology), the U.S. Environmental Protection Agency (EPA), and the U.S. Department of Energy (DOE) formed an agreement to clean up the Hanford Site, located in the state of Washington. By 1995, the three parties developed an initial comprehensive site wide groundwater remediation strategy with a vision to address contaminated plumes of hazardous and radioactive waste. The Hanford Site has more than 170 square miles of contaminated groundwater. Almost half exceeds the state and federal drinking water standards. The plumes are often commingled. The remediation is challenged by limited technologies, poor understanding of conceptual models, and subsurface contaminant behavior. This paper briefly describes the basic principles of the initial strategy, its application, the results of the decade-long operation, and the future path forward.

The initial strategy was based on a qualitative assessment to reduce immediate risk to human health and the environment; to support commonly held values of stakeholders, including tribal nations and the public; and to deploy available remediation technologies. Two different approaches were used for two distinct geographic, the river shore reactor areas and the central plateau few miles away. The strategy was to cleanup the major groundwater plumes in the reactor areas next to the Columbia River where chromium, strontium-90, and uranium already entering the river and to contain the plumes of chlorinated solvents and radionuclides in the central plateau. The strategy acknowledges the lack of cost-effective technologies to address the contaminants, and asked DOE to develop, test, and deploy cost-effective alternative technologies wherever applicable.

After more than a decade, the results are mixed. While the pump and treat provided a meaningful approach to address certain contaminants, it was too small in scale. Efforts to scale up these operations enhance characterization, and to deployment innovative technologies are progressing; albeit slowly due to budget constraints. A number of innovative technologies were identified to address source control and groundwater remediation across the Hanford Site.

In the 10 years since the initial strategy was developed, additional severe groundwater and vadose zone contaminations were discovered under the waste storage tanks on the central plateau and river corridor areas. These problems required changes to the strategy. Changes include complete integration of vadose zone and groundwater characterization and remediation activities and immediate needs for technologies to address the deep vadose zone source areas, as well as thick aquifer contamination – especially for chlorinated solvents and technetium-99.

The successes of the initial strategy show that even a strategy based on incomplete information can make progress on difficult issues. The regulatory agencies identified these issues early and

provided the needed direction to DOE to move forward with the overall mission of clean up. The cleanup of the Hanford site is a big challenge, not only for DOE, but also for the regulators, to ensure the tri-party agencies achieve the desired goals.

INTRODUCTION

The Hanford Site encompasses approximately 560 square miles northwest of the city of Richland along the Columbia River in southwest Washington State. The site is divided into several distinct areas: the 100 Areas along the Columbia River, the 300 Area along the Columbia River just north of Richland, and the 200 area comprising east and west area covering the central portion of the site and popularly known as the "central plateau". Nine production reactors were located along the river corridor. Until 1980s, the site was dedicated primarily to the production of plutonium for national defense.

In the 1990s, the site's mission changed to cleaning up site. DOE, EPA and Ecology signed a comprehensive cleanup and compliance agreement on May 15, 1989. The Hanford Federal Facility Agreement and Consent Order, or Tri-Party Agreement [1], is an agreement for achieving compliance with the Comprehensive Environmental Response Compensation and Liability Act (CERCLA) remedial action provisions and with the Resource Conservation and Recovery Act (RCRA) treatment, storage, and disposal unit regulations and corrective action provisions. The Tri-Parties collaboratively developed the Hanford groundwater remediation strategy as a means for multiple regulatory authorities and government agencies to move forward with active groundwater remediation on a sitewide basis.

