DEPLOYMENT OF AN INNOVATIVE THERMALLY ENHANCED SOIL MIXING PROCESS AUGMENTED WITH ZERO-VALENT IRON* 

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1 INTRODUCTION

An innovative in-situ soil treatment process, referred to as soil mixing/thermally enhanced soil vapor extraction (SM/TESVE), was used to remediate the 317 Area of Argonne National Laboratory-East (i.e., Argonne), which is contaminated with volatile organic compounds (VOCs). Following the initial soil treatment, polishing was required to reduce residual concentrations of contaminants.

A study of polishing methods was conducted. It determined that injecting metallic iron particles into the soil, in conjunction with soil mixing, would reduce residual VOC concentrations more effectively than the original conventional soil ventilation approach. After the effectiveness of iron injection was verified, it replaced the soil ventilation step.

The modified process involved mixing the soil while hot air and steam were injected into it. Off-gases were captured in a hood over the treatment area. During this process, an iron slurry, consisting of up to 50% iron particles in water with guar gum added as a thickening agent, was injected and mixed into the soil by the mixing equipment. Approximately 6,246 m³ (8,170 yd³) of soil was treated during this project. Confirmatory samples were then collected. In these samples, VOC concentrations were usually reduced by more than 80%.

2 TREATMENT AREA

The 317 Area is an active hazardous and radioactive waste processing and storage area. It is located on the extreme southern end of the Argonne site. From the mid-1950s to the early 1960s, liquid waste was placed in a unit in the 317 Area known as the French Drain. Since that time, this waste has migrated into underlying soil and groundwater. Figure 1 shows the current arrangement of waste management facilities in the 317 Area. Photo A shows the treatment area before in-situ soil treatment began.
FIGURE 1 Map of the 300 Area
2.1 PHYSIOGRAPHY

The treatment area is situated within the Wheaton Morainal Country of the Great Lakes Section of the Central Lowland Physiographic Province. This terrain has a gently sloping topography and shallow surface relief, which is typical of older Wisconsinan glacial drift deposits. This relief is modified in the vicinity of streams and tributaries. The natural topography of the treatment area consists of bluffs adjacent to the Des Plaines River floodplain.

2.2 LOCAL GEOLOGY

Argonne is situated in the Valparaiso end moraine that locally trends in a northwest-southeast direction. Geologic sections compiled from soil borings at and near Argonne (Olimpio 1984; Killey and Trask 1989) and nearby geologic exposures (Hansel and Johnson 1987) indicate that Quaternary sediments of the Wedron Group lie directly on dolomite bedrock of the Silurian Age. The Wedron Group is composed of sediments from two glacial depositional phases. The Lemont Drift was deposited directly on the bedrock surface about 16,200 years before present, and the Wadsworth Formation was deposited on top of the Lemont Drift about 15,500 years before present.
2.2.1 Wadsworth Formation

The contaminated soils that underwent treatment are situated in the Wadsworth Formation. The Wadsworth Formation is a poorly sorted glacial till characterized by brown to gray silty clay intermixed with minor amounts of gravel composed primarily of dolomite and shale. The till was deposited under subglacial and supraglacial environments. A few lenses of silt, sand, and gravel are present among the till. Each lens ranges from a few inches to a few feet in thickness. Lenses with a lateral extent of more than a few thousand feet have been reported at the Palos Forest Preserve and at the Advanced Photon Source (APS) site at Argonne (Olimpio 1984; Killey and Trask 1994). The total thickness of the Wadsworth Formation ranges from 6 to 10 m (20–33 ft) at the treatment area to 25 to 29 m (81–95 ft) at the APS site (Killey and Trask 1994) and at the 800 Area Landfill.

The Wadsworth Till is calcareous. An analysis of the grain-size distribution of 47 till samples collected near the APS site revealed that the fine-grained matrix of the till averages 16% sand, 45% silt, and 39% clay. The major clay mineral is illite, which accounts for more than 70% of the clay particles. The upper part of the Wadsworth Till has been weathered. Iron stains found along fractures in till and weathered brownish till were noted in well logs. The weathered zone varies from 2 to 8 m (8–25 ft) below ground surface (bgs).

2.2.2 Lemont Drift

The Lemont Drift contains a sequence of alternating sorted sediments and tills deposited in a near ice margin environment (Hansel and Johnson 1987). In the 317 Area at Argonne, the Lemont Drift can be divided into three units: two stratified preglacial fluvial deposits with a silty till unit between them. The bottom stratified proglacial unit lies on the dolomite bedrock. It contains a combination of layers of coarse-grained sediments ranging from silt, silty sand, and sand to sand and gravel, with a total thickness ranging from 1.5 to 9 m (5 to >30 ft).

2.2.3 Bedrock Stratigraphy

The uppermost bedrock unit across the Argonne site is the Racine Formation. The Racine Formation is the youngest formation of the Silurian Series and has a maximum thickness of about 91 m (300 ft) in northeastern Illinois (Willman 1971). Other, thinner dolomite formations make up the rest of the Silurian Age dolomite bedrock.

The Silurian system overlies about 1,291 m (4,000 ft) of Ordovician and Cambrian sedimentary rock with alternating carbonate, shale, and sandstone formations. The sandstones include the Ordovician St. Peter sandstone and the Cambrian Mt. Simon sandstone, which have historically been major sources of potable groundwater in northeastern Illinois.
2.3 HYDROGEOLOGY

2.3.1 Groundwater Occurrence and Flow Direction

In northeastern Illinois, groundwater is obtained from three major sources: glacial drift aquifers, shallow bedrock (dolomite) aquifers, and deep bedrock (sandstone) aquifers. Silurian dolomite is the primary water supply aquifer beneath the Argonne site. This dolomite yields water primarily from fractures, bedding planes, and solution cavities. In northeastern Illinois, the aquifer is generally recharged from overlying glacial deposits.

