Capturing a "Mixed" Contaminant Plume: Tritium Phytoevaporation at Argonne National Laboratory's Area 319

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Abstract

Large green plants have the capability to move significant amounts of soil solution into the plant body through the roots and evaporate this water out of the leaves as pure water vapor in the transpiration process. It is known that tritium, as tritiated water, is partly directly incorporated in biological tissues, and partly transpired by plants as tritiated water vapor.

An innovative application of engineered phytoremediation has been deployed at the Argonne National Laboratory (ANL) site in Illinois. At this site, tritium is present as a co-contaminant with Volatile Organic Compounds (VOCs) in the groundwater, approximately 30 ft (10 m) deep in the glacial subsoil. In 1999, the U.S. Department of Energy (DOE), through the Accelerated Site Technology Deployment (ASTD) Program funded the deployment of a phytoremediation system in the 317/319 areas with the objectives of minimizing water infiltration into the source soils, stabilizing the treated soil surface to prevent erosion, runoff, and downstream sedimentation; hydraulically contain tritium and VOCs migration with the groundwater, and continuing remediation of the residual VOCs in the plume.

The phytoremediation system installed involves the use of high-transpiring, deep-rooted phreatophytes to provide hydraulic control of the contaminated plume. While the fate of the VOCs in a phytoremediation system has already been demonstrated in a number of cases, this installation is pioneering the use of phytoevaporation for the removal of tritium from the subsoil.

A preliminary evaluation conducted by ANL prior to the inception of the project indicated that even assuming that all of the tritium (at the highest concentration in the plume) were transpired by plants, air emissions of tritium would result in an inconsequential exposure for a person at the site boundary, and be well within the National Emission Standards for Hazardous Air Pollutants (NESHAPS).

Soon after DOE funded the project, the U.S. EPA and DOE agreed to include this remediation technology deployment in the projects evaluated by the EPA Superfund Innovative Technology Evaluation (SITE) Program. Under this program, the EPA is independently monitoring and evaluating the technology’s performance at the ANL-E 317/319 sites, in addition to the scheduled monitoring activities conducted by ANL.

Phytoremediation at the 317/319 areas at Argonne was deployed in the summer of 1999 achieving a significant, 33% cost saving over the baseline traditional technology of capping and extraction wells. As the plants mature, performance data will validate further predicted cost savings on operations and maintenance, as the existing extraction wells will be closed and the plants will generate no secondary waste.

Introduction

Tritium is a soft (low-energy) beta emitter radionuclide. As such, it is easily shielded by human skin, paper, and approximately 6 mm of air. It is, however, hazardous when taken internally via ingestion, inhalation, and absorption. It decays to Helium-3 and has a half-life of 12.6 years. As it shares the chemical and physical properties of hydrogen, it is found as an environmental concern typically as tritiated water. As for most of the radionuclide contaminants, its radiological hazard exceeds the chemical hazard and thus levels of environmental concern in terms of radioactivity translate into minute amounts in terms of mass. Sources and an estimated inventory of tritium are reported in Table 1 (from: www.hfbr.bnl.gov/hfbrweb/hdbl1079a.html#ZZ5).
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Table 1. Sources and estimated inventory of Tritium.

<table>
<thead>
<tr>
<th>Source</th>
<th>Inventory</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural (cosmic rays, m steady state)</td>
<td>$70 \times 10^6$ Ci</td>
</tr>
<tr>
<td>Nuclear test explosions (1945-1975)</td>
<td>$3 \times 10^6$ Ci (most decayed)</td>
</tr>
<tr>
<td>Nuclear power and defense industry releases</td>
<td>$1.2 \times 10^6$ Ci/year</td>
</tr>
<tr>
<td>Commercial devices (radioluminescent, neutron generating)</td>
<td>$1 \times 10^6$ Ci/year</td>
</tr>
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Tritium is known to be directly incorporated in water and biological tissues. Its average biological half life in the human body is 7.5 to 9.5 days. In plants, it is taken up as tritiated water and subsequently mostly transpired as tritiated water vapor ([IAEA, 1981]). Studies conducted by the University of Heidelberg in natural ecosystems suggested that heavy plant growth might pull water from the soil at a rate so fast to considerably reduce tritium diffusion and therefore isotopic mixing in the groundwater ([IAEA, 1967]). A small portion of the tritium is accumulated in plants as cell water or into tissue. Work conducted at the Maxey Flats Disposal Site concluded that trees could be bioindicators of tritium contamination ([Rickard and Kirby, 1987]). In any case, the accumulation in plants appears to be of short duration (4 to 37 days) ([IAEA, 1981; Fresquez et al. 1995]).

Tritium contamination of groundwater is present at the 317/319 areas at Argonne National Laboratory-East (ANL-E), as a result of past operations. Low levels of tritium, as well as VOCs, have been detected in the groundwater in this area. The contaminated plume, approximately 30 ft (10 m) deep in the glacial subsoil, is migrating toward the southern boundary of the site through a series of sand layers, into the adjacent Waterfall Glen Forest Preserve of DuPage County.

