# Organic substances in the subsurface: Delineation, migration, and remediation\*

# Ishwar Murarka<sup>a</sup>, Edward Neuhauser<sup>b</sup>, Michael Sherman<sup>b</sup>, Barbara B. Taylor<sup>c</sup> and David M. Mauro<sup>c</sup>, John Ripp<sup>d</sup> and Terry Taylor<sup>d</sup>

<sup>a</sup> Land and Water Quality Studies, Electric Power Research Institute, 3412 Hillview Ave., Palo Alto, CA 94303, (USA)

<sup>b</sup> Niagara Mohawk Power Company, Syracuse, NY (USA)

<sup>e</sup> META Environmental, Inc., 49 Clarendon St., Watertown, MA 02172 (USA)

<sup>d</sup> Atlantic Environmental Services, Inc., Colchester, CT (USA)

#### Abstract

The Electric Power Research Institute (EPRI) and Niagara Mohawk Power Corporation are sponsoring a research program concerning the release, transformation, and migration of organic compounds at a site where coal tar from former manufactured gas plant (MGP) operations was disposed nearly thirty years ago. Work at this site, referred to as EBOS Site 24, has included: determination of the location and chemical content of the tarry source material, delineation of the groundwater contaminant plume, evaluation and implementation of innovative methods for sampling and analysis, and the remediation and restoration of the site. The results of the initial phase of research provided several important insights into the mechanisms of contaminant release and migration. For example, the shape of the groundwater contaminant plume at EBOS Site 24 was dominated by longitudinal advection with little contribution from transverse or vertical dispersion. A long-term monitoring program at EBOS Site 24 was initiated prior to the removal of the source material. The results of the baseline groundwater monitoring along the plume centerline were similar to the values predicted using EPRI'S MYGRT<sup>TM</sup> model for migration of contaminants. After the baseline monitoring was completed, all of the tarry source material was removed in 1991 and used in the production of asphalt and portland cement. The groundwater monitoring program will continue for several years and the field results generated during this time will be used to evaluate and/or calibrate the  $MYGRT^{TM}$  model.

## Introduction

In recent years, increasing concern has been expressed about the potential threats to human health and the environment from organic substances produced as by-products of former manufactured gas plant (MGP) operations,

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Correspondence to: Dr. B. Taylor, META Environmental, Inc., 49 Clarendon St., Watertown, MA 02172, (USA).

such as the mononuclear aromatic hydrocarbons (MAHs) and polynuclear aromatic hydrocarbons (PAHs) in coal tar. Although the presence of coal tar, and other process residues, at sites where former MGP plants were located has been known for some time, few sites have been investigated thoroughly and even fewer have been remediated.

Starting in 1987, the Electric Power Research Institute (EPRI) initiated a major research effort under the Environmental Behavior of Organic Substances (EBOS) program to help understand what happens to the organic wastes at MGP sites over time as some of their constituents are released to the environment, and what happens to the contamination in the downgradient aquifer after the wastes are removed from the site.

The focus of the EPRI-sponsored research began with a series of field efforts directed toward developing insights concerning the natural processes by which organic substances in tarry materials are released to, transformed in, and move through various environmental matrices. The understanding of these processes is fundamental to selecting the appropriate means of remediation and for determining the efficacy of that remediation. As a better understanding of these processes has evolved, part of the more recent research has focussed on predicting how well remediation efforts will work (based on computer-generated migration simulations using EPRI's MYGRT<sup>TM</sup> software) and determining how good those predictions are.

The EPRI field work started in late 1987 at a site in the Northeast, referred to as EBOS Site 24, with the assistance and cooperation of the owner utility, Niagara Mohawk Power Corporation. First, preliminary field work was carried out to test both conventional and innovative methods for sampling and analysis of soils and groundwaters. When the methods were well-established, a series of field studies were implemented to locate the tarry material and define the groundwater contaminant plume both aerially and vertically. After the location of the source area was defined, Niagara Mohawk contracted to have all of the known tarry material removed from the site. Finally, EPRI, in cooperation with Niagara Mohawk, recently initiated a long-term monitoring program which will examine the groundwater quality, biotransformation, and plume dissipation for the next ten years to assess both the efficacy of the remedial source removal and the ability of MYGRT<sup>TM</sup> to predict plume dissipation.