Groundwater within the glacial deposits typically flows in the coarser, more permeable units. These may include stratified glacialfluvial sand and gravel, reworked mixtures of sand and gravel with till, eolian and beach sands, and windblown silt (loess). In some instances, silt may be more permeable than the surrounding till units. Therefore, in a glacial environment, paths of groundwater flow are typically complex.

Given the heterogeneous nature of the coarse-grained glacial deposits, groundwater flows to the south-southeast toward local groundwater discharge areas, mimicking the local ground surface topography. Groundwater discharge areas include local drainage ways and creeks; groundwater seeps south of the treatment area; and the Des Plaines River, which is about 457.2 m (1,500 ft) south of the treatment area.

2.3.2 Hydraulic Gradients and Conductivity

Data collected in the RCRA facility investigation (RFI) conducted across the 300 Area showed that horizontal hydraulic gradients in the glacial deposits across the treatment area ranged from 0.011 ft per foot (ft/ft) to 0.058 ft/ft. Vertical hydraulic gradients in the treatment area were about 0.04 ft/ft in the glacial deposits.

RFI data also indicated that hydraulic conductivity values for wells screened in the glacial deposits ranged from $1.5 \times 10^{-6}$ to $3.6 \times 10^{-2}$ cm/s, indicating significant variation in permeability because of the varying physical characteristics of the granular deposits. Laboratory values obtained from using Shelby tube samples ranged from $4.5 \times 10^{-8}$ to $2.91 \times 10^{-6}$ and were consistent with the silty clay and the clayey silt materials encountered in the glacial till deposits.
2.4 SITE CONTAMINATION

RFI information indicated that the depth of detectable contamination in the treatment area soils varied from less than 2 m (8 ft) on the western edge of the contaminated area to 9 m (28 ft) on the southern and eastern edges. Figure 2 is an isoconcentration map showing the lateral distribution of VOCs based on data collected from 43 soil borings conducted before the contaminated soils were treated.

3 PROJECT RATIONALE

The RFI showed significant groundwater and surface water contamination emanating from the 317 Area French Drain. The overall remedial action was designed to minimize the exposure and release of contaminants to surface water, groundwater, and air. Soil treatment would significantly reduce the source of contaminants escaping to the groundwater and thereby reduce the potential chemical force that would drive the contamination to groundwater. Because of operational and regulatory issues related to ex-situ treatment of contaminated soil, an in-situ treatment method was selected. In-situ treatment would not require any permit modifications, would minimize waste handling and disposal activities, and would be less disruptive of critical operations in the 317 Area.

4 MOBILIZATION OF SOIL TREATMENT SYSTEM

Argonne teamed with In-Situ Fixation, Inc. (ISF), of Chandler, Arizona, to implement the scope of work for this project. ISF is an environmental remediation firm experienced in the in-situ treatment of contaminated soil and groundwater. ISF was responsible for all major aspects of the project work plan. Argonne conducted daily oversight and was responsible for all sample collection and analysis, project reporting, cost and schedule tracking, and evaluating the success of the project. Argonne selected appropriate polishing technologies for full-scale deployment.

Before soil treatment could begin, several activities were necessary to prepare the site to accept the treatment process. Rerouting of power lines, removal of a crane control tower, surveying of the area for radioactivity, and removal of radioactively contaminated surface gravel were completed before or simultaneously with contractor mobilization. These activities are discussed in detail in the following sections. More detailed descriptions of the treatment equipment are included in Section 5.
4.1 SITE PREPARATION AND SETUP

All equipment arrived on the site via flatbed semitractor trailers. To minimize exposure to elevated ambient radiation levels inside the 317 Area, the flatbed trucks were staged outside the 317 Area, adjacent to the north fence. A 20-ton crane, which was operated by Stevenson Crane and staged inside the 317 Area, off-loaded the equipment from the trucks. Equipment was placed inside the 317 Area, then moved by ISF to designated locations.

4.1.1 Work Zones

The boiler plant, vapor treatment plant, tool and maintenance equipment container, and diesel fuel tank were staged north of the treatment area. Figure 3 shows the work area layout. As work progressed, appropriate zones — exclusion, support, and decontamination — were established to facilitate the movement of project personnel throughout the work area and to give them access to project equipment for operation and maintenance.

- The exclusion zone included the soil treatment area and surrounding areas where treatment equipment was staged. The treatment area was bounded by the storm water diversion ditch to the north, the North Vaults to the south, the container storage area to the west, and the eastern edge of the North Vaults to the east.

- The support zone was located north of the treatment area and encompassed the rest of the work area, excluding the exclusion zone. The boiler plant, vapor treatment area, tool and maintenance equipment container, and diesel fuel tank were located there. The support zone was marked with yellow tape and orange snow fencing where access was possible.

- The decontamination zone was located southeast of the treatment area and North Vaults, just north of an Argonne wastewater treatment plant manhole. During soil sampling activities, soil sampling equipment was stored in a small tent just west of the decontamination zone.

Throughout field activities, a vehicle access zone was located north of the support zone. Deliveries to the work site took place in this zone. This area had been screened by Argonne Health Physics (HP) personnel before the project. Vehicles entering this zone did not have to be screened by HP before leaving the 317 Area.
FIGURE 3 Work Area Layout
4.1.2 Electrical Power Reroute

Overhead electrical lines and power poles were removed to allow unimpeded movement of the Caterpillar 245-D/auger assembly within the treatment area and of equipment to and from the decontamination zone.

4.1.3 Contaminated Gravel Removal

HP personnel surveyed the surface gravel areas that encompassed the treatment area. The purpose was to identify areas with elevated levels of radioactivity. Such material would be removed during the site preparation phase to prevent radioactive particles from becoming entrained in the subsurface soils during the soil mixing phase and to prevent possible contamination of equipment staged in the support zone.

