
SITE CLOSURE USING MONITORED NATURAL ATTENUATION AS FOLLOW UP TO AN ACTIVE SOURCE REMEDIATION IN ITALY

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SUMMARY

A multi-phase remediation project was applied at an operating manufacturing facility located in an environmentally sensitive area in Northern Italy. A soil gas investigation was first completed to delineate the source of contamination present in soils. The remediation project, approved by the Italian Authorities, included an active phase of remediation to remove the source of contamination followed by Monitored Natural Attenuation (MNA) to achieve site closure objectives. An air sparging / soil vapour extraction (AS/SVE) system was designed and installed inside and outside the facility after pilot testing. The system, intended to be operated to the limit of its technology, was installed to remove trichloroethene and associated degradation products from the vadose zone and shallow groundwater. Once the system monitoring parameters indicated asymptotic trends it was possible to move towards the natural attenuation phase of the project. The on-going MNA monitoring program is intended to collect necessary bioparameters recommended to control and evaluate biodegradation processes at the site. The active remediation phase successfully removed a significant mass of the target compounds and groundwater concentrations lowered by as much as 75 percent at some locations. Groundwater monitoring results obtained during the MNA phase indicated a global reduction of the concentrations of the compounds of concern. The MNA data suggests favourable conditions for biodegradation. The project illustrates the benefits of an integrated remediation strategy where MNA is being applied after mass removal to satisfy remedial objectives and regulatory requirements cost effectively.

INTRODUCTION

A multi-phase remediation project was designed and negotiated with the authorities to address soil and groundwater constituents at an operating manufacturing facility located in Northern Italy. The constituents to be remediated consisted of volatile organic compounds (VOCs), primarily trichloroethene (TCE) and its associated degradation

products, *cis*-1, 2-dichloroethene (DCE) and vinyl chloride. Phase 1 included an air sparging / soil vapour extraction (AS/SVE) system installation and Phase 2 consisted of monitored natural attenuation (MNA). The original source of VOCs was a release from a historical degreasing operation in the early 1970s.

The manufacturing facility occupies approximately 30,000 square meters (m²), of which approximately 90 percent of the surface is paved or covered with buildings. A layout of the facility is shown in Figure 1. Compounds of concern have been detected in shallow groundwater at concentrations as high as 258 micrograms per litre (µg/l). DCE and vinyl chloride have been detected at the highest concentrations, as high as 130 µg/l and 73 µg/l, respectively. The vadose zone consists of fill overlying heterogeneous clays and alluvial silts. Shallow groundwater is encountered at approximately 6 to 8 meters below ground surface (bgs). The surficial water-bearing zone consists of 5 m of sand and gravel; the underlying aquitard consists of 9 m of clayey silt; groundwater constituents have not been detected in deeper groundwater. Data collected indicates that shallow groundwater flow direction in the vicinity of the site is generally towards the Southeast.