This paper provides a summary of a portion of the work carried out by EPRI and utility contractors at EBOS Site 24 and the major findings of that work. It starts with an overview of the MGP problem and why such research efforts are needed.

## Background

From the early 1800s through the 1950s, the production of gas using coal, coke, or oil resulted in the generation of large volumes of various process by-

products at MGP operations throughout the United States, as shown in Table 1 [1-4]. As seen in this table, tars and tarry materials were a major part of the MGP waste products. Often, the standard practice of the day was to leave these tarry residues in holders, trenches, ponds, or pits on-site or to move them to off-site locations for disposal [5,6].

Gas production at MGP sites was phased out rapidly after World War II with the development of natural gas pipelines. The tarry remnants of MGP operations remained, even as the former MGP sites were converted for various utility purposes, such as utility maintenance areas or substations, demolished, abandoned, or sold to others. In recent years, however, the leftover wastes generated from the old MGP processes have come under increased scrutiny. This is partly a result of increased awareness of potential problems, as well as an increase in the sale or redevelopment of former MGP sites [3].

Since the concern about potential hazards from MGP wastes has surfaced only in the past several years, little has been known about the environmental fate of MGP-derived tarry materials. Attempting to study the fate of these materials is difficult since the coal or oil tar resulting from different processes can be physically or chemically different [6,7]. Furthermore, where and how the tars have been left for decades can impact their current state. For example, tar left in a deep holder that was demolished and covered over with rubble, fill, and asphalt can be significantly different from a similar tar that was placed in a shallow trench and left exposed to the environment. This is because from the moment of disposal, a variety of physical, chemical, and microbial processes affect the way in which the coal tar constituents are released to, transformed in, and transported through the environment. Natural processes such as dissolution, volatilization, adsorption, microbial degradation, dispersion, and advection combine to determine the ultimate environmental fate of tarry material constituents [1,8]. Table 2 illustrates some of the differences among the

#### TABLE 1

By-products	Processes			
	Coal carbonization	Carburetted water gas	Oil gas	
Petroleum oils			X	
Tars	×	×	×	
Spent oxides	×	×	×	
Ash/clinker	×	×	×	
Sludges/emulsions	×	×	×	
Ammonia liquor	×			
Lampback			×	

Typical MGP process by-products

Characteristic	Compound groups				
	MAHs	PAHs	Phenols		
Volatility	High	Low	Low		
Solubility	Medium	Low	High		
Biodegradability	Medium	Low	High		
Adsorption tendency	Low	High	Low		
Mobility	High	Low	High		

#### Comparison of MGP organic compound characteristics<sup>a</sup>

<sup>a</sup>There is significant variability in characteristics among individual compounds, such as the PAHs

environmental fate characteristics of three major types of MGP-related organic compounds, MAHs, PAHs, and phenolic compounds.

Understanding how natural processes play a part in the ultimate fate of MGP tar is very difficult. It is especially complicated because tars are very complex mixtures of hundreds, if not thousands, of different organic compounds. These compounds each have their own physical, chemical, and biological properties, which often are very different from each other. For example, one of the most common constituents in MGP tars is naphthalene, a two-ring PAH. It has a pure component, aqueous solubility of about 31 mg/L. This is dramatically contrasted with benzo(a)pyrene, a five ring PAH, which has a pure component, aqueous solubility of only about 0.004 mg/L [9].

Although some properties of the individual tar constituents, such as volatilization and dissolution, have been studied in laboratory experiments [9], the estimation of individual constituent parameters does not accurately describe how the tar mixture will act in the environment. Again, using the example of naphthalene, while the pure component solubility is about 31 mg/L, the aqueous solubility of the naphthalene in some MGP tars has been determined to be about 11 mg/L in laboratory experiments and about 13 mg/L based on Raoult's Law predictions [10].