During the production of plutonium, huge amount of solid and liquid wastes were generated. In total, site records show that 440 billion gallons of contaminated liquids were intentionally discharged to the soil and groundwater. About 1 million gallons of radioactive wastes from storage tanks have been unintentionally released, albeit in more concentrated form. As a result, over 170 square miles of groundwater beneath the Hanford Site are contaminated by hazardous and radioactive waste. Almost half exceeds over federal and state drinking water standards (figure 1). In the central plateau area groundwater contaminant plumes include uranium, technetium-99, iodine-129, tritium, chlorinated hydrocarbons (e.g. carbon tetrachloride, chloroform and trichloroethylene) and nitrate in the 200 West Area. Plutonium, cesium-137, strontium-90, technetium-99, iodine-129 and nitrate plumes have been found in the 200 East Area. In the river corridor, a major strontium-90 plume is present in the N-Area and hexavalent chromium plumes are present at 100-K, D, H and F areas. A major groundwater plume containing mainly uranium is also present in the 300 Area. The chromium, strontium-90, and uranium plumes currently discharge to the river. The most extensive contaminant plumes in groundwater are tritium, iodine-129, and nitrate, covering tens of square miles. They have multiple contamination sources. These contaminants are highly mobile. They are known as "sitewide" plumes. The largest portions of these plumes are migrating from the 200 East to the Columbia River. The nitrate and the tritium plumes both reached the river several decades ago. Several contaminant plumes overlap because of either merging plumes from different sources or because they were released as co-contaminants. The general direction of groundwater flow is from the higher elevation natural recharge areas southwest and west of the Hanford Site to discharge areas primarily along the Columbia River. However, the groundwater levels along the river corridor are heavily influenced by river stages causing changes daily, weekly, and seasonally. River stage may change up to about 8 feet within a few hours. As the river stage rises, river water flows into the aquifer /bank storage. When the river stage falls, water flows out of the aquifer via river bank springs which is composed of a mixture of groundwater and bank storage river water, changing the concentrations of near shore groundwater, river bank spring water, and Columbia River water. The river stage fluctuation has tremendous impact on any groundwater remediation designed for this mixing zone along the river.

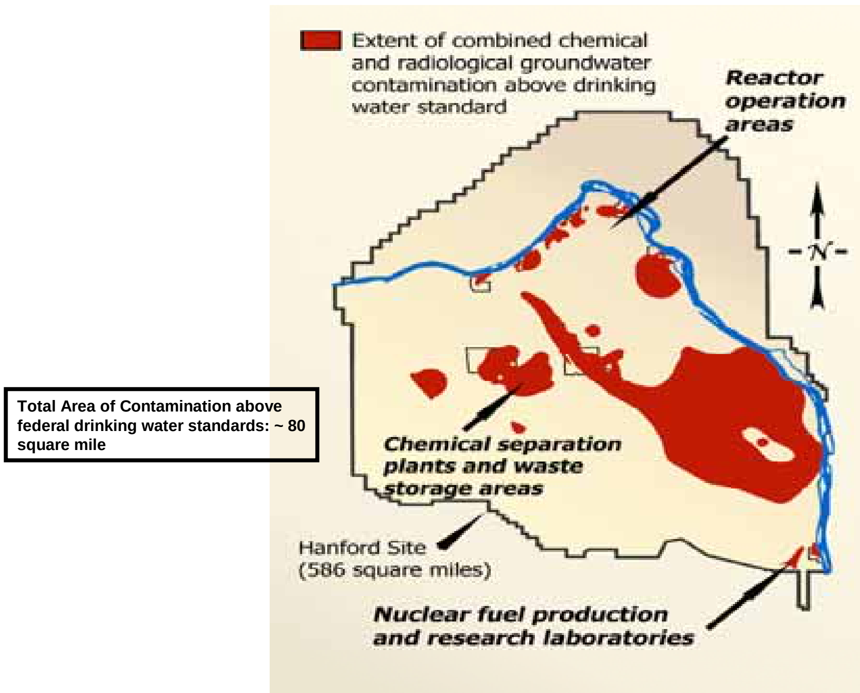

Fig. 1. Area showing the groundwater contamination above federal drinking water standards

INITIAL STRATEGY

The purpose of the initial strategy is to develop core principles to manage and accelerate groundwater remediation across the Hanford Site as an interim step to final remediation [2]. It provides a means of addressing issues of sitewide significance, and a broader perspective for planning remediation at individual operable unit level.

The strategy was developed with recognition that stakeholder, public and Tribal Nations values shape clean up objectives. The following major principles were developed based on the commonly held values and the recommendations from the future site uses committee [3, 4].