HP personnel used a shielded personal radiation monitor (PRM) 5-3 to measure gamma energy from radioactive particles in the surface gravels. A total of 33 areas of gravel in the treatment area were found to contain elevated levels of radioactivity. Areas showing elevated levels were delineated with blue spray paint. Levels ranged from 3,800 to 130,000 counts per minute (cpm). At the direction of HP, surface gravel in areas with readings that exceeded 10,000 cpm were excavated until monitoring showed levels to be below 10,000 cpm. Photo B shows Argonne Waste Management and HP personnel excavating areas of gravel having elevated levels of radioactivity.

Radioactively contaminated surface gravels were removed after the surface survey was completed. At three areas, two just north of the North Vaults and one in the northwest part of the treatment area, contamination was found to extend through the entire surface gravel interval, but it was at ambient levels at the interface with native soils. At these areas, a backhoe was used to remove radioactively contaminated gravels. At all other areas, gravel was manually removed with shovels. Contamination in these areas extended only a few inches into the surface gravel. Contaminated gravel was placed into four 208-L (55-gal) drums and four reinforced 1.5-m³ (2-yd³) polyethylene bags known as supersacks. Radioactively contaminated gravel was used by Waste Management personnel as void filler for low-level radioactive waste shipments.

4.1.4 Water Supply Construction

A water supply system was established during site preparation activities to provide a constant source of water for injecting steam and water during soil treatment activities. Argonne used water from an on-site process water treatment plant as a source throughout the entire project. Rigid polyvinylchloride (PVC) piping was hooked up to a connection inside the east end of the water
treatment plant. Water was conveyed above ground for approximately 549 m (1,800 ft) through wooded areas, then underground from the road north of the 317 Area to the boiler plant. Flow was controlled both at the CWTP and at the boiler plant in the support zone. During cold weather, the water line was flushed empty at the end of each day to prevent pipe damage as a result of freezing.

Before being used to generate steam, CWTP water was treated to remove silt particles and adjust the pH (to minimize mineral deposits in water lines). Water was conveyed through a series of water softening and additive tanks, filters, and holding tanks, then sent to the boiler where it was converted into steam.

4.2 TREATMENT SYSTEM SETUP

The soil treatment system consisted of a mobile soil mixing rig, steam (i.e., boiler) plant, and off-gas treatment system. When the treatment process was modified to include iron addition, an iron batch plant was constructed. The steam plant, off-gas treatment system, and iron batch plant were located in the support zone, north of the treatment area. Figure 3 shows the locations of these treatment system components.
The Interim Action Work Plan for treating soil in the vicinity of the 317 Area French Drain included the deployment of a soil polishing treatment system designed to reduce residual VOC concentrations even more after SM/TESVE. The polishing system that was proposed — soil ventilation (also known as conventional soil vapor extraction) — was found to be unsuitable for the soil conditions that existed after SM/TESVE. An alternate soil polishing system that involves the injection of metallic iron particles was developed during a series of treatment studies (Kibler 1997). These studies were performed in the treatment area prior to full-scale treatment.

The boiler plant was constructed first. Steam from the boiler was conveyed through a cast-iron line to the north edge of the treatment area. A flexible, iron-mesh-covered rubber line conveyed the steam to the swivels located at the top of the Kelly bars for injection into the mixed soils.

The off-gas treatment area was constructed after the boiler was completed. Off-gas containing VOCs was diverted to the off-gas treatment system located inside a tent immediately east of the boiler plant. Canisters containing activated carbon used to remove VOCs from the off-gas were located at the north end of the off-gas treatment area.

The iron batch plant was constructed during two separate mobilizations. At both times, the batch plant was located immediately southwest of the boiler plant. In the first mobilization, the batch plant was constructed to support an experiment testing the effectiveness of zero-valent iron as a polishing treatment. After the experiment, the batch plant was dismantled and returned to the vendor. The batch plant was built again to support the full-scale deployment of iron as a final polishing treatment across the treatment area. At this time, the iron batch plant was enclosed in a tent because of inclement weather.

Zero-valent iron powder was mixed with guar gum and water in the batch plant. Hot water was conveyed from the boiler to the batch plant. As soil mixing proceeded, the iron slurry was pumped to the Kelly bars, where it was combined with pressurized steam and injected into the subsurface soils. The iron batch plant was located close to the treatment area and the Caterpillar 245D excavator to allow ISF personnel to monitor auger advancement.

4.3 STORMWATER DIVERSION AND CONTROL

During ongoing treatment in the area, the potential to generate silt-laden storm water runoff was great as a result of disturbance of the soils in the area and the known flow of storm water through the area. The following sections describe the erosion control and water diversion efforts that occurred on the site during the duration of the work.
4.3.1 Original System

The original erosion control system was essentially a diversion system designed to channel the flow of storm water around the treatment site and dump the water back into the site's principal storm water flow system. Figure 4 shows the treatment area, the original flow pattern through the 317 Area, and the storm water diversion system placed over the area to control runoff. Before the treatment system was set up, storm water flowed from a low point (A) north of the 317 Area road, across the road, and into the 317 Area. Water then flowed south into the 317 Area in a sheet-flow pattern (B) and eventually collected in a low point near an existing storm water inlet (C). The water then flowed in an underground sewer to a discharge point (D) in a nearby intermittent stream. Before the onset of soil treatment, a system was set up to divert storm water around the treatment area and discharge it to the nearby intermittent stream. As shown in Figure 4, the diversion system consisted of a diversion channel (E) designed to intersect the sheet flow (B) coming from the north. Storm water was then directed through the diversion channel to storm water inlet (C). To catch overland flow to the east, a diversion berm (F) was put in place to direct flow into the storm water inlet (C). Storm water collecting in the treatment area (G) was collected and discharged to the 317 Area Pump Station (H), which was connected to Argonne's sewer system.

