Monitored Natural Attenuation; Moving Forward to Consensus

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Abstract

This paper describes the current status of the NICOLE project Monitored Natural Attenuation (MNA). The objective of the project is to provide a technical basis for a consistent measuring strategy for natural attenuation at industrially contaminated sites within NICOLE. The project intends to contribute to the understanding, and thereby acceptance, of natural attenuation as a part of a cost-effective and environmentally sound solution for contaminated sites. Starting in 1998, the existing protocols and guidelines for MNA have been listed and evaluated. It has become clear that there is much consensus amongst the existing protocols and guidelines on how to evaluate MNA. Common trends in the use of the lines of evidence and data requirements are reported here. This project is now progressing in its second phase where the possibility and efficacy of MNA at specific sites will be investigated. These sites are located in seven different countries throughout Europe. Field investigations started in summer 2000.

Keywords: monitored natural attenuation, NICOLE, NENA

INTRODUCTION

Chlorinated solvents and aromatic hydrocarbons represent the most prevalent organic groundwater contaminants in the United States, Canada and Europe (Pankow and Cherry 1996; Rijnaarts 1996). Over the past several years natural attenuation has received increasing attention as it became clearer that naturally occurring processes act to remediate contaminated groundwater. The term natural attenuation (NA) refers to ‘naturally occurring processes in soil and groundwater environments that act without human intervention to reduce the mass, toxicity, mobility, volume or concentration of contaminants in those media’ (US EPA 1999).

The most important destruction mechanism during natural attenuation is microbially mediated degradation. Non-destructive attenuation mechanisms include sorption, dispersion, dilution and volatilisation. During microbial degradation the contaminants are broken down ultimately into harmless components such as water, chlorine, and carbon dioxide. In the latter processes the contaminants remain, but the concentrations observed in the groundwater decrease. Inorganic contaminants such as heavy metals, arsenic and cyanide can be attenuated by chemical and physical processes but cannot be degraded.

Although the occurrence of natural attenuation processes that can contribute to decreasing concentrations of contaminants, and to risk reduction has been established, the use of these processes as a remedial strategy at contaminated sites is a rather new phenomenon in Europe. This use of naturally occurring attenuation processes as a remedial strategy has to be monitored in practice. This approach is called monitored natural attenuation (MNA). There is much debate on the efficacy, the reliability and the long-term sustainability of MNA. There is also much uncertainty as to how to measure attenuation in the field, and how to ‘prove’ in advance, that reliance on natural attenuation to achieve site-specific remedial objectives is reasonable.

In The Netherlands, the UK, and recently in Germany, national projects have been set up to investigate the technical basis for application of MNA, and to formulate decision support tools (BMBF Programm 2000; Carey *et al.* 2000; Sinke *et al*. 1998). Within NICOLE an MNA review project started in 1998 involving industrial partners Europe-wide. The objective of this NICOLE project is to provide technical evidence to

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support the acceptance of natural attenuation as a part of a cost-effective, and environmentally sound, solution for contaminated sites. The goal of the project is to provide a technical basis for a consistent monitoring strategy for NA at industrially contaminated sites within NICOLE. The project takes place in phases. In the first phase of the project existing protocols and guidelines on MNA have been reviewed to distil common features (Sinke and Hecho 1999). In the second phase of the project, which started in spring 2000, measurements will take place at eight demonstration sites to develop a protocol for NICOLE members to evaluate the possibilities for and likely effectiveness of NA at their sites: the acronym for this protocol is NENA (NICOLE Evaluation of NA). NENA will be based on existing guidelines and protocols and tested at contaminated industrial sites in several European countries.

OVERVIEW OF PROTOCOLS AND GUIDELINES

The evaluation of the potential applicability of MNA requires a reliable technical protocol to confirm and quantify its likely efficacy. In the USA, several initiatives, listed in Table 1, have been taken to develop technical protocols to assess sites with petroleum hydrocarbons, chlorinated hydrocarbons or recalcitrant compounds (ASTM 1997; RTDF 1997; US EPA 1999; Wiedemeier *et al*. 1994). These protocols have been updated and adapted regularly to keep up with scientific innovations and increasing expertise. An overview of current protocols is given by Rittmann *et al.* (2000). While the use of technical protocols gives recommendations for site-specific data collection and analysis to support remediation, such protocols for MNA are not agreed or accepted in many European countries or by their national regulatory agencies. There are different emerging European protocols and guidelines. These differ primarily in tone and emphasis, reflecting the different perspectives and responsibilities of the entities that developed them. For MNA to be allowed as a remedial strategy, the protocols and guidelines all have as their most important criteria:

- MNA has to be protective of human health and the environment; and
- MNA has to be effective within a reasonable time frame.