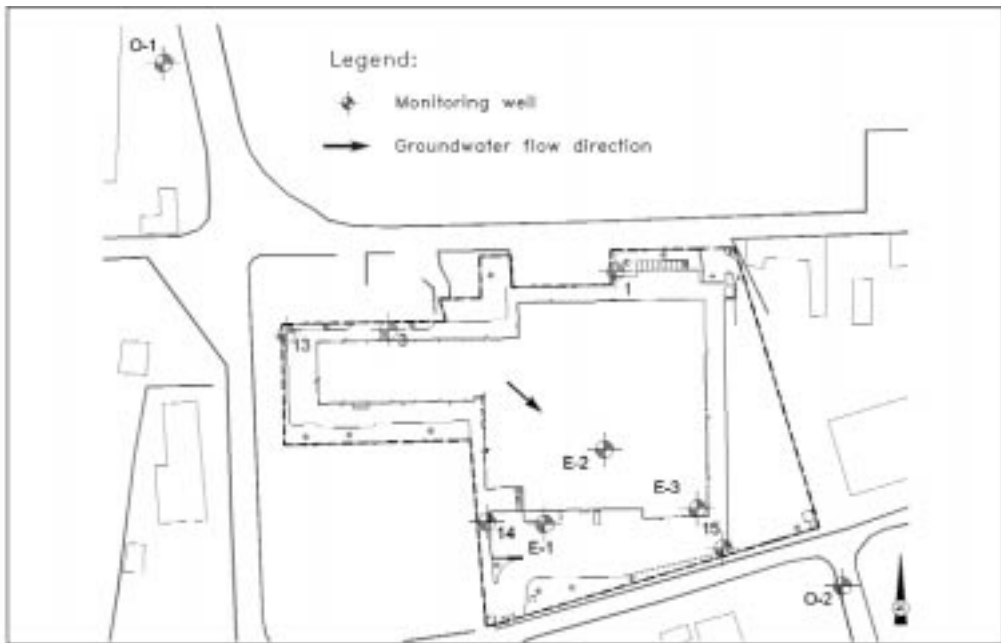


Figure 1. Facility layout and monitoring wells location.

The remedial objectives are defined by Italian National drinking water standards, which establish a limit of 30 µg/l for total halogenated hydrocarbons (including chlorinated hydrocarbons). The AS/SVE system was intended to recover as much VOC mass as much as possible, operating the system to the limits of its technology, as well as promoting aerobic biodegradation of volatile organic constituents. The subsequent MNA program is

intended to collect chemical, biological and geochemical data in order to control and evaluate biodegradation processes at the site with particular attention to any potential limiting factor.

Potential soil and groundwater remedial alternatives were evaluated for the site. AS/SVE was selected as the recommended Phase I remedial alternative for the following reasons: 1) Would address soil and groundwater in areas of highest concentration, 2) Contaminants would be either removed or destroyed, 3) High probability of success, 4) Provide significant remediation in short time frame (1 to 3 years), 5) Relatively easy to implement, and 6) Cost effectiveness and would cause limited disturbance to manufacturing operations.

AS/SVE PILOT TEST AND INSTALLATION

A pilot test of AS/SVE was conducted as the first step of implementing the technology full-scale. Costs of the pilot test were minimised by conducting a series of brief tests to confirm the effectiveness of the technology and provide order-of-magnitude design criteria for operating parameters such as vapour extraction rates, air injection rates, and induced impacts on soil vapour and groundwater in the subsurface. Extensive testing was not performed. As a trade-off, optimisation of the system would be performed during start-up of full-scale if the pilot test indicated AS/SVE was effective. Extensive testing was also not performed based on published data suggesting that air sparging systems can often be designed with minimal pilot testing using default design conditions (Gillespie, et al., 1999). The pilot testing indicated that AS/SVE could be effectively applied at the site.

Based on the pilot test results, an AS/SVE system was installed with a turnkey design/build approach using a minimal design document and close co-ordination of design engineers and construction staff in the field during installation. Fourteen SVE wells and 18 AS wells were installed. The AS/SVE system was started up in November 1999 and had been operated continuously using all 14 vapour extraction wells and 18 air sparge points for the following 12 months. An 18-hp blower was installed for the SVE system with an expectation that it would extract about 420 cfm vapour from the extraction wells. Following optimisation during start-up, the SVE system stabilised at extraction of 414 cfm vapour at an applied vacuum of 21 inches of water. At these conditions, the SVE system was operating at 98 percent of the expected design rate. Flows have remained steady at these levels within 10 percent. A 15-hp blower was installed for the AS system with an expectation that it would inject about 150 cfm air into the sparge points. Following optimisation during start-up, the AS system stabilised at an injection rate of approximately 132 cfm, or 87 percent of the expected design rate. During operation, flow rates have decreased as the injection pressure has been lowered.

AS/SVE RESULTS AND DISCUSSION

The effectiveness of the AS/SVE system was monitored by primarily tracking three parameters: 1) VOC concentrations in removed vapour, 2) total VOC mass removed, and 3) VOC concentrations in groundwater.