1. Protect human health, worker safety, and the environment
2. Protect the Columbia River and the near shore environment from degradation caused by inflow of contaminated groundwater.

3. Cease untreated discharges to the soil and groundwater. Treated discharges should not affect the existing contamination plume
4. Control the migration of plumes that threaten or continue to further degrade groundwater quality beyond the boundaries of the Central Plateau area of the Hanford Site.
5. Use available technology and start remediation
6. Develop new technologies to clean up contaminants less amenable to remediation with available technologies
7. Monitor groundwater throughout the Hanford Site well coordinated monitoring network system and continue monitoring all known contaminants that might undergo natural attenuation below drinking water standard.
8. Cleanup groundwater on a geographic basis, to level necessary to enable the future land use option to occur as per the Future Site Use Committee’s recommendation.

The Future Site Use Committee’s recommendation on clean up range from “unrestricted use” in the 100 and 1100 Areas, to “restricted use” for the 200 Area (central plateau) and “industrial use” in the 300 Area. A “buffer zone” concept was used for the Central Plateau which surrounds an exclusive area and is treated like an exclusive area.

On the basis of these values and guiding principles, the initial effort of sitewide remediation focused on:

- Maintaining a bias toward accelerated groundwater remediation using proven technology to control plume expansion, reduce contaminant mass, and better characterize aquifer response to remedial actions.
- Prioritizing work that will protect the Columbia River.
- Identifying control sources of contaminants in the vadose zone that impede groundwater remediation.

The succeeding phases of the remedial actions are oriented toward implementation of a final Record of Decision (ROD). This in turn will satisfy broader cleanup activities such as:

a. Comply with Achieve applicable relevant and appropriate requirements of CERCLA for the value of current and potential future beneficial uses for groundwater.
b. Develop alternative and containment remediation strategies if currently available groundwater remediation technologies prove in adequate or impracticable.
c. Restore groundwater next to Columbia River for unrestricted use.
d. Prevent further degradation of groundwater beyond the boundaries of the Central Plateau, and ultimately restore unrestricted beneficial use beyond the boundary

APPLICATION OF THE REMEDIATION STRATEGY

Initially, geographic and plume-specific approach for groundwater remediation was designed. The strategy focused on two major contaminants in 100 Area and four major contaminants in the central plateau. The strontium-90 plume located in the N-Area and hexavalent chromium in the D, H, and K Areas were selected in 100 Area. Groundwater contaminated with strontium-90 is discharging directly to the Columbia River and in areas accessible to public at concentrations many times higher than drinking water standards. Hexavalent chromium is also discharging directly to the river and is toxic to aquatic species [5]. In the 200 West Area of the central plateau region, carbon tetrachloride, chloroform, trichloroethylene, uranium and technetium-99 plumes were selected for active remediation. These plumes are expanding and have the potential to migrate to Columbia River at level several times drinking water standards. A soil vapor extraction of carbon tetrachloride was recommended as one of the initial source control measure for groundwater remediation in the 200 West Area. As with the other contaminant plumes, the initial remediation area was limited to the highly concentrated portion of the plume. This typically covers a very small portion of the entire plume. It was assumed that most of the mass is confined in this core area which eventually be reduced by the active remediation, thereby, affecting the plume migration significantly.

Since 1995, DOE has operated six pump and treat systems (figure 2) to remove carbon tetrachloride, chloroform, trichloroethylene, uranium, technetium-99, strontium, and chromium. DOE also used chemical barrier technology near the Columbia River to stop hexavalent chromium going to the river in the 100-D Area. Out of the four pump and treat operation in the 100 Area, three pump and treat are currently treating chromium while the other one is on stand by mode until a treatability study using apatite sequestration technology to treat strontium-90 in 100 N-Area groundwater is completed.