Clearly, although laboratory experiments on single tar compounds or model simulation concerning the rate of environmental processes in nature provide some important information, field-scale research at MGP sites can help fill in the information gap. This is particularly important in determining appropriate remedial alternatives and predicting how well they will work. Thus, EPRI has emphasized field-scale research at MGP sites.

#### EBOS Site 24

The EPRI research at EBOS Site 24 began in late 1987, about twenty-five to thirty years after coal tar was removed from an MGP holder, trucked to a

TABLE 2

rural area a short distance away, placed in a large trench at the side of a country road, and covered over with sand. The amount of tar deposited in the trench has been estimated to be anywhere between about 4,000 and 16,000 gallons, although no records of the actual volume disposed were found.

The tar was left forgotten in the covered over trench and indigenous vegetation grew back on top of the sand fill, giving no indication that this location was anything other than a pleasant open area covered with wild strawberry plants, small shrubs, and grasses. It was not until the early 1980s when a utility worker detected coal tar-derived organic compounds several hundred meters downgradient from the tar that an investigation of the site was initiated.

With support from and in cooperation with Niagara Mohawk, EPRI began its MGP site research at EBOS Site 24. The goals of the research effort were to examine and evaluate both conventional and innovative methods for sampling and analysis at former MGP sites, to help develop insights into the ultimate fate of tarry materials in the environment, and to determine the efficacy of remedial actions.

EBOS Site 24 was an ideal site to use for initial research on environmental processes as they control the release and migration of organic compounds from MGP tars. First, the site was located in a rural area and, other than a country road and electrical transmission lines which transverse through one corner of the site, there were no buildings or other structures to modify subsurface characteristics or influence the natural movement of groundwater. Second, the site had only a single tar source, thereby eliminating the problem of multiple sources and multiple plumes. Third, although there were discontinuous clay lenses in the saturated zone, the aquifer was consistently sandy throughout the site and was composed of coarse to fine silty sands, with a confining clay layer located below the silty sands at a shallow depth of about 22 feet (6.6 m). Finally, the sandy soil contained little organic carbon, indicating that adsorption reactions would not be dominant for chemical attenuation at this site.

### Location and volume estimation of the source tar

One of the first steps in the site investigation at EBOS Site 24 was to accurately determine where the tarry source was located. To do this, a square site grid was established at the site with grid points at 50 foot intervals in the source area and 100 foot intervals for the rest of the site. This is shown in Fig. 1. The grid was extended over the entire site and was used throughout the research effort to pinpoint sampling, piezometer, multilevel sampler, and monitoring well locations.

Anecdotal information from a former utility employee indicated that the coal tar trench was located north of the road in a clearing to the west of a small stand of trees. Thus, initial efforts to locate the source area focussed on that part of the clearing. It was soon discovered that anecdotal accounts often are



Fig. 1. Site grid for EBOS Site 24.

only partly correct. In this case, the tar indeed was found north of the road, but to the east — rather than the west — of the small stand of trees.

The determination of the areal tar boundaries and the estimation of the tar and tar-contaminated soil volume was made later in the project by a series of rapid borings in the vicinity of the source. The areal extent of the tar and contaminated soils is shown in Fig. 2. Based on the results of the borings, the volume of tar and tar-contaminated soils was estimated to be approximately 8,000 cubic yards (6100 m<sup>3</sup>), with most of the tar and highly contaminated material located at or below the water table at depths from about 7 to 22 feet.

## **Contaminant plume location and definition**

The data from piezometers and groundwater wells installed at various locations across the site showed that the groundwater flowed from the source in a southeasterly direction, under the road, and down to a downgradient seep area more than 300 yards away (Fig. 3). This area consisted of numerous small trickling seeps that converged to a small stream that eventually reached a ma-



Fig. 2. Location of tarry source material at EBOS Site 24.



Fig. 3. Groundwater contours.

jor river. Results of a study concerning the seep area showed the presence of naphthalene in the seep sediments, but no PAHs were detected in the stream water entering the river.

