Chapter

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Remediation

Because long-lived contaminants were released into the environment in the past, remediation is essential at each DOE site. Unfortunately, most of the hazardous materials, earmarked for remediation, have lifetimes of hundreds of thousands of years, or forever, in the case of some toxic chemicals, greatly complicating the cleanup process and putting future generations at risk.

Remediation can be divided into three basic types of action: removal, containment and treatment. Plans differ at each of the sites depending on the characteristics of the environmental media as well as varying levels of contamination. The presence of mixed wastes complicates the cleanup options due to the challenge of remediating both hazardous and radioactive wastes. In this chapter, we present a series of remediation options. Following this we discuss long-term stewardship at each of the 13 DOE sites.

Removal

Removal is the only way to completely restore the environment and prevent toxic chemicals and radionuclides from seeping into the groundwater and reaching humans and animals. Direct removal of existing contamination should therefore be carried out wherever and whenever possible.

Removal could be the most effective solution at substandard landfills containing nuclear and/or hazardous waste and contaminated soil. At Oak Ridge Reservation, for example, the plutonium and thorium contaminated soil is excavated and disposed of offsite where allowed. Much of the exhumed waste material has gone to the Envirocare facility in Utah, though DOE has been searching for less expensive alternatives, such as the Cotter uranium mill in Canon City, Colorado, where there is substantial community opposition.

Some Oak Ridge landfills were placed directly within the water table, guaranteeing leakage from rusting drums and cardboard boxes. Excavation and removal enables the residually contaminated soil areas left behind to be within allowable concentration limits.[1]

But there is a problem. The material must then be treated or be safely disposed of at its new location. Treatment of exhumed soil may mean incineration that poses additional problems. Placing landfill contents in aboveground storage facilities removes the contamination source, but, contrary to frequent assertions by DOE, long-term maintenance will still be required. Concrete bunkers degrade and crack with time, necessitating ongoing repairs and maintenance.

As is now evident at the Hanford site even carbon steel tanks have a specific lifetime that is shorter than the long-lived radionuclides stored inside.

Another problem: Because non-contained contaminants spread in the environment, the volume of material that is contaminated and must be removed is typically considerably greater than the waste that was originally disposed of. The previous DOE practice of dumping wastes into unlined landfills, holding ponds, or directly into the soil has created a need for enormous amounts of space for properly contained and controlled disposal sites that comply with modern regulatory standards. These sites will require long-term monitoring, with periodic surveys and tests to ensure the maintenance of contaminant levels.

Excavation of contaminated materials will require close attention to worker safety. Depending upon the nature of the wastes at each of the sites, workers will need to be properly suited, masked up where necessary, with all proper safety practices followed.

Removal and aboveground storage of subsurface contamination is still the only alternative that efficiently prevents contaminants from migrating to the groundwater, as opposed to the comparatively incomplete methods of containment and treatment (see below).

Containment

Because removal of wastes and soils can be very costly and may pose a risk to workers, DOE attempts to leave waste in place wherever possible. This containment policy is deeply flawed. According to a recent study, "The DOE's current management practices, budget assumptions and contracting strategies are a blueprint for catastrophic releases of nuclear and toxic wastes."[2]

To protect the environment and human health, the radionuclides and/or chemicals have to be held in place, or contained, for a long period of time. During this long time-frame the materials must be monitored. This period has to be long enough for the contaminants to degrade, so that if they should reach a human or the environment, their capacity for damage will have decreased to a point below allowable limits.

Strategies of containment include (but are not limited to) impermeable caps, slurry walls, groundwater extraction pumps, disposal cells and chemical immobilization in place, all of which are briefly described in the following paragraphs.

Impermeable caps

Buried wastes can be mounded to assist water runoff and capped with layers of impermeable material to prevent surface water from seeping in and dissolving the contaminants. The use of impermeable caps as a solution has three problems. One, the "impermeable" cap may not be impermeable for the requisite decades or, in some cases, centuries required. Two, caps give a false sense of security and often result in a reduced or no monitoring regime. Three, caps do not address the problem of ingress of water horizontally from underground water sources.

Over the long time periods during which these toxic materials must be sequestered from the environment, the caps (often no more than layers of clay, topped by plastic sheets and soil) will degrade. The degradation may occur from cracking, which is a particular problem in environments with freeze and thaw cycles. Caps are also weakened by penetration by plant roots, animal burrows, settling of the landfill contents as containers degrade and erosion of the landfill beneath the cap. Caps do not allow DOE to walk away, saying the contamination problem is solved; they require long-term maintenance, involving repairing and replacing the caps for decades to come.