The pump and treat operation in the 200 East Area to treat mainly technetium, strontium, plutonium and cobalt-60 was abandoned after about eighteen month of operation. The system was found to be cost prohibitive since wells could not produce enough groundwater. Tri-Parties used standard CERCLA regulatory processes for treatability testing, followed by interim RODs [6]. The remedial action objectives varied from meeting 10 times of drinking water standard in the 200 West Area for uranium and technetium to 10 ppb ambient aquatic water quality criteria for chromium in 100 Area. In 100-N Area, the main goal was to stop strontium-90 going to the river although it was realized that the pump and treat is not the cost effective groundwater remediation technology. The strategy emphasized the lack of cost-effective technologies to address the contaminants of uranium, strontium-90, technetium and chromium and asked DOE to develop, test, and deploy cost-effective alternative technologies during the implementation of this initial strategy phase.

LESSONS LEARNED

More than a decade has passed since the implementation of the above strategy. Two 5-Year ROD reviews have been conducted to evaluate the effectiveness of this strategy [7]. Conventional pump and treat has provided enough information and seemed to be the appropriate remediation tool to move forward at all locations except for the 100-N Area. However, while pump and treat successfully addressed target contaminants, it was too small in scale. Large scale above ground treatment capacities were needed to treat groundwater contaminated with chlorinated solvents, chromium and radionuclides.

The rate of strontium-90 in N-Area was significantly slower than expected and strontium-90 release from groundwater to the river remained the same. Studies have shown that the strontium-9 moves extremely slowly and the discharge to the river came from strontium-90 in the riparian and the adjoining area. The current pump and treat cannot work in these areas due to the fluctuation of the Columbia River and its impacts on the bank storage. The remediation technology must address the riparian zone strontium-90 to protect the Columbia River. This will require innovative technologies.