In the USA, the EPA, in its role as regulatory agency, recognises MNA as a viable method of remediation for soil and groundwater which can be evaluated and compared to other methods of achieving site remediation as a part of the remedy selection process. An EPA directive has been produced, intended to promote consistency in how MNA remedies are proposed, evaluated, and approved for protection of human health and the environment (US EPA *et al*. 1999).

Table 1. Overview of evaluated protocols and guidelines

Protocol/guideline	Compound
RTDF 1997	Chlorinated solvents
*Wiedemeier *et al*. 1996, 1998	Chlorinated solvents
*NOBIS 1998[1], 2000[2]	Chlorinated solvents and BTEX
ASTM 1997	Petroleum hydrocarbons
AFCEE 1995[3]	Petroleum hydrocarbons
*OSWER 1997[4], 1999[4]	Petroleum hydrocarbons

*New, adapted editions of these protocols are available.
1. Sinke *et al*. 1998
2. Sinke *et al.* in press
3. Wiedemeier *et al.* 1995
4. US EPA 1999

General set-up of protocols

The protocols reviewed generally describe a stepwise framework to demonstrate the feasibility of MNA at a given site. The protocols include tables with parameters that have to be used and methods to determine them. The parameters to be collected include field data on hydrology, spatial distribution of contaminants and presence of receptors.

In all protocols it is stated that to support remediation by MNA, the proponent must scientifically demonstrate that attenuation, or even better degradation, of site contaminants is occurring at rates sufficient to be protective of human health and the environment. The protocols use the data that are measured in the field as a basis in the decision framework and generally work with 'lines of evidence', i.e.:

1. *First line: historical contamination data*
   Historical groundwater data and/or soil chemistry data that demonstrate a clear and meaningful trend of decreasing contaminant mass and/or concentration over time at appropriate monitoring or sampling points. In the case of a groundwater plume, the decreasing concentrations should not be solely the result of plume migration. In the case of inorganic chemicals, the primary attenuation mechanism should also be understood.
2. *Second line: application of screening models*
   Hydrological and geochemical data can be used in an analytical screening model to demonstrate indirectly the type of natural attenuation process active at the site and the rate at which such processes will

reduce contaminant concentrations to required levels.

3. *Third line: additional laboratory or field data*
   Data from field or microcosm studies which directly demonstrate the occurrence of particular natural attenuation processes at the site and its ability to degrade the contaminants of concern.

First line of evidence and plume life cycle evaluation

The first and most important line of evidence for the occurrence of natural attenuation is obtained by evaluating field data. Depending on the type of contaminant and the situation, different factors can be used as indications of the occurrence of NA. For instance, chlorinated solvents such as trichloroethene (TCE) can be dechlorinated stepwise under anaerobic conditions and the presence of daughter-products such as cis-1,2-dichloroethene (cis-1,2-DCE) and vinylchloride (VC) are a strong indication of NA. Also changes in redox conditions can be a strong indication of the occurrence of NA. A very important type of evidence can be obtained by data on the historical plume behaviour. The interpretation of the plume behaviour and the reliability of the often scattered historical data are not straightforward in all cases as different analytical methods may have been used over time, and wells may not have been positioned optimally. For sites with multiple source regions and mixed contaminants, the historical development of the plumes can seldom be retraced. Nevertheless, in a number of well-described and investigated cases, trends can be observed in the development of a plume; these plumes can be grouped in different categories as illustrated in Figure 1.

- expanding: residual source present. Mass flux of contaminants exceeds the assimilative capacity of the aquifer;
- stable: insignificant changes. Active or passive remediation processes are controlling the plume length;
- shrinking: residual source (nearly) exhausted. Active or passive remediation processes significantly reducing plume mass;
- exhausted: average plume concentration very low and unchanging over time.