The third problem with caps, ingress of water from underground sources, may happen when the groundwater table rises and reaches the buried wastes. Now the uncontained wastes can simply be flushed away. Many factors can cause the water table to fluctuate: increased or decreased precipitation, nearby construction that changes the configuration of the land, flooding in nearby rivers, etc.

An example of the problem with "impermeable" caps is the six waste pits at the Fernald site in Ohio. Two have a water cover and are not yet remediated. A water cover is an unremediated settling pond. In a settling pond contaminants are released to a pond, where the pollutants supposedly "settle-out" or sink down into the bottom of the pond.

At Fernald chemicals are added to precipitate out the uranium. The settled material has the consistency of toothpaste, with a covering of water on top. When the ponds are remediated, they are dried out and covered with an impermeable cap. Of the 4 other pits at Fernald, one has a synthetic cap and three others have soil covers. Unfortunately, capping these wastes does not reduce the risk to the groundwater because each of these pits is located near or directly above the aquifer.

With inadequate knowledge of the underground hydrologic regime, as well as overly optimistic assessments of the behavior of synthetic cover materials, so called "impermeable" caps have not been entirely successful at Fernald and many other sites.

Slurry walls

Slurry walls are underground barriers meant to prevent groundwater from migrating towards discharge areas such as surface waters or groundwater wells, or to prevent ingress of groundwater. In some cases slurry walls can effectively prevent migration in a specific direction, but they do not completely contain the contamination.

Like an underground dam, slurry walls are designed to hold back groundwater movement. But slurry walls generally must operate in concert with groundwater extraction pumps; or else groundwater will back up and eventually overtop or bypass the slurry wall. The volume of contaminated material therefore continuously increases over time.

In short, groundwater extraction pumps must operate indefinitely if slurry walls are to be effective.

Another type of slurry wall, a porous wall, is meant to selectively remove specific contaminants, such as strontium-90, while allowing groundwater to pass through. An attempt at an old (shutdown in 1972) reprocessing factory in West Valley, New York was not successful. The underground plume moved around the wall. Furthermore, over time, porous slurry walls may degrade, becoming so clogged that they no longer adequately filtrate contaminants.

Groundwater extraction pumps

Sump pumps can sometimes extract groundwater at a rate sufficient to stop the off-site migration of contaminants in a groundwater plume, or alter its flow, and are often used in conjunction with interceptor trenches or slurry walls. Instead of slowly moving toward the natural discharge region or a drinking water well, the groundwater flows towards the pumps. To effectively contain the contaminant, the flow toward the pump must exceed the diffusion rate of the contaminant in water.

At Pantex, pump-and-treat is one of the primary methods of remediation of the groundwater contamination. Volatile organic compounds, semi-volatile organic compounds, fuels, explosive compounds, and dissolved metals are the materials that are removed from the groundwater with pump-and-treat systems. As we have seen above, this method does not remove radionuclides and metals that adhere to soil. The pump-and-treat method can be used with ion exchange resins to selectively remove some chemicals, but, throughout the DOE complex, pump-and-treat is almost exclusively used for volatile organics that can be vaporized and captured on charcoal filters. For example, TCE is being removed at Paducah via charcoal filters and stripping.

With few exceptions, pump-and-treat puts radionuclides back into surface or ground waters, in a continual cycle. For example, at Lawrence Livermore lab, tritium is pumped from the front of an advancing plume and reinserted at the rear. As long as the process is maintained and continued, the groundwater plumes are restricted from traveling further offsite. Clearly as long as these operations continue, full decommissioning of a site is impossible. It is important to note again that it is possible to remove strontium, cesium and uranium from groundwater using ion-exchange resins. This is done in other operations, such as in situ uranium mining operations or in removal of cesium and strontium from high-level liquid waste tanks, but the DOE has chosen not to employ this technology elsewhere in the complex.

A major complication with the pump-and-treat method is that it requires a very long time to meet cleanup goals, typically 50 to 100 years. For example, because of the geology and nature of the contaminants, pump-and-treat may not reach acceptable groundwater levels in the Snake River aquifer till the year 2095.[3] At this juncture, DOE's remediation plans have a much shorter time frame with plans at most sites to "finish" remediation and terminate active cleanup programs long before the end of this time span.

Another complication with groundwater extraction pumps: the removed contaminants must be packaged and isolated from the environment, essentially forever.

If the groundwater extraction rate is high enough, it is possible for the groundwater plume to be completely pumped out of the ground and treated, resulting in a fairly complete cleanup, providing the removal efficiency is high. However, such high extraction rates are technically difficult to maintain and are very costly. Furthermore altering the natural groundwater flow can have significant environmental impacts, such as drawing down the aquifer.