4.3.2 Modified System

During active treatment operations in the 317 Area, Argonne personnel discovered that storm water flow (predominantly from snow melt and light rain) was not being diverted as planned by the diversion trench. Rather, it was flowing through the surface gravel, past the diversion trench, and into the treatment area. This resulted in a larger than expected volume of water entering the treatment area. In turn, water contacting the treated soils in the treatment area was found to be contaminated with residual VOCs from the treatment area. To prevent potentially contaminated water from leaving the site, Argonne and ISF made several changes to the storm water diversion system. They lined the diversion channel with plastic sheeting to prevent storm water from flowing into the gravel and appearing again in the treatment area, and they created a water collection basin on the east side of the treatment area. They also placed several temporary plastic holding tanks in the 317 Area to act as settling basins and prevent silt from being discharged to Argonne's sewer system. The water collection system from the treatment area worked as follows: water collected from the treatment area was retained in and around a small pit dug in the soil east of the treatment area. The pit measured approximately 3 m (10 ft) long by 1.5 m (5 ft) deep by 0.9 m (3 ft) wide and was surrounded by silt fencing and straw bales. Photo C shows the water collection basin and associated runoff controls downgradient of the treatment area.
Storm water entering the pit was pumped to the temporary plastic settling tanks. The water was allowed to settle and was siphoned to the 317 Area pump station. Further improvements included placing silt fences and straw bales in the intermittent stream east of the 317 Area. These structures were placed as a safeguard to prevent any silt-laden water from leaving the site.

4.4 COLLECTION OF INITIAL AND FINAL (VERIFICATION) SOIL SAMPLES

Initial and final (verification) soil samples were collected by using a Simpco Drill Team® probe rig mounted on the front of a Gradall 544D-10 forklift. The Drill Team rig is a hydraulically powered machine that uses static force and percussion to advance a 3- or 5-cm (1- or 2-in.) split spoon, or other small-diameter sampling tube, to the prescribed sampling depth. A work platform consisting of a 1-m (4-ft) wide steel grate with railings was welded to the base of the rig to serve as a work platform and sampling support equipment storage area. The rig was moved to each sampling location with the forklift. Photo D shows the soil sampling rig over a treated soil column.

The split spoon is a stainless-steel tube that has either a 4- or 5-cm (1- or 2-in.) outside diameter, is either 61 or 122 cm (24 or 48 in.) long, and is fitted with a replaceable, transparent cellulose acetate butyrate liner. The sampler is driven through the desired interval to fill it, then it is retrieved. Initial soil samples were collected by using the following procedure. The disassembled soil sampling tool and probe rods were scrubbed with a nonphosphate detergent, rinsed with potable water followed by a deionized rinse, and air dried. The sample was reassembled with a new, clean
liner. The sampling tool was lowered in the borehole to the top of the next sampling interval; the sampler was then advanced for the length of the sampler (either 61 or 122 cm [24 or 48 in.]) and retrieved. The soil sample inside the liner was removed by either opening the split spoon, or, if a sample tube was used, by pulling the liner out one end of the tube.

At each boring location, soil was field screened for VOCs on a continual basis. Field screening for VOCs was used to identify sample collection intervals and as a process control technique for estimating, in real-time, the degree of treatment achieved. Field screening was conducted by measuring organic vapor concentrations in the headspace above a sample of the soil. Headspace organic vapor measurements were performed with either a Microtip Photovac® MP-1000 or Mini-Rae® photoionizing detector. Soil samples were placed into a clean widemouth jar covered with aluminum foil and heated in an oven to approximately 60°C (140°F) for about 15 minutes. Following heating, the instrument probe was gently pushed through the foil, and the organic vapor content of the headspace gas was measured.

4.5 SITE RESTORATION ACTIVITIES

This section describes the activities that took place to stabilize the treatment area and restore the support areas following removal of the treatment equipment.
4.5.1 Site Grading

During treatment system dismantlement and final verification sampling and after all equipment was removed from the site, ISF began site grading. Initially, grading activities were confined to pushing muddy soils that flowed off of the treatment area back onto the treatment area to prevent any significant flow from leaving the treatment area during storm events. Photo E shows muddy soils being consolidated with the main body of the treatment area.

Site grading continued after the storm water retaining system and decontamination pad were removed. Grading activities included consolidating all soils in the treatment area and grading the soils surrounding it to promote gentle sheet flow from the area. After consolidation, gravel was placed in a 3 to 6 m (10–20 ft) wide strip around exposed perimeter of the treatment area. (No gravel was placed between the treatment area and the vaults because of inaccessibility.) Gravel was also placed over disturbed portions of the equipment staging area to prevent erosion in these areas.

4.5.2 Storm Water Diversion System Removal

The storm water diversion system was removed by filling in the original diversion trench and removing the berm from the east end of the diversion system. After the berm was removed, the area was graded to facilitate the gentle sheet flow of water. ISF then removed gravel and
accumulated silt and clay from the decontamination pad and placed the materials back into the treatment area. Following removal of the overlying gravel, the underlying plastic sheeting and plastic collection sump were removed, placed into a rolloff box, and shipped off site as a special waste. Following removal of the plastic, the area was regraded, and a layer of gravel was placed over the former decontamination pad. In addition, the storm water collection trench located east of the treatment area was backfilled with the soils removed from that location, and the area was graded with a layer of gravel. The erosion control structures placed around the collection trench and into the intermittent stream were left in place to provide erosion protection until restoration of the area was complete. Four 5,678-L (1,500-gal) plastic tanks used to settle out silt from storm water collected from the treatment area were cleaned out. The clay and silt contained in the tanks were placed onto the treatment area.

4.5.3 Seeding

The treatment area was allowed to dry until the surface was dry. On May 30, 1998, ISF employed a Case 550G bulldozer to rough-grade the treatment area. The bulldozer's track treads pulverized the upper 5 or 8 cm (20 or 30 in.) of surface soils in preparation for seed and fertilizer application. The south side of the treatment area was pulled back to reduce the slope along the vaults. The soil was prepared by adding nutrients to facilitate growth. Following fine grading, fertilizer with a 1:1:1 nitrogen:phosphorous:potassium ratio, combined with a seed mixture consisting of birdsfoot trefoil, perennial ryegrass, and reed canarygrass, was distributed in a quantity of 11 kg (24 lb) per acre. The seed mixture was designed to penetrate the upper 2 m (6 ft) of surface soils. The fertilizer/seed mixture was applied to the graded treatment area via a hydroteed application system. Photo F shows the treatment area being graded before seed application.