One of the major developments for field sampling at MGP sites from this research was the method for determining the lateral and vertical placement of groundwater monitoring wells directly within the plume or at the plume edges. This method used naphthalene as an indicator compound because it was the PAH found in the highest concentration in the source tar and because it is the most soluble and has the least potential for soil adsorption of all of the PAH compounds detected. Thus, it was the most abundant and mobile of the coal tar-derived PAHs for the EBOS Site 24 source tar.

The method for determining where groundwater wells should be placed (both laterally and vertically) and what screen length should be used had several steps. First a series of transects were established to cut across the site in such a way so that they were approximately perpendicular to the expected direction of the groundwater plume (Fig. 4). As a result of the placement of some monitoring wells during the initial reconnaissance phase of the work and the impediment of heavily wooded areas, not all of the transects were parallel.

Second, several borings were made along each transect, starting from the edges and working toward what was thought to be the centerline of the plume. During the boring process, soil samples were collected at appropriate depths and taken to the field laboratory where they were extracted and analyzed. (A



Fig. 4. EBOS Site 24 transects.

field method of soil and groundwater extraction and analysis for PAH compounds was developed during this research effort. It consisted of a micro extraction technique followed by gas chromatography with flame ionization detection (GC/FID). The results of this method compared favorably with conventional Soxhlet extraction followed by gas chromatography with mass spectrometric detection (GC/MS)). Next, the results of the initial borings were used to determine where future borings would be made or from what depth samples should be collected for analysis.

Finally, once all of the data were examined, the areal location of the boring with the highest concentration of naphthalene along each transect was selected for the approximate well location. Then, by studying the vertical distribution of naphthalene concentrations in that boring a determination was made concerning what depth and screen length would place the monitoring well in the zone with the highest naphthalene concentration. Typically, two or three relatively short well screens (usually 2 or 2.5 feet in length) were placed at a single areal location determined to be at or near the plume centerline. Fringe wells were also placed at the edges of the plume in order to clearly delineate the outer boundaries of the plume.

This method of well placement proved to be an excellent technique for placing monitoring wells where the highest concentration of PAHs were found in the plume or at the plume boundaries with a fairly high degree of certainty. However, since soil analysis results were needed within a day or so of collection, it could only be used effectively in conjunction with a rapid turnaround field laboratory.

As an example of how this method worked, Fig. 5 shows the maximum concentration of naphthalene detected in the soils at five locations along Transect C. The place where the highest naphthalene concentration was detected was designated the centerline for Transect C and the soil results from different depths at that location were reviewed to determine the best well depths and screen lengths to use for the monitoring wells. Figure 6 shows how the naph-



Fig. 5. Maximum naphthalene concentrations in soils across Transect C.



Fig. 6. Soil ( $\bigcirc$ ) and groundwater ( $\blacktriangle$ ) naphthalene concentrations at the centerpoint of Transect C. ( $\Box$ ) Well screen.



Fig. 7. Different groundwater well configurations. (a) Conventional placement of two wells located at the water table and confining layer to detect the presence of L-NAPL and D-NAPL, (b) a single well with a long well screen, (c) a single well with a short screen placed in the contaminated zone, and (d) a multilevel sampler ports at specific intervals.

thalene concentrations in the soil changed with depth, the placement of the three groundwater wells, and the groundwater naphthalene concentrations found in the three wells. As shown in this figure, the vertical extent of the groundwater plume is very narrow and that the well with the highest naphthalene concentration is the middle well, between the water table and the confining layer.

One of the interesting results from this method was that standard well placement techniques may not always result in proper well placement. For example, often wells are placed so that they are screened at the water table to detect the presence of lighter-than-water non-aqueous phase liquids (L-NAPL) or at the confining layer (or bedrock) to detect the presence of denser-than-water nonaqueous phase liquids (D-NAPL). However, for most locations at EBOS Site 24, such as Transect C illustrated in Fig. 6, the placement of wells at the water table and confining layer would have resulted in groundwater samples with much lower concentrations of naphthalene than in the plume. This is because most of the highest naphthalene concentrations were observed at locations between the water table and the confining layer (except for the near source locations). Wells placed at the two extreme locations or long well screens would have significantly underestimated the contamination present in the plume, this is shown schematically in Fig. 7.