VOC Concentrations in Vapour. Vapour concentrations were detected at levels as high as 400 ppm during the pilot test. During long-term operations, VOC concentrations in vapour have declined to 0.1 ppm. The steady reduction in concentrations that has occurred from start-up (post pilot test) through the 12-month operating period demonstrates that the SVE system at the end of the first year was recovering vapour with low concentrations of VOCs. The SVE system had reached the limits of its effectiveness under continuous operation (Figure 2).

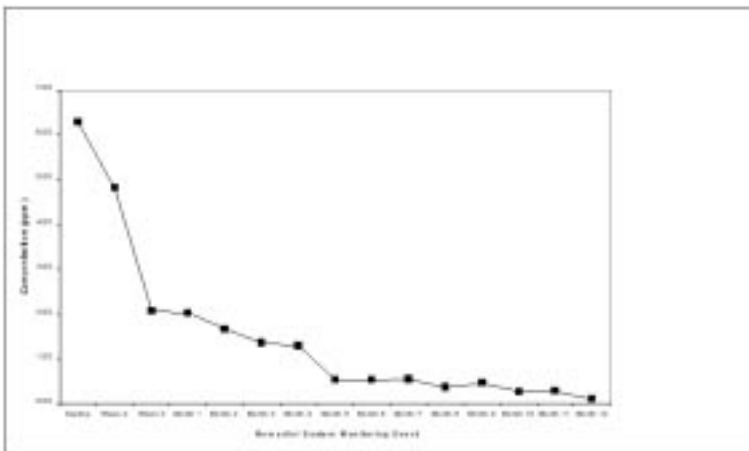


Figure 2. VOC concentration in removed vapour

VOC Mass Removed in Vapour. The primary purpose of the SVE system was mass removal, which was calculated based on vapour concentrations. As VOC concentrations declined, the VOC mass removed also decreased. After 12 months of operation, a total of 28.6 kg of VOCs was removed in the extracted vapour since start-up. Data demonstrates that the mass removal rate after one year of operation reached an asymptotic level indicating that further SVE would not significantly enhance the mass removal rate (Figure 3).

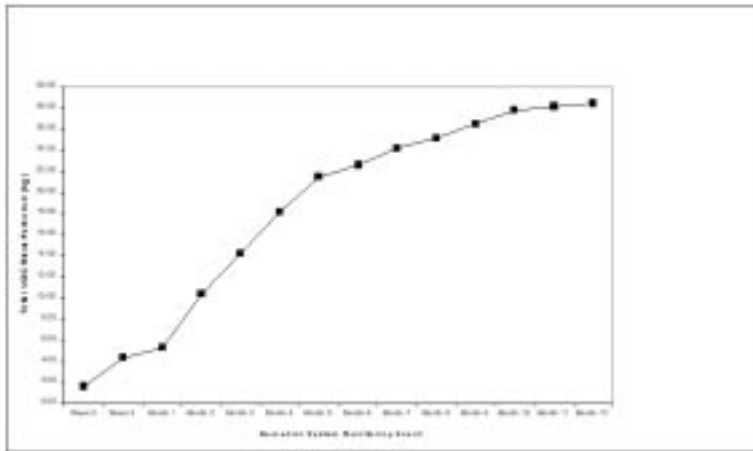


Figure 3. Demonstration of asymptotic SVE mass removal rate

VOC Concentrations in Groundwater. The apparent result of mass removal via AS/SVE was to decrease VOC concentrations in groundwater. VOC concentrations also decreased in groundwater samples collected from wells within the area remediated by the AS/SVE system. VOC concentrations as high as 258 µg/l were historically detected in monitoring wells. During the last system monitoring event, total VOCs were detected at concentrations of 41 µg/l or less. This data demonstrates that concentrations have dropped an average of 82 percent (Figure 4).