5 TREATMENT SYSTEM DESCRIPTION

This section provides a general description of the equipment used to perform the SM/TESVE of soils in the 317 Area at Argonne.

5.1 DUAL AUGER® SYSTEM

The mixing system consisted of a modified Caterpillar 245D excavator (backhoe) that supported and powered a soil mixing assembly. The soil mixing assembly consisted of two overlapping drill flights and mixing blades affixed to a hollow shaft (Kelly bars) that conveyed the heated air, steam, or slurry mixture to the mixing area. The shafts and cutting blades rotated in opposite directions to pulverize the soil and blend in the hot air and steam or reactive slurry mixture into the soil undergoing treatment. Surrounding and encapsulating the mixing augers was a 0.6-cm
Photo F Final Grading of the Treatment Area

(¼-in.) thick steel shroud designed to contain gaseous and particle emissions emerging from the column. Up to 100,000 foot-pounds of torque was transferred to the Dual Auger from the backhoe through the aid of a modified hydraulic system. The backhoe also raised, lowered, and positioned the shroud. Exhaust ports and pressure gauges were fitted on the shroud to control flow through the system and ensure capture of the off-gas. Photo G shows the soil mixing assembly. Photo H shows a detail of the shroud over the soil column undergoing mixing.

The augers could treat soil to a depth of 11 m (35 ft) bgs and an area of 3 m² (35 ft²) on the surface. Actual mixing occurred in a 1.5-m (5-ft) section containing the auger flights and paddles. Steam and hot air were injected through nozzles located at the top of the Kelly bars, through the Kelly bars, and then through nozzles located near the auger flights.

5.2 STEAM AND HOT AIR GENERATION

The boiler plant, which included the boiler, water treatment system, and a 3,785-L (1,000-gal) plastic holding tank, was located inside a rigid-frame tent. Steam was generated by a 3,500-kPa (500-psi), 260°C (500°F) diesel boiler that connected to the auger assembly via a series of steel pipes and flexible, high-temperature, stainless-steel pressure hoses. Makeup water for the boiler passed through a water filtration and softening system to prevent scaling and sediment deposits in the unit.
Photo G  Soil Mixing Assembly (Rubber lines carry steam, air, water, and iron slurry to the top of the Kelly bars)

Hot air was generated by an 11-m$^3$ (375-ft$^3$)/min air compressor that heated the air to 66°C (150°F) at 875 kPa (125 psi). The hot air helped to carry the steam to the Kelly bars and through the system to the nozzles underground.

5.3 OFF-GAS TREATMENT

During treatment, hot air and steam containing VOCs rose from the treatment area below the surface, through overlying treated soil, and escaped into the auger shroud positioned over the treatment column. At this point, the off-gas treatment system took over to remove contaminants entrained in the steam and air. The off-gas treatment system was composed of a shroud that was connected by flexible 25-cm (10-in.) hoses to a water knock-out tank and was further connected in series by rigid piping to a particle filter, followed by a high-efficiency particulate air (HEPA) filter.
that removed small particles. Two activated-carbon canisters used to remove the VOCs accomplished final treatment. The off-gas treatment system was powered by a vacuum blower unit positioned between the HEPA filter and the activated-carbon canister units. This configuration resulted in a vacuum system on the upstream side of the blower to pull untreated steam and air from the shroud to the blower unit. The blower unit then forced the air through the activated-carbon canisters and out to the atmosphere.

Air monitoring at the exhaust port of the activated-carbon canisters was used to determine when breakthrough occurred (i.e., when the activated carbon's capacity to absorb contaminants was exhausted). When monitoring suggested that breakthrough had occurred, the exhausted canister was replaced with a new one.
5.4 IRON SLURRY DELIVERY

Zero-valent iron powder of 40 mesh size was mixed with guar gum in a mixing basin. Photo I shows granular iron being added to the mixing basin. Hot water was conveyed from the boiler through cast-iron pipes to the mixing basin, where it was combined with the iron powder and guar gum to form a colloidal slurry of prespecified viscosity. The iron slurry was then conveyed through a rubber line to the 1,893-L (500-gal) holding tank on the west side of the iron batch plant. As soil mixing proceeded, iron was measured in 38- or 76-L (10- or 20-gal) lifts, then pumped through rubber lines to the swivels, where it was combined with pressurized steam and injected into the subsurface soils.

The iron batch plant was located close to the treatment area and the Caterpillar 245D to allow ISF personnel to monitor auger advancement. Iron was injected into the subsurface in the above volumes at every foot of auger advancement. At the end of each day, pipes containing iron slurry and any unused iron remaining in the holding tank were flushed to the storm water diversion ditch located approximately 3 m (10 ft) south of the iron batch plant. Photo J shows the iron batch plant and the treatment area in the background.

![Photo I Preparation of Iron Slurry in the Iron Batch Plant](image)
In general, Argonne employed two variations of the soil mixing treatment method. The first of these used prolonged soil vapor extraction to remove the majority of VOC contaminants. This system used the SM/TESVE technology exclusively. The second variation involved injecting an iron slurry mixture into the soils while active SM/TESVE soil mixing proceeded to depth. Detailed descriptions of each treatment system variation are provided in the two subsections that follow. Figure 4 is a process schematic showing the SM/TESVE system, including the iron addition component.

6.1 SOIL MIXING

Originally, as discussed in the Interim Action Work Plan for treating soil in the vicinity of the 317 Area French Drain, the SM/TESVE process was to be used first, followed by installation and operation of a soil ventilation system. The soil ventilation system was to be a polishing step that would follow the active mixing by SM/TESVE. It was designed to be a series of air injection and extraction wells screened through the treated soils. Air blowers would force air through the injection wells, and a vacuum blower would then extract contaminated air from the extraction wells and transport it to awaiting activated-carbon canisters. However, because of the specific soil mixing system used (which used steam injection to heat the soil rather than hot air as originally anticipated)
and because a more effective polishing technology was developed, the planned soil ventilation system was never implemented as a polishing step for the soils treated with SM/TESVE.