## **Investigation conclusions**

The results of the field investigation at EBOS Site 24 showed that a fairly narrow plume of tar constituents moved away from the source area and that naphthalene moved the furthest distance of any of the PAH compounds studied. Figure 8 depicts the areal extent of the groundwater naphthalene plume across the site. Other PAH compounds did not appear to move through the saturated zone as readily as naphthalene and some did not show any significant movement away from the near source area. This is shown in Fig. 9. In this figure, the concentrations of naphthalene, acenaphthylene, and anthracene in groundwater are shown as a function of distance from the source. It is easily



Fig. 8. Aereal extent of the groundwater naphthalene plume.



Fig. 9. Groundwater concentrations of naphthalene  $(\bigcirc)$ , acenaphthylene  $(\triangle)$ , and anthracene  $(\Box)$  at wells in the plume.

seen that the high molecular weight PAHs, such as anthracene and benzo[a]pyrene, were not observed at any significant distance away from the source.

At EBOS Site 24, it was apparent that the primary release mode of coal tar constituents to the environment was by dissolution. Thus, the solubility of a particular constituent dictated how much of that compound was allowed to move with the groundwater. Once dissolved, the material moved along with the groundwater flow in an advective manner with very little vertical or transverse dispersion. Although the contaminant plume was near the water table in the near source area, the plume dropped gradually as it moved downgradient and tended to be situated in similar soils located above the clay confining layer, as shown in Fig. 10. And, since this site contained very little organic carbon, adsorption did not appear to play a major role in retarding the movement of the coal tar constituents with the groundwater. Furthermore, there was no evidence of L-NAPL or D-NAPL at any locations away from the immediate source area.

One process which may be important with respect to the fate of some coal tar constituents is subsurface microbial transformation. Research performed at this site by scientists from Cornell University showed that there were indigenous, PAH-degrading, microorganisms in the constituent plume (11). The researchers collected soil samples from locations both in contaminated areas and an uncontaminated background area and from the saturated and unsaturated zones. They then measured the potential of the indigenous organisms to degrade naphthalene and phenanthrene. The results showed that mineralization of naphthalene and phenanthrene occurred only in the samples collected from within the contaminant plume. No such mineralization was observed for the soils from the on-site background location.



Fig. 10. Location of detectable naphthalene concentrations in soils downgradient from the source.

#### Source removal

In 1991, Niagara Mohawk contracted with Atlantic Environmental Services, Inc., to oversee the removal of all of the source tars and tarry soils, as well as the restoration of the source area. This work began in May 1991 with the site preparation. First, the entire site was secured using fencing and a locked access gate. To enclose the entire work area, the country road adjacent to the tar trench had to be closed for the duration of the site work and all traffic had to be rerouted from the area.

Next, plastic liners were located near the excavation area so that contaminated soil removed from the source area could be stockpiled until loaded into trucks for transport. In addition, a plastic-lined wastewater pond was constructed to hold all of the water generated during the excavation dewatering. Then, sheetpiles were driven to a depth of about 30 feet, well below the known confining layer, to completely surround the area where tar and tarry soils were located and to form a cofferdam. A schematic drawing of the construction site layout and the location of soil samples is shown in Fig. 11.

Soils were removed from the excavation area using backhoes and were temporarily placed on the stockpile until they were placed in trucks and transported off-site. When the excavation reached the water table, water was pumped from the cofferdam area to the lined wastewater pond.

A total of approximately 9,400 cubic yards (15,000 tons) of tarry soils, tarcontaminated soils, and adjacent clean soils were excavated from the source area. The excavated soils were tested several times and were found not to exhibit any hazard characteristic as defined by the EPA, including: toxicity, ignitability, reactivity, or corrosivity. As a result, the excavated soils could be



Fig. 11. Remedial construction layout and sample location.

used in processes to form asphalt or portland cement and would not be required to undergo treatment and/or disposal at a hazardous waste facility.