A stabilised or receding plume is direct evidence for natural attenuation. Several studies addressed the changes in plume length and concentration for dissolved petroleum hydrocarbon plumes and plumes of chlorinated solvents (Mace *et al*. 1997; Newell *et al*. 1990; Rice *et al*. 1995). A general trend in these studies has been that BTEX plumes are shorter than plumes of chlorinated ethenes or other chlorinated solvents (Newell and Connor 1998). Most of the studies on plume behaviour were carried out at sites where aerobic conditions prevail. Generally aerobic conditions are favourable for the degradation of petroleum hydrocarbons including BTEX. In the USA, the majority of the petroleum hydrocarbon plumes appear to be either stable or shrinking and only as little as 5–10% are still expanding (Newell and Connor 1998). Plumes at approximately 75% of 604 sites investigated in Texas, California and Florida are under 75 m long.

Second line of evidence

Site investigation data should first be used to develop a site-specific conceptual model representing the groundwater flow and solute transport system. The model is typically used to quantify groundwater flow, sorption, dilution and biodegradation. The results of these calculations are used to make a prognosis for the impact of NA over time. Therefore, all available data must be organised so that the effect of NA is shown clearly. The model should predict plume behaviour, to identify mass loss of contaminants in relation to groundwater flow, and additional data to be collected at specific locations.

Third line of evidence

This line includes additional evidence from field, laboratory or mesocosm systems, for example, of the occurrence of biological activity. Not all protocols require this type of evidence. Clearly, additional evidence might help to strengthen the other observations. Several new techniques are currently being developed that

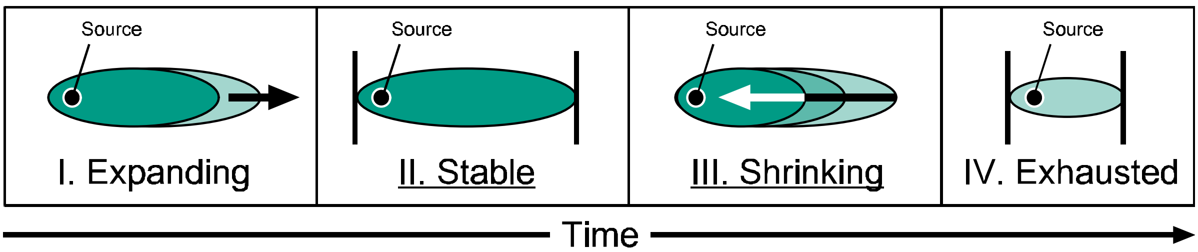

Figure 1. The life cycle of a contaminant plume that is naturally attenuated with the four succeeding stages of expanding, stable, shrinking and exhausted (adapted from Newell and Connor 1998).

might be used to demonstrate that degradation occurs or prove the presence of the bacteria in the aquifer capable of degrading the site contaminants. Some examples are:

- measurements on stable isotopes such as the ratio $^{13}C/^{12}C$ that can be used to demonstrate the degradation of petroleum hydrocarbons;
- RNA probes that detect the genetic codes present in specialised bacteria that can degrade chlorinated solvents;
- methods to determine the activity of oxygenases that are the enzymes needed to degrade the chlorinated solvents co-metabolically.

SEQUENTIAL STEPS IN INVESTIGATING NATURAL ATTENUATION

All of the protocols and guidelines reviewed use a tiered procedure to investigate NA at a site. The aim is to make as early as possible and with as little expense as possible, a good estimation of the probability that MNA is an appropriate strategy to achieve site-specific remediation objectives within a reasonable time frame. The number of steps, the amount of detail and the sequence of steps, differ between the different protocols but the protocols are essentially similar (Table 2).

Table 2. Overview of sequential steps as used in most protocols and guidelines

Step	Line of evidence	Activity
1	1st	Collect information on available site data, identify additional data requirements and collect data
2	1st	Assess potential for NA and develop hypothesis to explain NA
3	3rd	Field and laboratory experiments
4	2nd	Simulate NA using transport & fate model to predict future plume behaviour
5	2nd	Test hypothesis and carry out risk evaluation
6		Integration of NA in long-term management strategy & verification of MNA

Typically, the first steps focus on obtaining information for the first line of evidence while at a later stage the guidelines focus on the modelling required for the second line of evidence. The third line of evidence (i.e. experiments under field conditions or in the laboratory to demonstrate biological capabilities to degrade the contaminants of concern) is not included in all protocols.