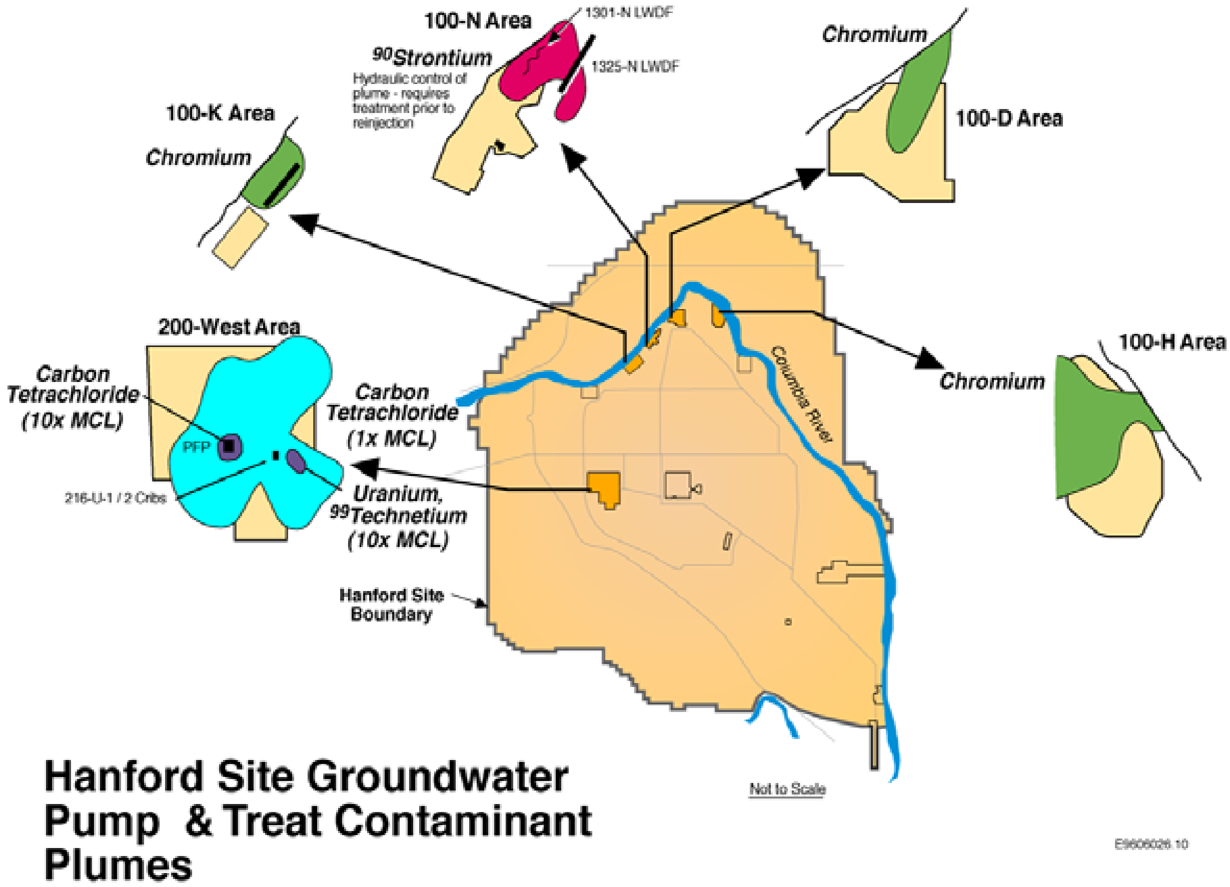

Fig. 2. Location of the pump and treat operations at the Hnaford Site

We also have learned that to combat the increasing concentration of chromium in the 100-D Area, we must remove source of contamination there. However, we have yet to identify and delineate it. The chemical barrier popularly known as "ISRM (In-Situ Redox Manipulation)" did show great potential in reducing hexavalent chromium to trivalent. Recently its performance was found to be unsatisfactory, requiring more innovative studies to improve its performance.

Since the development of initial strategy in 1994, we have had a number of surprises. The initial strategy was based on a general understanding that contaminants were confined within the top 30-35 feet of the aquifer. While the lateral extent of contamination is delineated by the current groundwater monitoring wells installed at the top 35 feet of the unconfined aquifer, we still do not know the vertical extent of a number of plumes. It seems a number of contaminants were found at greater depth than originally assumed. This would significantly impact current approach of remediation. Some of the key discoveries and challenges identified since 1994 were:

- Leaking tanks have impacted the groundwater. Some of the immobile contaminants such as cesium, cobalt, and uranium have migrated deeper than expected. There appears to be a big inventory of radionuclides in the tank farm vadose zone and its impact on groundwater is not known.
- The groundwater contamination under certain tank farms is very high and has reached a great depth. For example the concentration Tc-99 in the T tank farm in the 200 West Area was found to be 182 000 pCi/L, the highest ever found at the Hanford Site. The drinking water standard for Tc-99 is 900 pCi/L. This contamination has reached a depth of about 170 feet below the water table [8].
- There is a large inventory of technetium-99 in the 200 East Area B/C Crib deep vadose zone which eventually will impact the groundwater. The contamination may eventually reach the Columbia River if nothing is done to treat this deep vadose zone contamination.
- Most carbon tetrachloride in the 200 West Area is in groundwater down to a depth of about 150 feet below the water table. Carbon tetrachloride is present as the dense non aqueous phase liquid in the vadose zone. The plume is spreading faster than expected.
- The geochemical conceptual models and contaminant fate and transport behavior of uranium were found to vary significantly across the site. More site specific characterization, investigation and field studies are required to understand its behavior to implement appropriate remediation technologies.
- The vadose zone source of chromium in 100-D Area is yet to be delineated and remediated. The removal of this source is vital to success of groundwater remediation in this area.
- There are major uncertainties in the Hanford groundwater flow field. The conceptual model should be established with credible field data and field observation. Proper understanding of the flow field is vital to the long term remediation goal.

FUTURE PATHFORWARD:

It is clear that we must go beyond conventional remedial technologies and test and deploy innovative technologies to address contamination in the deep vadose zone and groundwater. In order to move forward, we need an integrated approach of characterization and remediation for vadose zone and groundwater. We need to look across the site covering both the tank farm and the non tank farm sites. We must know more about the nature and extent of contaminants in the vadose zone and groundwater, especially in the central plateau and 100 D Areas.