Two facilities did trial studies using the contaminated soils to produce portland cement and one facility did a trial study using the soils to produce hot batch asphalt. All three facilities used thermal desorption techniques with rotary kiln technologies, all three performed the trial burns satisfactorily, all passed audits by the contractor, and all had secure, covered, on-site storage for the contaminated soils. Based on total costs, however, one facility, the asphalt batching plant, was used to treat most of the soils (about 11,500 tons) at a cost of about \$106/ton. One of the facilities generating portland cement was used as a secondary and backup treatment plant and processed about 3,600 tons of contaminated soil for about \$172/ton. The other facility producing portland cement was used only for the trial burn study and processed about 40 tons at a cost of \$210/ton.

The wastewater generated during the excavation dewatering was tested and

found acceptable for the local wastewater treatment plant without pre-treatment. Thus, the approximately 737,000 gallons (2800 m<sup>3</sup>) of water collected during the remedial work were removed from the site and taken to the wastewater treatment facility for disposal.

By mid-October of 1991, all of the contaminated soils, water, and liners had been removed from the site. After removal of the tar and tar-contaminated soils, the excavated hole was filled with clean native sand and clean soil resulting from excavation activities in areas other than the contaminated location. The sheetpiles were removed, decontaminated, and taken from the site. The EPRI research wells and piezometers which were destroyed during the excavation process were replaced. The disturbed area was leveled, and the ground was fertilized and seeded.

#### Long-term monitoring

Two rounds of groundwater monitoring were completed in 1989 and 1990 prior to the removal of the source in 1991. The naphthalene results for this baseline monitoring program are shown in Fig. 12. Note how the concentration of naphthalene at Transect A in 1990 is much less than it was during the monitoring from 1989. The reason for this change is that the water table level fell so much between 1989 and 1990 that the well with the highest naphthalene concentration at Transect A could not be sampled in 1990 because it was dry. Thus, data from another well at that location had to be substituted in the 1990 study. The very low water table level also might have impacted the well at Transect B, to a lesser degree.

Figure 13 shows the mean naphthalene concentrations at the plume center-



Fig. 12. Naphthalene concentrations at the groundwater plume centerline prior to source removal.  $(\Box)$  September 1989,  $(\nabla)$  June 1990, and  $(\bigcirc)$  mean concentration prior to source removal.



Fig. 13. Measured mean (O) and  $MYGRT^{TM}$  predicted ( $\diamondsuit$ ) naphthalene concentrations along the plume centerline.



Fig. 14. Naphthalene mean concentrations along the plume centerline prior to  $(\bigcirc)$  and after  $(\triangle)$  source removal.

line prior to the source removal along with the predicted naphthalene concentrations from the EPRI MYGRT<sup>TM</sup> Code 2.0 model. MYGRT<sup>TM</sup> is a user-friendly IBM PC code designed to evaluate and predict the migration of organic and inorganic chemicals in groundwater [12]. As shown in Fig. 13, the MYGRT<sup>TM</sup> prediction of naphthalene concentrations at EBOS Site 24 was very close to the measured concentrations prior to source removal. In future studies,  $MYGRT^{TM}$  will be used to predict the dissipation of the contaminant plume over the next ten years. During this time, groundwater samples will be collected, analyzed, and the results of these samples will be used to calibrate and/or verify MYGRT<sup>TM</sup> predictions.

The first round of sampling after removal of the source material was com-

pleted in November 1991. The naphthalene results for this sampling event are shown in Fig. 14.

## Summary

The results of the EPRI and Niagara Mohawk Power Corporation work at EBOS Site 24 have provided considerable insights into the mechanisms for release, migration, and transformation of MGP tar constituents in the environment. Future work at the site will continue to generate data designated to help understand these mechanisms and to understand the impact of remedial source removal.

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