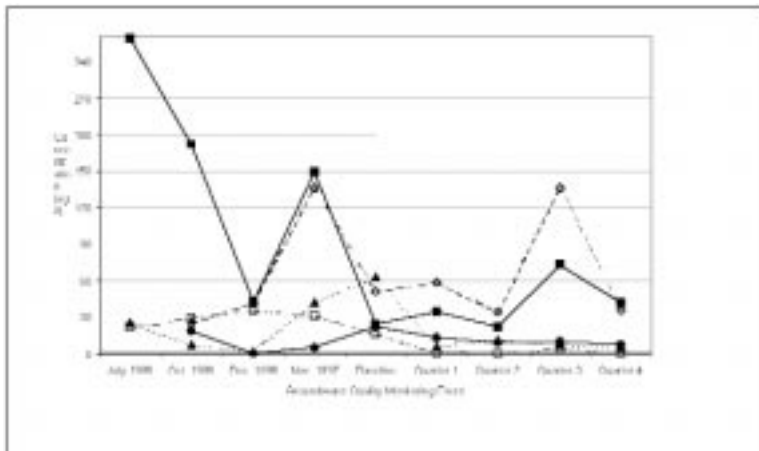


Figure 4. VOC concentrations in groundwater at key monitoring wells

After one year of system and groundwater monitoring, the results demonstrated that the AS/SVE system had reached the limits of the technology. The authorities therefore approved initiation of the monitored natural attenuation phase of the integrated remedial

project. Thus, a one year natural attenuation and groundwater monitoring program was developed and subsequently approved by the authorities. The on-going program is intended to 1) verify achievement of remediation objectives and 2) collect necessary bioparameters recommended to control and evaluate aerobic and anaerobic biodegradation processes due to naturally occurring micro-organisms in soils and groundwater.

NATURAL ATTENUATION MONITORING PROGRAM

Natural attenuation refers to naturally-occurring processes in the environment that act to reduce the mass, toxicity, mobility, volume or concentration of contaminants in various media. These in-situ processes include biodegradation, dispersion, dilution, adsorption, volatilisation, and chemical or biological stabilisation or destruction of contaminants. Intrinsic bioremediation is the process by which contaminants are transformed from toxic to non-toxic by-products through biologically-mediated reactions that occur naturally in the groundwater system. Whereas physical attenuation processes reduce the contaminant concentrations and their overall toxicity in groundwater, intrinsic bioremediation includes biological and chemical processes that destroy contaminant mass in the aquifer. Loss of contaminant mass will reduce the volume of contaminants present and result in overall plume shrinkage.

Proving that intrinsic bioremediation is occurring at a site is typically based on collection of site-specific information related to geology and hydrogeology, extent and distribution of contaminants, and collection and analysis of specific chemical and physical attributes of the aquifer matrix and groundwater. Several investigators have developed lines of evidence which they believe can adequately demonstrate that intrinsic bioremediation is occurring at a site (Rifai et al., 1995; Wiedemeier, et al., 1995, 1996; Wilson, 1996; Nyer and Duffin, 1997). The lines of evidence at the site include:

1. Bioindicators of anaerobic conditions,
2. Loss of electron donors/acceptors,
3. Loss of contaminant mass and presence of breakdown products, and
4. Geochemical parameters.

During each event the following parameters are investigated as fingerprints of on-going biodegradation processes occurring at the site.

Field Analyses

pH
Temperature
Conductivity
Dissolved Oxygen
Redox Potential (ORP)
Iron (II), Sulfate and Chloride– Hach Kit

Laboratory Analyses

VOC
TOC (total organic carbon)
Nitrate
Dissolved Gasses - methane, ethane, ethene

The on-going MNA work plan includes the collection of groundwater chemistry data from seven selected wells. As part of each monitoring event, groundwater samples are collected from on-site monitoring wells 14 and 15, on-site groundwater extraction wells E-1, E-2 and E-3, and off-site monitoring wells O-1 and O-2. Figure 1 identifies monitoring and groundwater supply well locations. To ensure that data collected are suitable for the project use and meet project quality objectives, specific field procedures have been adopted during purging and sampling activities.

Purging. It is generally accepted that water in the well casing is non-representative of the aquifer and needs to be purged prior to collection of groundwater samples. The type of pump used and the velocity with which water entered the pump intake were selected considering the high sensitivity of some bioparameters to water aeration (Dissolved Oxygen and ORP). To minimise stress (resulting in water drawdown) and water aeration, insuring bioparameters validity, purging activities were conducted using dedicated tubing, a peristaltic pump and an in-line low flow cell. To determine water stabilisation, field measurements of pH, temperature, conductivity, ORP and Dissolved Oxygen were conducted every five minutes. During the purging activities, to ensure the groundwater system was not under stress, resulting in water level drawdown, groundwater elevation measurements were recorded every five minutes.