Another problem: the extracted water has to be treated before discharge, but this is not done for all contaminants. For instance, the groundwater extraction and treatment program at the Portsmouth Gaseous Diffusion Plant removes about half of the volatile organic compounds in the extracted water and none of the radionuclides, before discharging the water to the nearby Scioto River. Clearly, this example of remediation does not adequately protect the public. At LLNL, as mentioned previously, pumps remove tritium-contaminated water, only to move it to another location.

Disposal cells

Disposal cells are large underground containers that are lined to keep water out from all directions, physically encapsulating the contaminants. Should water seep in (through cracking, corrosion, defects, earthquakes, floods, etc.), the cells are designed to drain to a catch basin where contamination levels can be monitored. Disposal cells can be an effective way to contain contamination, if placed above the water table and properly maintained.

Containment of contaminants that degrade within a relatively short time frame can be very efficient and more cost-effective than removal or treatment. However, containment of long-lived elements is not practical, because the facilities (pumps, layers, basins, controls, etc.) must be maintained continuously over extremely long period of times, due to the long hazardous lives of many radionuclides and toxic chemicals.

It is reasonable to assume that at some point in time, institutional control over any site will be lost, and it cannot be assumed that the future occupant of the land will have the capability, know-how and funding to maintain the containment. Institutional control refers to a site being controlled by a governmental agency or other administrative unit. This "control" might involve periodic monitoring, occasionally repairing a fence or concrete bunker, or other forms of maintenance. Once institutional control is lost, the site would not be monitored or maintained in any way.

In short, the function and maintenance of containment facilities cannot be guaranteed for several thousands of years, and the off-site migration of contaminants with half-lives of thousands or millions of years is therefore unavoidable. The inevitable loss of institutional control and subsequent failure of containment facilities have to be taken into account in any responsible risk analysis of a remediation plan.

Immobilization

In some rare cases, it is possible to immobilize contaminants so they remain in place. It is being done successfully for chromium in groundwater at the Portsmouth Gaseous Diffusion Plant, though it not clear how long chromium will remain immobilized. Essentially, the valence state of chromium is changed with chemical reductants. (Valence state refers to the number of free electrons that the material can be combined with.) Using this technology highly toxic chromium +6 is reduced to relatively harmless chromium +3. The highly mobile CrO_4^2 becomes the highly insoluble solid $Cr(OH)_3$.

Decontamination Treatments

A concern with clean-up technologies is the lack of knowledge of the origin and extent of the contamination. With time, institutional memory has been lost. Furthermore, there often is inadequate knowledge and/or controversy regarding the hydrologic regime underneath a site. For example, is water gaining access to waste via precipitation penetrating through landfill covers or from an unknown underground source? These lacunae in knowledge can greatly complicate, possibly completely jeopardize, the design of an adequate cleanup program. At Fernald there is still considerable disagreement regarding the cause of some of the underground contamination. Does it originate from the Waste Pit area or some other location?

Given the newness of some of the technologies being employed, very often the "latest" in clean-up technologies is shortly seen as a disaster. But contractors who have spent a considerable amount of the taxpayers' money are reluctant to admit to such failures. Nor do they want to jeopardize lucrative DOE contracts. Further, the threat of lawsuits may also make contractors less than candid about their unsuccessful or failed projects. Finally, the DOE is reluctant to admit to failures that might jeopardize future funding requests.

Theoretically, the best way to remediate a contaminated site is to completely decontaminate it, that is, remove in situ the contaminants from soil and groundwater. An example of in situ decontamination would be steam stripping where the idea is to remove volatile organic compounds from contaminated groundwater by injecting steam directly into the polluted water. In other words, the contamination is removed without pumping out and moving the contaminated water to another location.

The concept is attractive because, supposedly, there would be no follow-up costs. (This contrasts with the long-term costs to excavate and remove wastes that then have to be re-packaged and placed in containers and/or repositories. These, in turn, have to be constructed and maintained.) However, most of the decontamination in situ strategies are in a very experimental stage. It is unclear if they offer the nice, tidy, perfect solutions so often described by DOE and their contractors.

The number of possible decontamination methods is as varied as the number of possible contaminants. In the following paragraphs, we briefly describe vacuum extraction wells, steam stripping, hydrous pyrolysis/oxidation, in situ chemical oxidation, enhanced evaporation, phytoremediation and the use of anaerobic bacteria. Each of these methods has its own drawbacks or unanswered questions.

Vacuum extraction wells

These wells use a vacuum to increase the amount of volatile organics that can be pumped from a groundwater recovery well[4]. This is especially useful in low-permeability groundwater areas where the water-conducting layer consists of small particles, such as clay. The vacuum helps volatilize organic chemicals from the water, i.e. to move contaminants from the soil or water phase into the gaseous phase. Vacuum extraction wells can be used in combination with other remediation technologies.