The following phases are typical:

1. The collection of basic information such as historical, technical and field data. The structured evaluation of these datasets and their graphical representation, allows the identification of data gaps.
2. A qualitative procedure can be applied to assess the potential for NA. Some of the protocols (Wiedemeier *et al*. 1998; Sinke *et al*. in press) use a type of quick scan giving points for field parameters but these ‘score lists’ only serve as indications for the occurrence of NA and can never be used as such to ‘demonstrate’ the efficacy of NA. In the case of chlorinated solvents, the presence of daughter-products is a strong indication of the occurrence of degradation processes. Also the redox conditions are included here as, for example, the degradation of petroleum hydrocarbons is favoured under aerobic conditions. Special attention has to be paid to the evaluation of the sustainability of the processes, which is important to assess the capacity of NA to persist in the long term.
3. A third step, not included in all protocols, includes different types of experiments under field conditions or in the laboratory to demonstrate biological activity and the abilities of the soil biota to degrade the contaminants of concern.
4. The next phase is to simulate NA processes using a transport and fate model to predict the long-term behaviour of the plume. In some cases, simple models can be used such as BIOSCREEN but in other cases a more complex modelling approach using MODFLOW and RT3D is more suitable.
5. The comparison of field data and the model output allows conclusions on the efficacy of NA and enables the risk assessment of current and future plume conditions.
6. Finally, NA can be implemented as a remedial strategy, provided that a contingency plan has been formulated. A long-term monitoring strategy has to be designed to track the plume development and to validate or refine the modelling results.

Summary of data requirements for the protocols

The different protocols are similar with regard to the parameters recommended to be measured for the first and second lines of evidence. Differences in the number and frequency of the measurements are relatively minor. Even the protocols for chlorinated solvents and petroleum hydrocarbons are largely alike and differ mainly in the pollutants and daughter-products to be measured. However, where the protocols differ is that some of them tend to be more prescriptive, defining the analytical methods to be used (Wiedemeier 1995) while others indicate more generally the approach to follow (ASTM 1997; Sinke *et al*. in press; US EPA 1999).

The type of data to be collected to characterise a site and to determine the possibilities for NA can be divided into two categories:

- hydrogeological (determines transport of the contaminants);
- chemical and biological (determines fate of the contaminant).

Hydrogeological: the properties of an aquifer that have the greatest impact on contaminant transport include hydraulic conductivity, hydraulic gradient, and porosity. In Table 3 the typical parameters needed to characterise the hydrogeology of the site and the presence of potential receptors are listed. These data are used as the basis for the site-specific conceptual model.

Chemical and biological parameters: measurements of the concentration and spatial distribution of the major contaminants and, in the case of chlorinated solvents, the daughter-products necessary for characterising the extent of the pollution (Table 3). In addition, the distribution of contaminant in the aquifer needs to be understood, and whether it is present as pure product (LNAPL or DNAPL), or whether it is sorbed or dissolved in the groundwater. Finally, the conditions in the aquifer, as indicated by its chemical and biological properties, determine which reactions may proceed: sorption, degradation, chemical transformation, etc. The amount of organic carbon determines the sorption, and consequently, the retardation of contaminants.

Redox conditions determine the type of degradation process that might proceed in the aquifer. Aerobic conditions favour the degradation of petroleum hydrocarbons and lower chlorinated solvents such as DCE and VC but do not favour the degradation of the higher chlorinated solvents such as PCE and TCE. For the aerobic degradation of petroleum hydrocarbons, the bal-

Table 3. Parameters that are advised to be measured to characterise the hydrogeology and receptors of the site by the different guidelines

Parameter	Chlorinated solvents			Petroleum hydrocarbons	
	AFCEE 1996[1]	RTDF 1997	NOBIS 1998[2]	AFCEE 1995[3]	ASTM 1997
Characterisation of aquifer					
Regional hydrogeology	yes	yes	Yes	yes	yes
Site lithology & stratigraphy	yes	yes	Yes	yes	yes
Porosity	yes	yes	Yes	yes	yes
Water level elevations	NS	NS	NS	NS	yes
Direction of ground water flow	yes	yes	Yes	yes	yes
Travel rate of groundwater	yes	yes	Yes	yes	NS
Range of seasonal water level fluctuations	NS	NS	NS	yes	yes
Depth of pollution	yes	yes	Yes	yes	yes
Extent of unsaturated soil impact	NS	NS	NS	NS	yes
Potential for vertical migration	NS	NS	NS	NS	yes
Groundwater recharge and discharge areas	NS	yes	NS	yes	NS
Push-pull tests or pumping tests #	NS	NS	yes*	yes	NS
Characterisation of receptor					
Survey of nearby water supply wells	yes	yes	note 1	yes	yes
Location of potential receptors	yes	yes	note 1	yes	yes
Identification of preferential flow pathway	NS	yes	note 1	yes	yes

Parameters marked with * are optional, with # under development, NS: Not Specified or mentioned in the protocol.
1. Wiedemeier *et al*. 1996
2. Sinke *et al*. 1998
3. Wiedemeier *et al*. 1995
Note: In The Netherlands the collection of this information is prescribed regulatory and is present for every known site.