Sampling. Upon parameter stabilisation, low flow sampling was conducted. The in-line low flow cell was disconnected and sampling flow rate adjusted to minimise aeration, bubble formation and turbulent filling of sample bottles. Volatile parameters (VOC and dissolved gasses) were sampled first. For each event, a total of 28 primary and one blind duplicate groundwater samples were collected and submitted to laboratory analysis. During each MNA event, additional samples were collected for field measurement of MNA specific parameters (Iron II, chloride and sulfate) using specific field test kits.

MNA RESULTS AND DISCUSSION

For each line of evidence, the results of the first two monitoring events conducted (MNA-Q1 and MNA-Q2) are described below.

1. Anaerobic Conditions. Typically, degradation of PCE and TCE is occurring under anaerobic conditions through a reductive dechlorination process. Anaerobic conditions in an aquifer are indicated by several factors, including: (1) low DO; (2) low reduction-oxidation (redox) potentials; and (3) the presence of methane. The first two conditions are present within groundwater at the site.

Dissolved Oxygen. DO concentrations less than 0.5 mg/l (Wilson et al., 1996) to 1.0 mg/l (Wiedemeir et al., 1996) generally indicate anaerobic conditions in an aquifer. According to Vance (1997), DO levels less than 0.18 mg/l provide an indication that conditions are favourable for methane generation. At the site, with the exception of one single measurement (1.33 mg/l) concentrations were less than 1.0 mg/l, indicating anaerobic conditions in the aquifers. However, in only one case DO level was less than the suggested threshold level for methanogenic activity (0.12 mg/l) Therefore, DO conditions confirm that conditions are favourable for anaerobic processes at the site, which would encourage biodegradation of chlorinated hydrocarbons.

Redox Conditions. The measurement of anaerobic conditions was also determined by evaluating redox. Redox is a significant indicator of microbial respiration and biotransformation. Generally, redox levels below +100 mV, measured on an ORP meter, signal the utilisation of electron acceptors other than oxygen. At the site, with the exception of one reading, all the redox values were below 50 mV showing favourable conditions for natural attenuation. Lowest redox potentials are located in the area corresponding to the heart of the VOC plume (wells E-2, 15 and E-3) and were measured as low as -197.2 mV. Globally, although some measurements do not appear on the negative end of the ORP scale, most of the measurements are on the low positive side of the scale, which is suggestive of anaerobic activity. Therefore, low redox potentials and low DO concentrations indicate good quality field procedures and thus optimal sampling conditions.

Presence of Methane. The presence of methane in an aquifer suggests that biodegradation of organic matter is occurring via methanogenesis (Wiedemeir et al., 1996). Methanogenesis represents the last in a series of reactions where anaerobic microbes sequentially use DO, nitrate, manganese, iron, sulfate and carbon dioxide to oxidise organic matter (Simpkins and Parkin, 1993; Wiedemeir et al., 1996). After the energetically more favourable electron acceptors have been depleted in the treatment zone (DO through sulfate), carbon dioxide is used as the electron acceptor and methane is produced. Therefore, the presence of methane in groundwater is indicative of strongly reducing conditions (Wiedemeir et al., 1996). The results of the first MNA event (MNA-Q1) indicate that methane was detected at concentrations ranging from 0.002 mg/l at O-1 (background) to 0.011 mg/l at E-3. Although values remain below the favourable range, it appears that highest methane concentrations were found in monitoring wells located downgradient where groundwater chemistry results show VOC contamination (wells 15 and E-3). Higher detections of methane concentrations correlate with wells exhibiting low DO and low redox values. The MNA-Q2 results indicate that methane was detected in all

the wells selected. Concentrations ranged from 0.001 mg/l at E-2 to 0.006 mg/l at 14. Similarly to MNA-Q1 results, higher concentrations of methane appear to be correlated with downgradient wells.