Understanding the nature and extent of contamination and fate and transport are important for making decisions about how best to protect the Columbia River. The future approach should include the following issues:

- Use innovative approach to remediate groundwater contamination of Sr-90 in the N-Area, uranium in the 300 Area and chromium contamination in 100 Area.
- Develop and deploy technologies to address deep vadose zone source areas in the central plateau and the river corridor

- Deployment of cost effective remediation approach to address thick aquifer contamination – especially for chlorinated solvents and technetium-99 in the central plateau and
- Develop a better understanding of the conceptual model for the uranium to address the tank farm and 300 Area.

Tri-Parties have identified a few technologies to address these issues and to move forward with site specific field scale demonstrations. We placed priority on protecting the Columbia River from contaminants such as Sr-90, chromium and uranium.

Two technologies were aimed at improving the performance of the ISRM barrier where chromium breakthrough has occurred. The cause of premature barrier breakdown was determined to be heterogeneities in the aquifer, whether laterally discontinuous units with high permeability and lower inherent reductive capacity (because of lower iron content) were reoxidized faster than the less transmissive layers. In this conceptual model, the barrier wells would have to be reinjected periodically to reestablish the reducing environment in these more permeable layers. Micron-sized iron will be injected to mend deteriorating portions of barrier. Also microbial substance will be injected into the subsurface to stimulate indigenous bacteria to reduce concentration of dissolve oxygen, nitrate, and hexavalent chromium in the up gradient of the ISRM barrier which will reduce the electron acceptor flux to the ISRM barrier thereby increasing the longevity of the barrier. We will also deploy an alternative treatment technology, electrocoagulation in a test system to improve and expand the existing pump and treatment system. Studies have shown that electrocoagulation holds promise for cost-effectively treating groundwater to the remedial action goal, which is <20 ppb. The focus of this work will be to evaluate the operability, robustness, and treatment efficiency to expand the current pump and treat system from 200 to 500 gallon per minute. The ultimate goal of these technologies, if proved successful, will reduce or completely eliminate chromium contamination reaching the Columbia River in the 100-D Area.

In 2004, Ecology proposed that DOE test apatite technology and phytoremediation to address the riparian zone Sr-90 in the 100-N Area. The apatite sequestration technology involved injection (through wells) of calcium-citrate-phosphate solution, which after a week will precipitate apatite, a natural calcium-phosphate mineral. Apatite will absorb the Sr-90, isolate the Sr-90 and keep it from entering the river. The other technology is phytoremediation. Regulators as well as the DOE identified phytoremediation as a potential technology both for the removal of Sr-90 from the soil of the riparian zone and as a filter for groundwater along Columbia River. Recent greenhouse and growth chamber studies have demonstrated the viability of phytoextraction to remove Sr-90 from this areas soil and groundwater.

Another technology testing is now focused on methods that would reduce the concentration of uranium in groundwater in the 300 Area. Methods include many techniques to stop uranium using chemical-based technologies. Initial screening identified phosphate technology as the best candidate for further evaluation. Treatability testing will follow to evaluate the efficacy of using phosphate injections to treat uranium contaminated groundwater in-situ. DOE is now preparing a detailed test plan. The results of the testing will help for estimating costs, defining implementation challenges, and determining the technology's ability to meet remedial objectives.

Regulators recommended a robust active remediation effort in the second 5-Year ROD review. Recommendations include the emerging plumes of technetium-99 in T tank farm area and uranium and technetium-99 plumes in the 200 Area B-BX tank farm and increase the current pump and treat systems. Regulators asked DOE to seek appropriate funding from the congress to implement a robust clean up plan reflecting the above requirements.

CONCLUSION

The 1994 groundwater remediation strategy was based on qualitative assessment of risk to the human health and the environment, stakeholders and tribal nation's values. The overarching goals and objectives associated with the strategy are also the basic principles of the future groundwater clean up. New discoveries and the lessons learned from the decade long implementation have provided valuable information on how to move forward in the future more effectively and wisely. We have learned that the better we integrate the better the results. The groundwater remediation program must become more robust not only by enhancing the current systems but also by addressing the emerging groundwater plumes and vadose zone sources under the tank farms. The innovative technology deployment must play a major role in cleaning up vadose zone and groundwater contamination. Tri-parties have already identified a number of technologies and their field scale demonstrations are now under way. To move forward in the future, is crucial.

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