Many questions still remain unanswered about this technology. Are all the hazardous materials trapped in filters or are they being released to the environment? How are the extracted gases trapped? What contaminants are released to the environment? Is this not a way of simply dispersing the contaminants to a wider area?

Steam stripping and hydrous pyrolysis/oxidation

Steam stripping is a method used to remove volatile organic compounds from groundwater. Steam is injected into a contaminated groundwater plume, heating the area to above the contaminants' boiling point.[5] The vaporized contaminants can then be removed by vacuum extraction wells (see above).

Since the steam stripping operates in tandem with vacuum extraction wells it suffers from some of the same defects and questions, in terms of how much material is actually being removed and how much is being re-inserted into the environment in another location.

A simultaneous process called hydrous pyrolysis/oxidation destroys contaminants that are not removed by the extraction wells, but are susceptible to heat.

In situ chemical oxidation

In situ (in place) chemical oxidation is used to break down volatile organic compounds in groundwater.[6] Chemical oxidants such as potassium or sodium permanganate, or hydrogen peroxide, are first injected into the ground. Then the oxidants will retrieve electrons from (oxidize) the volatile organic compounds, converting them into nontoxic chemical compounds.

In situ treatment methodologies are used at Pantex to reduce the concentration of contaminants in the Ogallala Aquifer.

Tests at Portsmouth showed that this method is effective, but that there is also a danger of the oxidants migrating away from the wells, which could result in additional environmental damage. To prevent this, extraction wells must be positioned around the injection wells.

Phytoremediation

This system was proposed by Ohio EPA to decontaminate volatile organic compounds in groundwater at the Portsmouth facility.[7] The idea is to plant several tree species in rows above the groundwater plume. More than one species should be chosen in case an entire species is destroyed, due to a species-specific susceptibility to parasites or chemicals in the groundwater. (The fact that several different types of tree species were recommended shows the limits of current knowledge about this technique.)

Mature trees consume about 3,000 gallons of groundwater per day and acre. Organic compounds are removed from the water and captured in the trees' root systems. These compounds do not accumulate in the trees, but are transported upwards to the leaves. Here the compounds are broken down by ultraviolet light and subsequently transpired, along with the water vapor through the leaf pores.

This technology is sufficiently new that its effectiveness is not known. One important question is about the residual chemicals that are transpired with the vapor. Which chemicals are broken down and which ones are re-released?

Biotechnology involving bacteria

This experimental approach is likely to encounter stiff opposition from environmentalists. Efforts are under way to genetically engineer radiation-resistant organisms that can degrade radioactive waste. The organism would combine a natural resistance to radioactivity found in certain bacteria with other bacteria that have the ability to oxidize and precipitate heavy metals, including uranium and plutonium. The hope is that, if the precipitated metal can somehow be collected, the cleanup will be complete. Even if this cannot be achieved, the soluble radionuclides are at least removed from the aqueous phase and are thus less likely to migrate off-site.

All of the candidate bacteria are anaerobic, which means they live in oxygen-free media. Instead of oxygen, they use heavy metals as electron acceptors, thereby reducing the heavy metals. In the case of uranium, the soluble uranium (VI) is reduced to uranium (IV), a precipitate. At the same time, the bacteria needs donors, which can be chemicals present in contaminated groundwater. Hence, such a reaction between radionuclides and chemicals involving anaerobic bacteria could precipitate heavy metals and simultaneously break down (by oxidation) toxic chemicals.

There are many major questions about this experimental technology. What are the potential hazards associated with releasing these genetically engineered, radiation-resistant, organisms into the environment? What else are the organisms resistant to? What impact will they have on other natural organisms such as plants, humans, fish, and insects? What happens if these organisms get into groundwater and aquifers?

This technology is not fully developed. DOE will want to test the organisms at DOE nuclear weapons sites. Whether this is appropriate or not should be a subject for full public debate.

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[1] US DOE. "NDAA Report to Congress on Long-term Stewardship", January 2001: 29.

[2] Pollet, G & Weida, B., *Missing the Path to 'Cleanup' :False Economic Assumptions and Failed Contracting practices in the Department of Energy's Nuclear Weapons Environmental Programs* , March 1998 :1

[3] INEEL, « Technology Needs Statement, », http://www.inel.gov/st-needs/need-detail.asp?id=877.

[4] US DOE, Portsmouth Annual Environmental Report for 1999, DOE/OR/11-3052&D1, January 2001, p. 3-5.

[5] US DOE 2001, p. 3-6.

[6] *Ibid*, p. 3-4.

[7] *Ibid*, p. 3-2 f.