Based on the detection of methane, combined with low DO concentrations and low redox values, anaerobic conditions are present at the site. Although anaerobic, conditions at the site do not appear to have favourable DO for methanogenesis.

2. Loss of Electron Donors/Electron Acceptors

Electron Donors. Reductive dechlorination of PCE and TCE requires that a supply of electron donors be available to provide energy or carbon to the subsurface microbial population. Carbon sources may include fuel hydrocarbons or naturally-occurring organic carbon in the soil. Results of the determinations conducted as part of the MNA monitoring events indicate that total organic carbon concentrations are constantly below favourable values ranging from 0.92 mg/l to 2.52 mg/l. Based on concentrations of total organic carbon (TOC) measured in the aquifer microbial activity may be limited by the presence of organic carbon.

Electron Acceptors. The succession of electron acceptors, as discussed above in reference to methanogenesis, refers to the terminal electron acceptors used by microbial populations for the biodegradation of organic carbon. Electron acceptors which are more thermodynamically favoured by microbes are used first, followed by electron acceptors sequentially less favourable. DO is the electron acceptor used first during aerobic biodegradation of a substrate. After DO is depleted, nitrate may be used as an electron acceptor for anaerobic biodegradation via denitrification. Following nitrate, manganese (IV) and iron (III) are reduced to manganese (II) and iron (II), and sulfate is reduced to sulfide. Reduction of carbon dioxide via methanogenesis is the terminal electron acceptor in the sequential progression. Parameters correlated with electron acceptors that were sampled at the site include DO, nitrate, iron (II), sulfate and methane.

As previously discussed and based on analytical results, groundwater seems to be characterised by a general anaerobic condition but it appears that neither biodegradation via sulfate reduction nor methanogenesis is occurring at the site.

Nitrate concentrations indicate that denitrification processes are likely occurring. All data collected as part of the two MNA monitoring events, with the exception of one value, are within the favourable range. For both events, lowest nitrate concentration are located in the heart of the plume in correspondence with downgradient monitoring wells (wells 15 and O-2).

Dissolved iron concentrations detected during the two natural attenuation monitoring events seem to indicate sufficient iron to support biodegradation processes at downgradient wells. Once again, iron presence appear to be correlated with higher VOC concentrations.

Nitrate and iron reduction appear to be actively occurring within the main body of the VOC plume at the site. Nitrate and iron reducing conditions are evident in extraction wells E-1, E-2 and E-3 for the two monitoring events, indicating a co-occurrence of electron acceptor conditions. The electron acceptor conditions at the site are likely due to the nature of natural microenvironments present in the aquifer. Pockets of nitrate-reducing activity may be located in various portions of the aquifer, whereas iron-reducing conditions may be prevalent in others. Therefore, when groundwater is sampled from a monitoring well screened across a section of an aquifer, the water sample is a composite of that portion of the aquifer and may screen pockets of nitrate and iron-reducing conditions.

3. Loss of Contaminant Mass and Presence of Contaminant Breakdown Products

The two monitoring events conducted indicated that PCE was never detected above laboratory MDL. TCE concentrations increased (2 to 3 µg/l) at almost all the downgradient wells sampled. It is expected that PCE and TCE would represent parent compounds for chlorinated hydrocarbons detected in groundwater at the site. Detection of cis-DCE and vinyl chloride are strong indicators of biodegradation because these compounds are not typically found in solvents used at facilities. The detection of cis-DCE at significantly higher concentrations than its isomer, trans-DCE, further indicates its presence is caused by biodegradation rather than as an impurity in the originating solvent source. DCE concentrations remained high at wells 15 (from 37 to 53 µg/l) and E-3 (from 18 to 89 µg/l). VC was detected at the same wells in both monitoring events but concentrations decreased (from 18 to 4µg/l at well E-3 and from 2 to <1µg/l at well 15).

The detection of significant concentrations of degradation products compared to TCE concentrations indicates that biodegradation is occurring in the heart of the VOC plume.