The SM/TESVE system was operated in a manner that maximized its vapor stripping features so it would remove as much contamination as possible from the soils before implementation of the soil ventilation system. The process of treating the soils with the SM/TESVE system involved two steps. The first step consisted of coring into the subsurface and beginning treatment of the soil. The second step consisted of mixing and stripping VOCs from the soil by mixing and circulating hot air and steam through the soil at specific depths in the soil column. Argonne used initial (pretreatment) sampling data to identify zones in the subsurface where the contaminant loading was the highest. These zones were then correlated with VOC concentrations in the off-gas treatment system by measuring the off-gas with an on-line photoionization detector located in the off-gas treatment line. When off-gas readings showed high VOC levels at a particular depth, circulation of the augers continued at that depth until VOC levels in the off-gas measuring system dropped. When the mixing and stripping in a zone was sufficient, the augers were allowed to penetrate to a lower depth. The method of mixing and treating the off-gas continued in the new zone at a greater depth. This process continued until the desired total depth was reached or until the augers could not penetrate any deeper because of an unknown subsurface obstruction. This SM/TESVE system was used to treat approximately 1,032 m³ (1,350 yd³) of contaminated soil before the switch from prolonged soil vapor extraction to the iron addition method was made.

6.2 ZERO-VALENT IRON INJECTION

Because of the extended time spent mixing and stripping the soil, the amount of moisture added to the soil by steam condensation was much greater than anticipated. After mixing, the soil column was very wet and resembled stiff mud. Photo K shows the muddy consistency of the soils after mixing. This high moisture content would probably have interferred with the proposed soil ventilation system to such an extent that this approach to polishing was abandoned.

The iron-injection system was developed as an alternative to the soil ventilation system. The iron treatment system was designed to work by injecting small, metallic iron particles directly into the contaminated subsurface immediately after SM/TESVE. When the iron would react with the subsurface water, condensed steam, and chlorinated organic compounds in the soil, it would dechlorinate these compounds. Originally, this treatment method was considered as one of three possible polishing steps to follow the soil mixing and steam stripping step.

During the soil treatment experiments (one of which was iron injection), it was found that the drops in residual contamination after treatment were most dramatic in iron-treated holes. The other polishing steps, including the one originally proposed for the project (i.e., soil ventilation) did not perform at the same level as iron injection. In further investigations, it was determined that iron
was highly effective, even with little or no SM/TESVE treatment. On the basis of these findings, the concept of injecting the iron into the soils after coring and mixing by means of steam stripping was abandoned in favor of injecting the iron solution into the soil during mixing. This process eliminated the need for mixing and stripping at prescribed depths and transferred the focus to coring and injecting iron into the soils as fast as possible.

The zero-valent iron-injection system and procedure used in the majority of treatment cells is described below. The coring and mixing system was used to enter and mix the soils. Once iron was included in the process, less time was spent stripping the highly contaminated zones. Iron injection was accomplished by fitting an additional fluid line into the nozzles located on top of the auger assembly.

To be injected into the soils, the iron had to be suspended in a liquid solution to facilitate pumping. The liquid solution had to have sufficient viscosity to suspend the iron particles evenly throughout the mix, but it could not be so viscous as to make pumping difficult. Guar gum, a natural substance derived from a soybean-type plant, was used to provide the required viscosity and carrying characteristics needed for this application. Guar gum is also commonly used in the environmental industry for this type of work because of its tendency to rapidly degrade in a subsurface environment. The guar gum, iron, and water were thoroughly mixed in a portable mixing plant. The mixing plant was connected to the auger assembly by flexible, steel-lined tubing leading from a grout pump located at the mixing plant to the auger assembly. As the auger assembly proceeded into the ground, personnel at the mixing plant, in radio contact with the machine operator, regulated the flow of iron
into the subsurface by opening and closing a valve between the grout pump and the line leading to
the auger assembly. This system ensured that the proper amount of iron was injected into the
subsurface as the depth of the augers increased.

The iron concentration injected into the ground amounted to approximately 1.5 to 2% by
volume of the soils mixed in each treatment column, i.e., about 0.75 to 1 ton of iron per treatment
column. Mixing and injection of iron continued to a target depth of 8 m (25 ft) bgs, or until the auger
could not penetrate any deeper because of an obstruction. The iron injection rate was regulated so
that the prescribed volume of iron (1.5–2%) was uniformly injected into the treatment column. When
the depth of mixing reached 8 m (25 ft), or an obstruction was encountered that prevented further
advancement, the auger assembly was withdrawn, and mixing was begun at the next cell.

6.3 RATIONALE FOR USING ZERO-VALENT IRON TREATMENT
DURING RATHER THAN AFTER MIXING

After studying the analytical results from the zero-valent iron polishing experiments,
Argonne personnel reviewed the costs of treating soil with iron injection. They found that the costs
would exceed budgeted costs by nearly four times if iron was injected after mixing and steam
stripping. Because the iron treatment process was more successful in treating contamination than
other processes, Argonne decided to conduct experiments on various methods that might be used to
inject iron into the ground during mixing. These experiments are summarized below.

1. In the first two experiments, ISF cored and mixed the soils in an attempt to
reach a desired depth as quickly as possible. After the desired depth was
reached, the iron solution was injected into the soils while the augers were
being removed from the ground.

