Monitored Natural Attenuation for Inorganic Constituents at Coal Combustion Residuals Sites

Technical Brief

INTRODUCTION

Monitored natural attenuation (MNA) is a remediation alternative that has been a component of corrective action for cleanup of both organic and inorganic constituents in groundwater since the 1990s. Where site conditions are conducive to MNA, it has the potential to provide a more sustainable, and potentially lower-cost, alternative to other remediation technologies such as pump and treat. MNA also provides a polishing phase of remediation in cases where other remediation technologies target specific areas with high concentration or high risk.

From existing case histories, the Electric Power Research Institute (EPRI) evaluated MNA for 24 inorganic constituents of interest with respect to coal combustion residuals (CCR), with emphasis on arsenic, boron, and selenium. Although only three case histories were identified for CCR facilities, extensive information exists, particularly from the mining and mineral processing industries, that can be applied to MNA for CCR (EPRI 2015a). This technical brief describes the MNA process, and the significant effort involved for a full demonstration that MNA can be a viable component of a groundwater corrective action at a CCR management facility.

USEPA MNA PROTOCOLS

The USEPA defines MNA as follows:

> …the reliance on natural attenuation processes (within the context of a carefully controlled and monitored site cleanup approach) to achieve site-specific remediation objectives within a time frame that is reasonable compared to that offered by other more active methods. The "natural remediation processes" that are at work in such a remediation approach include a variety of physical, chemical, or biological processes that, under favorable conditions, act without human intervention to reduce the mass, toxicity, mobility, volume, or concentration of contaminants in soil or groundwater. (USEPA 1999, 2015)

USEPA does not consider MNA a no-action response. When properly implemented, MNA can help to restore an aquifer to beneficial uses by removing constituents from groundwater and immobilizing them onto aquifer solids. Decisions to utilize MNA as a remedy or remedy component are supported by site-specific data and analyses that go beyond data collection and analysis for a groundwater monitoring program (USEPA 1999, 2015).

According to USEPA (2015) requirements for MNA, the following active steps must be taken. These steps constitute USEPA's four tiers of MNA (USEPA 1999, 2007a):

1. Demonstrate that the groundwater plume is not expanding.
2. Determine the mechanisms and rates of attenuation.
3. Determine that the capacity of the aquifer is sufficient to attenuate the mass of constituents in groundwater and that the immobilized constituents are stable and will not remobilize.
4. Design a performance monitoring program based on the mechanisms of attenuation, and establish contingency remedies (tailored to site-specific conditions) should MNA not perform adequately.

Based on MNA case histories for inorganic constituents, time frames range from 5 to 150 years, with 10 to 40 years being the most commonly cited.

EPRI (2018a) provides a framework for performing a four-tiered MNA analysis as part of a groundwater corrective action at a CCR landfill or impoundment. It includes a flow chart outlining the tiered approach (Figure 1), types of field and laboratory tests that can be performed to meet the objectives of the MNA tiers, and a detailed case example for a full MNA study including field and laboratory tests and their results.

Source Control

USEPA considers source control a prerequisite for an MNA remedy. Source control helps ensure the timely attainment of remediation objectives. USEPA expects that source control measures will be evaluated for all impacted sites and that source control measures will be taken at most sites where practical