Presence of Ethene and Ethane. Ethene and ethane are produced during reductive dechlorination and represent the final step of chlorinated hydrocarbon degradation. These compounds are non-toxic and break down to carbon dioxide and water. Ethene was never detected above laboratory MDL. Ethane, which is the by-product of vinyl chloride oxidation, was detected only once at 1 µg/l.

4. Geochemical Parameters

Chloride. According to Wiedemeir et al. (1996) chloride concentrations in excess of two to three times background indicate chlorinated solvent reduction. Wilson et al. (1996) also state that reductive dechlorination results in accumulation of inorganic chloride, and in aquifers with low background of chloride, the concentration of inorganic chloride should increase as the chlorinated solvents degrade. Considering MNA-Q1 background concentration (21 mg/l at well O-1) and downgradient concentrations detected at wells 15 (37 mg/l) and O-2 (65 mg/l) it seems that reductive dechlorination is occurring at downgradient locations of the site. The MNA-Q2 data were similar although slightly lower chloride concentrations were detected downgradient.

pH, Temperature and Conductivity. Groundwater parameters that were monitored during sampling of wells included pH, temperature, and conductivity. Measurements of pH and temperature indicated conditions within favourable ranges for biodegradation. Groundwater temperatures have remained consistently between 14.4 and 19.9°C, showing conditions supportive of microbial growth. Groundwater pH values have remained in the favourable range of 5 to 9. The pH values observed do not indicate any particular gradient correlated with high levels of acids generated during biodegradation at this location. Measurements of conductivity did not indicate any significant trends.

REMEDIAL OBJECTIVES

Data indicates that total halogenated VOCs have been historically and consistently detected at the site at much higher concentrations as compared to concentrations since commencing with the system operation and subsequent natural attenuation phase of the remedial project. The following table provides a summary of the groundwater chemistry results for centrally located on-site wells for total halogenated VOCs (results are µg/l):

Well ID	July 1996	Oct. 1996	Dec. 1996	Nov. 1997	Quarter 1	Quarter 2	Quarter 3	Quarter 4	MNA-Q1	MNA-Q2
14	ns	19	<MDL	5	13	9	10	7	2	2
15	ns	25	41*	136*	58*	34*	136*	34*	39*	55*
E-1	25	7	2	42*	6	11	7	5	4	5
E-2	22	29	36*	30.9*	0.9	<MDL	4	1	<MDL	2
E-3	258*	172*	43*	148*	34*	27	73*	41*	36*	96*

NS: Not sampled during indicated event.

<MDL: Less than laboratory method detection limit.

* Concentration exceeds remedial objective of 30 µg/l.

Analysing the results of the first two MNA monitoring events it appears that a global reduction of total VOC concentrations has occurred during MNA application. For the wells E-3 and 15 total VOC concentrations are still above remedial objectives but have remained within historical ranges of fluctuation indicating there was no rebound after the system shut down.

CONCLUSIONS

The active phase of the remediation (AS/SVE system) successfully removed a significant mass of VOCs (almost 29 Kg). The source reduction phase was a critical first step for application of the MNA phase of the project and regulatory acceptance.

During the first six months of MNA, the natural attenuation process seems to have globally reduced total VOC concentrations. The natural attenuation groundwater chemistry data indicated that concentrations of total halogenated hydrocarbons detected in the on-site

and off-site wells do not exceed the limit of 30 µg/l, with the exception of on-site monitoring well 15 and on-site extraction well E-3.

With the exception of the south east corner of the site (wells 15 and E-3), biological indicators and MNA specific parameters collected show favourable conditions for biodegradation. Something seems to be limiting sequential degradation of cis 1,2- DCE at these locations. Evaluation of the bioparameter data suggest the limiting factor in the south east area of the site is likely an electron acceptor or substrate, based on low TOC values.

The two monitoring events, showing comparable results for both VOCs and bioparameters, demonstrate data is repeatable and thus likely representative. Based on the fact that MNA is entirely applicable for the rest of the site and there is no migration or increase in overall VOC trends, MNA will be continued at the site.

The project illustrates the benefits of an integrated remediation strategy where MNA is being applied after mass removal to satisfy remedial objectives and regulatory requirements cost effectively.

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