2. In a third experiment, only water was used during the coring, mixing, and
injection of iron. This was done in an effort to reduce costs by eliminating the
need for steam. Penetration and mixing of the soil at this location was
extremely difficult, and penetration was stopped at 4.3 m (14 ft) bgs. ISF then
moved to an alternate location, and attempted to core and mix again without
steam. Again, penetration was difficult and stopped at 4 m (14 ft) bgs. ISF
then opted to introduce steam into the process at this location. When steam
was used, drilling conditions improved; ISF reached a total depth of 6 m
(20 ft) bgs. ISF injected a 2% by volume iron solution into the soils at this
location. ISF set up another location and cored, mixed, and injected iron (with
steam) to a depth of 6 m (20 ft) bgs. At this location, ISF injected a 1% by
volume iron solution into the soil. Analytical results from these additional
treatment columns verified the results from the original experimental cell.
3. In a fourth experiment, ISF tried a new process in which the iron was injected into the soil while coring proceeded downward until the total depth of the treatment column was reached. The augers were then removed, and thoroughly mixed soils containing the iron solution were left in the hole. ISF began this new process at a new location. During the experiment, ISF cored to a total depth of 4 m (12.5 ft) bgs before encountering an underground obstruction. It then injected a 2% iron solution into the soil. ISF then moved to another location and injected a 2% iron solution to a depth of 7 m (23 ft) bgs before hitting an obstruction. Posttreatment analytical results from this iron injection method were superior to the results obtained in the first three experiments. Treatment times were shortened from an average of 300 minutes (when the SM/TESVE method alone was used) to approximately 90 minutes (when the new simultaneous injection method was used).

On the basis of the analytical results from the soils treated in the last two iron experiments, and because less time was required to treat soils with the iron injection method used in these cells, Argonne decided to employ the method used in these last two experiments to treat the remaining soils. This method was fully deployed on January 24, 1998. Approximately 5,329 m$^3$ (6,970 yd$^3$) of soil was treated by using the SM/TESVE method with iron addition.

7 DISCUSSION

During the planning phase of this project, several remedial alternatives were evaluated as corrective actions for the contaminated soil in the 317 Area. In-situ soil treatment was preferred over the other alternatives, including off-site disposal of soil, use of institutional controls, and construction of an engineered barrier. This section discusses the advantages and limitations of deploying soil mixing coupled with iron injection to treat VOC-contaminated soils in the 317 Area at Argonne.

7.1 ADVANTAGES

7.1.1 Efficiency of Iron Addition in Removing Residual VOCs

Examination of the analytical data from the initial and final sampling of the treatment areas indicated that dramatic reductions in VOC concentrations were achieved. A comparison of figures showing field VOC screening results before and after treatment indicated that essentially the entire main hot spot was removed, thus reducing headspace VOC levels by 90 to 99% in most locations treated.
In many cases, the SM/TESVE process, augmented by iron addition, attained efficiencies of more than 95% in removing specific chlorinated organic compounds. Despite this high degree of VOC removal, cleanup objectives specified by the Illinois Environmental Protection Agency (IEPA) Tiered Approach to Corrective Action Objectives (TACO) were not met because of the large initial concentrations of contaminants in the soils. To achieve these levels, a removal efficiency of more than 99.9% would have been required.

The site will be monitored from time to time in the next few years to determine whether residual concentrations will decrease further over time; in the meantime, it appears that additional corrective measures will be required before the corrective action process for this solid waste management unit (SWMU) is completed. Current corrective action activities, including monitoring of the groundwater downgradient of the former French Drain treatment area and operation of the groundwater extraction system adjacent to the Argonne property line, will continue for the near future. Other future actions will concentrate on preventing groundwater migration through the contaminated areas and on further reducing or containing VOC concentrations in the source area. These actions will be implemented to prevent further contaminant spread by release to groundwater and surface water.

7.1.2 Comparability of In-Situ Soil Treatment with Argonne Operations

As discussed above, the 317 Area is an active hazardous and radioactive waste processing and storage area. As such, the area in the immediate vicinity of the treatment area is frequently used by Argonne Waste Management personnel and vehicles during waste packaging activities. Of the remedial alternatives considered, in-situ soil treatment was the least disruptive to critical Argonne operations in the 317 Area. Excavation and off-site disposal of contaminated soils (another alternative considered by Argonne), while completely removing the risk posed by the contaminated soils, would have required a large, deep excavation in the 317 Area and numerous soil shipments from the site. The in-situ soil treatment discussed above occurred concurrently with routine Argonne Waste Management operations.

7.1.3 Comparability of In-Situ Soil Treatment with Existing Argonne RCRA Permits

When the work plan for this project was submitted to the IEPA, Argonne had not yet received the final RCRA Part B Permit. This project was intended to be an interim action to reduce the source of contaminants in the 317 Area French Drain. Because the soil was not removed, it was not considered waste. As such, in-situ treatment of the soil was not subject to RCRA permitting requirements, and a RCRA hazardous waste treatment permit was therefore not required. The treatment process, however, generated off-gases that were treated in the off-gas treatment system.
Construction and operation of this system was included in a joint construction operating permit issued by the IEPA well before field activities began.

7.1.4 Relatively Low Cost of In-Situ Soil Treatment

The primary objective of the in-situ soil treatment in the 317 Area was to remove most of the contaminants from the soils associated with the French Drain, which would, in turn, reduce the potential source of contamination of groundwater and surface water in the area. The in-situ soil treatment discussed here, coupled with iron injection as a polishing step, was designed to be an interim action conducted in anticipation of follow-up actions such as a future groundwater extraction system and isolation or additional treatment actions to prevent the release of residual materials to the groundwater. The in-situ soil treatment method was determined to be the most cost-effective way to achieve these project goals. Excavation and off-site disposal of contaminated soil, considered as a remedial alternative, were found to be cost-prohibitive because of the assumed presence of added radioactivity in the soil, which would cause the soil to be classified as a mixed waste. Construction of engineered barriers, also considered as a remedial alternative, was also determined to be cost-prohibitive because of its long-term operations, maintenance, and monitoring costs.