Table 4. Recommended parameters to characterise the dimensions of the contamination and the biological and chemical conditions in an aquifer in soil (S), groundwater (W) or gas phase (G) for chlorinated solvents and petroleum hydrocarbons

Parameter	Chlorinated solvents			Petroleum hydrocarbons	
	AFCEE 1996[1]	RTDF 1997	NOBIS 1998[2]	AFCEE 1995[3]	ASTM 1997
Characterisation of contaminant					
PCE	Yes (W)	Yes	Yes	NS	NS
TCE	Yes (W)	Yes	Yes	NS	NS
DCE	Yes (W)	Yes	Yes	NS	NS
VC	Yes (W)	Yes	Yes	NS	NS
Ethene, ethane	Yes (W)	Yes	Yes	Yes (W)	NS
Chloroethane	Yes (W)	Yes	*	NS	NS
1,1,1, trichloroethene	Yes (W)	Yes	*	NS	NS
1,1 dichloroethene	Yes (W)	Yes	*	NS	NS
Aromatic hydrocarbons (BTEX)	Yes (W,G)	Yes	Yes	Yes (S, W)	Yes (S,W)
Trimethylbenzene	NS	NS	NS	Yes (S, W)	NS
Methyltertiairybutylether (MTBE)	NS	NS	NS	NS	Yes (S,W)
PAHs	Yes (W)		Yes	*	NS
Other contaminants			Yes	NS	Yes
Carbon dioxide	Yes (G)		NS	NS	Yes (W)
Ketones	NS	Yes	NS	NS	NS
Volatile hydrocarbons	Yes (G)	Yes	NS	yes (S, W)	Yes
Characterisation of total carbon as sorbent and energy source					
Dissolved organic carbon	Yes (W*)		Yes	NS	NS
Total organic carbon	Yes (S,W)		Yes	Yes	Yes
Quality natural organic matter #	Yes (W)	NS	*	NS	NS
Volatile fatty acids	NS	Yes	NS	*	NS
Characterisation of redox					
Oxygen	Yes (W,G)	Yes	Yes	Yes (W)	Yes (W)
Nitrate	Yes (W)	Yes	Yes	Yes (W)	Yes (W)
Manganese		Yes	NS	NS	Yes (W)
Iron(II)	Yes (W)	Yes	Yes	Yes (W)	Yes (W)
Sulphate	Yes (W)	Yes	Yes	NS	Yes (W)
Sulphide	Yes (W)	Yes	NS	Yes (W)	Yes (W)
Methane	Yes (W)	Yes	Yes	NS	Yes (W)
Hydrogen #	Yes (W)	*	*	Yes (W)	Yes (W)
Oxidation reduction potential	Yes (W)	Yes	NS	Yes (W)	Yes (W)
Biologically available iron #	Yes (S)	NS	NS	NS	NS
Propane/propene	NS	Yes	NS	NS	NS
Biological indicators					
Microcosms	*	*	*	*	NS
Oxygenates, RNA probes #	NS	*	*	NS	NS
Dehydrogenase	NS	NS	NS	*	NS
Microbial counts, biomass	NS	*	NS	NS	NS
Other parameters					
Alkalinity	Yes (W)	Yes	NS	Yes	Yes
Calcium, potassium	NS	Yes	NS	NS	NS
Sodium	NS	Yes	NS	NS	NS
Ammonia	NS	Yes	NS	NS	NS
Phosphate	NS	Yes	NS	NS	NS
PH	Yes (W)	Yes	Yes	Yes	Yes
Temperature	Yes (W)	Yes	Yes	Yes	Yes
Conductivity	Yes (W)	Yes	NS	Yes	Yes
Chloride	Yes (W)	Yes	NS	Yes	NS
Nitrite	NS	Yes	NS	NS	NS
Toxic metals	NS	Yes	NS	NS	NS