In 1999, the U.S. Department of Energy's Office of Environmental Management, through the Accelerated Site Technology Deployment (ASTD) Program, jointly funded the deployment an innovative phytoremediation system in the 317/319 areas with the following objectives: (1) minimize water infiltration into the 317 French Drain area soils, some of which were treated previously by soil mixing, thermal desorptions and iron addition; (2) stabilize the treated soil surface in the 317 French Drain area to prevent erosion, runoff, and downstream sedimentation; (3) hydraulically contain groundwater migration and continue remediation of the residual VOCs within the source area, and (4) hydraulically contain the VOCs and tritium plume south of the 319 area landfill.

Large green plants are capable of moving significant amounts of soil solution into the plant body through the roots and evaporate this water out of the leaves as pure water vapor in the transpiration process ([Chappell, 1998; Wullschleger et al. 1998]). Plants transpire water to move nutrients from the soil solution through the roots (which function as a highly dispersed, fibrous uptake system) to leaves and stems, where photosynthesis occurs, and to cool the plant. While the use of trees to hydraulically control and remediate contaminated groundwater plumes at depths in the range of five to more than 30 ft has been successfully applied at commercial installations ([Nyer and Gatilff, 1996]) for the remediation of VOCs and excess nutrients, its application to treat tritium contaminated groundwater has never been conducted before.

**Technological Approach and Expected Results**

The use of trees to remediate and contain contaminated groundwater has been successfully demonstrated in treating contaminated groundwater. Applied Natural Sciences, Inc, (ANS) demonstrated the use of phreatophytic trees (i.e., plants such as poplars and willows that do not rely on precipitation water but seek water deep in soils) with its TreeMediation® and TreeWell® systems, that use a unique and patented process to enhance the aggressive rooting ability of selected trees to clean up soil and groundwater up to 50 ft deep. Under a CRADA, ANL-E and ANS researched phytoremediation applications since 1994.
The 317 and 319 areas are located on the extreme southern end of the ANL-E site, immediately adjacent to the DuPage County Waterfall Glen Forest Preserve. The 317 area is an active hazardous and radioactive waste processing and storage area. In the late 1950s, liquid waste was placed in the unit known as the French Drain. Since that time, this waste has migrated into underlying soil and groundwater. The principal environmental concern in the 317 area is the presence of several VOCs in the soil and groundwater and low levels of tritium in the groundwater beneath and downgradient of the site. The 319 landfill and French Drain area are located immediately adjacent to the 317 area. The principal environmental concern in the 319 area is the presence of radioactive materials in the waste mound, in the leachate in the mound, and in the groundwater downgradient of the landfill. Several interim actions have already been implemented to reduce the VOC and tritium releases from these areas, as the result of the Resource Conservation and Recovery Act (RCRA) Facility Investigation (RFI) conducted from December 1994 through September 1996. Currently, existing mechanical extraction wells remove approximately 20,000 m³/yr of contaminated groundwater, which are sent to ANL’s water treatment plant. While the VOC contaminants are degraded at this facility, tritium concentrations are diluted with other wastewater from the lab and discharged in accordance with regulatory limits.

The hydrogeology at the 317/319 sites is a complex framework of glacial tills interlaced with sands, gravels, and silts of varying character, thickness, and lateral extent. The subsurface is a complex arrangement of approximately 60 ft of glacial geologic deposits over Silurian dolomite bedrock. The glacial sequence is comprised of Lemont drift overlain by the Wadsworth Formation. Both units are dominated by fine-grained, low-permeability till. Permeable zones of varying character and thickness are present in each. These materials range from silty sands to sandy, clayey gravels to gravelly sands. In some locations, pure silt was encountered. If deep enough, this silt was saturated, and it is assumed to play an important role in the flow of groundwater in the study area. The permeable zones have a wide range in shape, their thicknesses range from less than one ft to roughly 15 ft and they have limited lateral extent (Quinn et al. 2000).

On the basis of a preliminary agronomic assessment, hybrid willow and hybrid poplar trees were selected for the system. In the summer of 1999, a total of approximately 800 trees were planted in three locations: the 317 French Drain area, south of the 317 French Drain area and 319 area landfill (the 317 and 319 Hydraulic Control areas), and in the waste trench south of the 319 area landfill. Approximately 160 hybrid poplars were planted in the area of interest of tritium contamination. This system is expected to prevent the further generation of contaminated groundwater in the source area by degrading the contaminants, and to prevent the further migration of these plumes by removing groundwater from saturated zones downgradient from the source area. Figure 1 shows the location of the plantings. The installed system consists of plantings of hybrid willows and special deep-rooted hybrid poplars. The willows were planted in the source area (317 French Drain area) deeper (16-20 ft) than is normal for horticultural plantings, but without some of the special modifications used with the deep-planted poplars.

In the 317 and 319 Hydraulic Control areas, poplar trees were planted in boreholes spaced 16 ft apart drilled down to the contaminated aquifer using ANS’s TreeWell® system. This technology was selected, in consideration of the hydrogeological setting of the site, to target root growth in the contaminated glacial-drift permeable unit approximately 30-ft deep. The poplars were planted in two-ft diameter caisson boreholes lined with plastic sleeves in order to direct the roots exclusively to the main contaminated aquifer. These boreholes were filled with a mixture of topsoil, sand, peat, and manure to promote root growth and tree development. The capillarity of the mixture provides an added benefit of drawing water to where it is available to the young trees. All boreholes were also provided with aeration tubes to ensure a supply of air to the growing roots. Figure 2 presents a diagram of a TreeWell® installation.