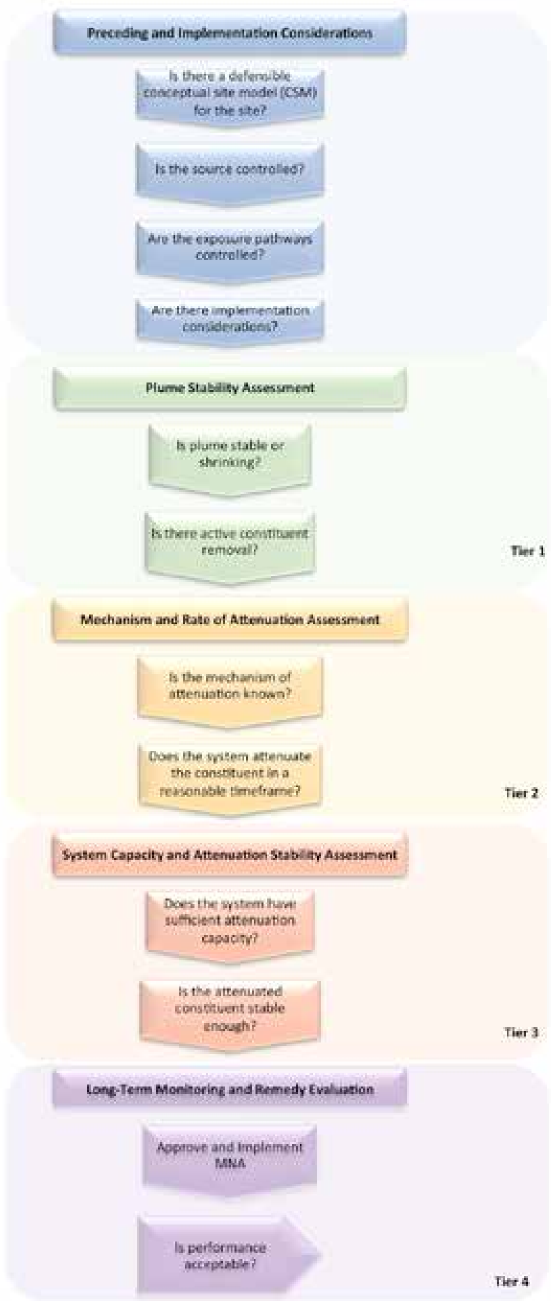

Figure 1 – MNA Decision Framework Overview
(Source: EPRI 2018a)

(USEPA 1999, 2015). Methods for source control at CCR management facilities can include one or more of the following: decanting and dewatering for ponds, capping, removal of CCR, liners, in situ solidification/stabilization (ISS), barrier walls, and permeable reactive barriers (PRBs).

Tier 1 – Demonstrate That the Groundwater Plume Is Not Expanding

USEPA states that sites where constituent plumes are no longer increasing in extent, or are shrinking, are the most appropriate candidates for MNA remedies (USEPA 1999, 2007a, 2015). Plume stability may be determined by groundwater monitoring over time. Several methods may be used to determine plume stability, and direct measurement methods are preferred, according to USEPA. For example, the areal extent of a plume may be mapped in plan view or contoured. Contouring may be helpful in demonstrating natural attenuation, particularly if concentrations of constituents are decreasing within the plume boundary (see Figure 2).

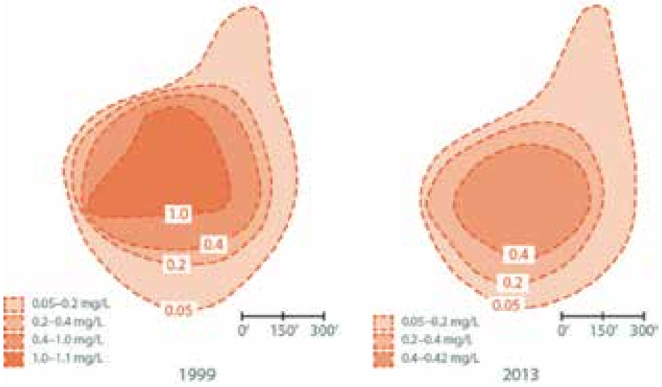

Figure 2 – Dissipation of Arsenic Plume Through Time, Map View
(Source: EPRI 2015a)

Graphs of constituent concentrations as a function of time may also show decreasing trends that document natural attenuation. These would be supplemented by upgradient and downgradient well data within the plume to show that constituents are actually attenuating, rather than the plume simply migrating downgradient. Groundwater modeling may be used to predict plume extent over time. This type of modeling is most effective when the transport model is calibrated and periodically compared to post modeling data from groundwater monitoring.