Parameters marked with * are optional, marked with # under development
NS: not specified in protocol
1. Wiedemeier *et al*. 1996
2. Sinke *et al*. 1998
3. Wiedemeier *et al*. 1995

Relevant Internet Sites

Name	Website	Information
Air Force Center for Environmental Excellence	www.afcee.brooks.af.mil	Many digital presentations on plume studies
Air Force Center for Environmental Excellence	www.afcee.brooks.af.mil/er/ert/natural.htm	Technology transfer on MNA
NNAGS Network for Natural Attenuation in Groundwater and Soil	www.shef.ac.uk/~NNAGS	Information on MNA in the UK. Scientific backgrounds, projects, members and publications
NATO-CCMS	www.nato.int/ccms	Information on NATO-CCMS pilot projects
NICOLE	www.NICOLE.org/NICOLE2/projects/NICOLEindex.shtml	Overview of NICOLE projects
NOBIS	www.bouwweb.nl/cur/nobis	Information on current projects on remedial technologies and NA
RTDF Remediation Technologies Development Forum	www.rtdf.org	General overview of remediation technologies
Technology Information Office	www.clu-in.org	Information on innovative treatment technologies and site characterisation
UK Environmental Agency	www.environment-agency.gov.uk/gwcl/ngwclc.htm	Information on UK National Groundwater & Contaminated Land Centre
US EPA	www.epa.gov	EPA site
US EPA	www.epa.gov/oerrpage/resources/gwdocs/ (www.epa.gov/oerrpage/resources/gwdocs/protocol.htm) – access via PDF file	Technical protocol for evaluating MNA of chlorinated solvents in groundwater
US EPA	www.epa.gov/swerust1/mna/index.htm	OSWER directive 9200.4-17P; Use of MNA at Superfund, RCRA corrective action and underground storage tank sites

ance between the amount of organics present and the flux of oxidising equivalents (oxygen, nitrate, etc.) determines the efficacy of the degradation and thereby the plume behaviour. Under very reducing conditions (e.g. sulphate reducing or methanogenic) reductive dechlorination of the higher chlorinated solvents (PCE, TCE) is favoured. Under these reduced conditions, organic carbon is used as electron donor (energy source) for the dechlorination of chlorinated solvents which are effectively used as electron acceptors. The amount of the energy source, present as dissolved organic carbon, total organic carbon or as co-pollutant (BTEX), relative to the amount of electron acceptors, i.e. contaminant, determines the likelihood and duration of the degradation process. Also, the quality of the source is important but this cannot yet be determined with standardised methods. However, the amount of volatile fatty acids may give information on the 'quality' of the energy source.

Several other parameters can be determined that help to describe the conditions present in the aquifer. Generally, these parameters are not measured in all samples or are only measured in a later tier to get additional, more detailed information:

- levels of phosphate and ammonia give an indication on the nutritional state of the bacteria;
- pH: when the pH is too low (< 5) or too high (> 9) the bacterial activity might be hampered;
- heavy metals: the presence of large amounts of heavy metals might inhibit the bacterial activity;
- temperature directly influences the rate of chemical, physical and biological processes;
- alkalinity: an increased alkalinity compared to background indicates the production of carbon dioxide;
- conductivity and chloride concentration: an increase in these values compared to background indicates the release of chlorine from chlorinated solvents.

CONCLUSIONS

A number of protocols and guidelines have been published recently about the evaluation of MNA as a remedial approach at contaminated sites. There is much consensus on the data requirements and the evaluation process for determining the feasibility of MNA as a contaminated land management approach. However, the actual application of MNA as a remedial strategy is still infrequent in Europe. This could be due to a lack of well-documented demonstration sites. Within the

NICOLE network several industrial partners are currently investigating the possibility of applying MNA at specific sites. These sites could be used as European demonstration sites. Probably at some of the sites MNA will be a good and reliable approach as the sole measure, while at others MNA will be used in addition to active methods such as pump and treat. At some sites source control may be necessary while at others the remaining source can be left in place. Finally, at some sites MNA may not be applicable at all, due to unfavourable conditions. The evaluation of all these data at well-investigated sites will form the basis of the NENA (NICOLE Evaluation of Natural Attenuation) protocol which is intended to evaluate whether MNA can be used as the (sole) remedial strategy or not, and as a tool to contribute to the discussion with the authorities on the conditions and restrictions for the application of MNA.

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