Planting phreatophytic trees at the capillary fringe in the year 1999 is expected to provide full hydraulic control by the year 2003 (see below) and be self sustaining for the full-expected life of the engineered plantation. Hybrid poplar and hybrid willow trees typically have a life span of about 40 years. The Path to Closure Plan committed ANL-E to have all remedial work at the 317/319 areas completed by October 2000.
Figure 1. Planting locations at the 317/319 areas.

Figure 2. Diagram of a Tree Well installation.
ANL-E installed 48 groundwater monitoring wells on the phytoremediation project site to track the performance of the phytoremediation system. Soon after DOE funded the project, the U.S. EPA and DOE agreed to include this remediation technology deployment in the projects evaluated by the U.S. EPA Superfund Innovative Technology Evaluation (SITE) Program. Under this program, the U.S. EPA will independently monitor and evaluate the technology’s performance at the ANL-E 317/319 sites in addition to the scheduled monitoring activities conducted by ANL-E. Monitoring activities have started at the completion of the construction phase. Root development will be observed through specially designed viewing ports (minirhizotrons).

At the end of the remedial process, when a final analysis will verify the absence of the contaminants in the biomass, the trees will be cut down at ground level, chipped, and air dried. The roots will be left in place to decay through natural processes and the chips will be reused on site as mulch for the planting of native prairie species, in accordance with the planned final restoration of the area.

**Planning Considerations**

Preliminary to the implementation of the system, a modeling study was conducted to assess potential air emission hazards, according to existing regulations (NESHAPS 40 CFR 61 Subpart H). Using maximized assumptions on tritium concentration in the groundwater and plant transpiration rates, emissions via transpiration were calculated for the four years from the time of planting to the time of canopy closure. Results are reported in Figure 3. The derived exposure rates to the nearest member of the public were calculated as required using the U.S. EPA CAP-88 PC program and resulted in values ranging between $6.32 \times 10^{-6}$ mrem/yr in the first year to $2.58 \times 10^{5}$ mrem/yr during year four. As the

![Figure 3. Tritium transpiration rates at maximum concentration.](image)
regulatory standard is 10 mrem/yr, the added exposure was considered inconsequential. In summary, air emission models indicated that even assuming that all of the tritium (at the highest concentration ever found in the plume) were transpired by plants (at the highest transpiration rates), air emissions of tritium would be well within the National Emission Standards for Hazardous Air Pollutants (NESHAPS). At the same time, tritium in the groundwater plume would be efficiently controlled.

To support the deployment of the phytoremediation system, a groundwater flow model was also developed. Flow modeling was conducted initially to model the natural, transient changes in the flow field caused by seasonal changes in recharge to the aquifer. The model was calibrated to approximately 10 years of water level measurements from site monitoring wells. Anticipated effects of the phytoremediation system were included. The model, updated to include the as-built configuration of the phytoremediation system, indicates that the as-built plantation will provide hydraulic containment by the fourth year of growth even during the winter months when the trees are dormant (Quinn et al. 2000).

Cost Savings and Other Advantages

The conventional, baseline method of remediation of the 317/319 areas originally planned for deployment in lieu of phytoremediation included placing an asphalt cap over the VOCs source area and installing extraction wells (pump-and-treat) downgradient of the source areas, from which contaminated water would be withdrawn and discharged to a lift station, which pumps water to Argonne's waste treatment plant.

The phytoremediation installation was installed with a cost saving of 33% compared to the expected cost of the baseline approach. The plant-based system is expected to have lower operating and maintenance costs also: preliminary evaluations put the cost savings in O&M over the lifetime of the deployment at 40% compared to the baseline approach. A significant cost saving (as well as a reduction in risks of spills and worker exposure) is the avoidance of secondary waste (pumped groundwater) and related treatment. These cost savings will be demonstrated as the extraction wells are shut off, expectedly in 2003.

In addition to this actual reduction in cost, a number of technical reasons made the phytoremediation choice more advantageous versus the baseline technology. As mentioned before, the subsurface at the site may be comprised of units of widely varying lateral or vertical extent, with gradational or sharp transitions in permeability. The fibrous nature of roots allows the trees to penetrate and remediate both the relatively fast-flowing pore spaces and the less permeable zones. Fundamentally, this distinguishes phytoremediation from extraction wells, which remove water mainly from the most permeable aquifer media.

Phytoremediation was considered more acceptable than the baseline also because of the ability of trees to actively promote and assist in the degradation of the contaminants at the VOC source area, which the baseline asphalt cap would not do, with expected reduction in cleanup times. The presence of vegetation was also considered an optimal fit with the planned future land use of the contaminated site and adjacent areas, as the phytoremediation plantation will contribute to increase soil fertility to host subsequent prairie species.

Literature Cited