7.2 LIMITATIONS

7.2.1 Damage to Dual Auger System from Subsurface Geologic Conditions

At the beginning of the field work, a large boulder (about 9 ft long, 7 ft high, and 6 ft wide) destroyed the augers. Their damage resulted in project down-time of about five weeks. Moreover, daily mixing of tight, clay soils resulted in numerous, shorter periods of down-time. Routine soil mixing damaged gear chains, auger motors, and gears. Repair times were long since, in most instances, replacement parts had to be specially fabricated for the soil mixing equipment because it was unique.

When the augers damaged by the boulder described above were being repaired, a hydraulic "shear pin" system was put into place. This was designed to cut power from the auger drive train whenever an obstruction or tough soil conditions were encountered. This system tended to slow the work, because when the rate of penetration was increased, pressures in the system approached the limit for the shear pin system. Although it resulted in a 50% reduction in productivity, the shear pin system did prevent severe damage on several occasions, and some repair down-time was avoided. However, the time originally budgeted for this work doubled.
Penetration was also hampered by safeguards placed into the auger system drive train to prevent damage to the augers and drive train system itself when subsurface obstructions (predominantly boulders) were encountered. In addition, because of the characteristics of the clay, penetration into the clay and mixing required at least two to three hours.

7.2.2 Damage to Iron Injection Lines

During deployment of the iron polishing treatment, damage to rubber and steel injection lines occurred routinely. The iron slurry was pumped through rubber lines to the swivels at the top of the Kelly bars, where it was combined with pressurized steam and injected into the subsurface soils. Because of the abrasive nature of the iron slurry, holes formed in the rubber line segment after the slurry mixed with the pressurized steam. Down-times for repair of these rubber lines typically lasted 1 to 2 hours. Less frequently, the schedule-80 steel line delivering the combined steam/air/iron slurry treatment media to the augers developed a hole from the iron. These repairs lasted slightly longer, about 2 to 3 hours. As a result of this damage, the iron lines were reconfigured to reduce the amount of pressure exerted by the pressurized steam and iron solution on the line’s interior walls. This reconfiguration reduced the number of damaged lines.

7.2.3 Excessive Mud and Runoff

The SM/TESVE soil treatment method, coupled with iron injection, required large amounts of water to be introduced into the subsurface. Areas undergoing soil mixing became completely saturated, and consequently, the subsurface became very soft. As the augers were withdrawn from a mixed soil column, large amounts of mud were released from the auger shroud, and they spilled over the ground surface. This condition affected related project activities in several ways, as discussed below.

Before an area of soil was mixed, surface gravels that might contain radioactive particles were removed to prevent their entrainment into the subsurface. The low viscosity of the soils after they were mixed caused mud to spill over to untreated areas, hampering gravel removal.

As soil treatment proceeded, a large area of treated soil — now mud — became exposed to erosion. This situation was aggravated by the fact that this project took place in the winter, when the precipitation level is high. An elaborate erosion control system was therefore used to contain all silt-laden surface runoff. About 77,000 gallons of runoff were captured by the system and disposed of in Argonne’s Laboratory Wastewater Treatment Plant, via a manhole adjacent to the treatment area. In the absence of the plant, this runoff would have been contained and shipped off-site for disposal, which would have added significantly to the overall project cost.
The soil treatment process resulted in an area of mud averaging about 20 ft deep and about 100 by 200 ft in area. The soft nature of the soil following treatment precluded any routine waste management activities from taking place in this area. In fact, only small front-end loaders, such as those used during site restoration work, were able to drive on the treated soil. Substantial settling has to occur before the area can be opened for unrestricted use.

The extensive area of mud also created health and safety concerns for project personnel. The mud could not support the weight of a person. In addition, mud spillover obscured the boundaries between areas of mixed and unmixed soil. To mitigate this hazard, the treatment area was cordoned off with orange snow fencing and posted with hazard signs. Another hazard included the generation of hydrogen-rich gas (greater than 90% hydrogen in some cases) from the iron-chlorinated solvent reaction. This gas easily bubbled through the mud and was released to the atmosphere, where it could potentially affect nearby workers. Efforts to mitigate this potential hazard included posting of warning signs about flammable gas around the treatment area and regular monitoring of the lower explosive limit in the work area atmosphere. Monitoring did not reveal any health hazard.

8 SUMMARY

A thermally enhanced soil mixing process was used to remediate the 317 Area French Drain at Argonne, which is contaminated with VOCs. After the initial soil treatment step, an iron slurry, consisting of up to 50% iron particles in water with guar gum added as a thickening agent, was injected and mixed into the soil by mixing equipment. This soil treatment method reduced VOC concentrations in soil by more than 80%.

Deployment of the soil mixing/thermally enhanced soil vapor extraction (SM/TESVE) system, followed by iron injection, required the construction of an extensive infrastructure. This consisted of a soil mixing and off-gas treatment train, boiler plant, and iron slurry batch plant immediately adjacent to the treatment area. Other support facilities constructed for this project included a decontamination area, sampling equipment tent, tool shed, and a bermed diesel fuel tank area.

The in-situ SM/TESVE process, augmented by iron addition, represented the most cost-effective and least disruptive method of achieving project goals, as stated in the IEPA approved work plan for this project. This treatment system had several advantages over other remedial alternatives that were considered. It attained removal efficiencies of more than 95% for several chlorinated organic compounds. Deployment of this treatment system in an actively used facility did not interfere with site Waste Management operations. Not only did the system achieve project goals of significant source reduction, but it was also determined to be the most cost-effective treatment method. Issues related to permitting and the costs associated with off-site disposal of the soil and
long-term operations and maintenance costs of engineered barriers precluded other alternatives from being implemented at this site.

Limitations of this treatment method were restricted to the physical characteristics of the soil matrix following treatment and the local subsurface geologic conditions. The large amounts of mud generated during this project required an extensive runoff control system downgradient of the treatment area and posed health and safety risks to project workers. Very tight clays and the presence of large boulders in the subsurface resulted in several periods of project down-time to repair damaged equipment. Efforts to reduce the risk of equipment damage from difficult geologic conditions reduced the number of incidences of damage but lengthened the time needed to treat contaminated soils.

9 REFERENCES