Tier 2 – Determine the Mechanisms and Rates of Attenuation

The second tier of MNA analysis consists of two parts: 1) identify the specific mechanisms controlling constituent attenuation; and 2) estimate the attenuation rates. Attenuation mechanisms can be placed in two broad categories, physical and chemical.

Physical mechanisms include dilution, dispersion, flushing, and related processes. All CCR constituents are subject to physical attenuation mechanisms. In its most recent guidance, USEPA (2015) discourages using dilution and dispersion as primary MNA mechanisms, as these mechanisms disperse contaminant mass rather than immobilize it. However, USEPA (2015) goes on to say that dilution and dispersion may be appropriate as a polishing step, e.g., at the boundaries of a plume, when source control is complete, another remedy is being used at the site, and appropriate land use and groundwater use controls are in place. Physical attenuation mechanisms are important for MNA application at CCR sites, because constituents such as boron, chloride, and lithium are poorly chemically attenuated, such that physical mechanisms may be the only attenuation processes operative for these constituents.

Common chemical mechanisms of attenuation for CCR constituents include adsorption to, or coprecipitation with, oxides and hydrous oxides (oxyhydroxides) of iron and manganese; coprecipitation with, and adsorption to, iron sulfides such as pyrite (FeS_2); and precipitation as carbonates, sulfides, sulfates, and/or phosphates (USEPA 2007b). Table 1 shows six classes of chemical attenuation mechanisms and the constituents to which they apply.

Analyses of both groundwater and aquifer solids can be performed to identify attenuation mechanisms. Many tools and techniques are available to determine mechanisms of attenuation; a few are listed in Table 2. Many of these tools and techniques are also applicable to Tier 3 of the MNA investigation.

Table 1 – Reported Chemical Attenuation Mechanisms by Constituent

Constituent[1]	Precipitation	Coprecipitation	Ion exchange	Sorption to sulfide minerals	Sorption to aluminum, iron, and manganese oxides and oxyhydroxides	Sorption to other minerals or organic matter
Antimony		✓		✓	✓	
Arsenic	✓	✓		✓	✓	✓
Barium	✓	✓	✓		✓	
Beryllium	✓				✓	
Boron			✓			
Cadmium	✓	✓	✓	✓	✓	✓
Calcium	✓		✓			
Chloride						
Chromium	✓	✓		✓	✓	✓
Cobalt		✓			✓	✓
Copper	✓	✓		✓	✓	✓
Fluoride	✓		✓		✓	
Lead	✓	✓	✓	✓	✓	✓
Lithium						
Manganese	✓	✓	✓		✓	
Mercury	✓	✓		✓	✓	✓
Molybdenum					✓	
Nickel	✓	✓	✓	✓	✓	✓
Radium		✓	✓		✓	✓
Selenium	✓			✓	✓	✓
Sulfate	✓				✓	
Thallium					✓	
Vanadium				2	2	2
Zinc	✓	✓	✓		✓	✓

Notes:

1. *Based on literature review, likely not complete for some constituents; this table shows more mechanisms for some constituents than EPRI 2015a, due to review of additional literature since the publication of EPRI 2015a*
2. *Sorption with no mineral association reported for vanadium*

Table 2 – Selected Techniques and Key Analytes to Determine Attenuation Mechanisms (Tier 2) and Stability of Immobilized Constituents (Tier 3)

Solids: Soil, Aquifer Media, Precipitates	Groundwater
• XRD to identify attenuating mineral phases and chemical composition • SEM and associated electron diffraction to identify distinct mineral forms (e.g., framboidal pyrite) and chemical composition • Sequential chemical extraction to determine association of constituents with attenuating phases (e.g., arsenic with iron oxides) and stability • K_d (solid-water partition coefficient) analysis to determine sorption capacity • Batch or column tests using site soil and groundwater or simulated site groundwater	• Key Analytes – Indicator parameters: pH, ORP, DO, alkalinity – Major cations and anions, constituents of concern – Sulfate and sulfide, ferrous and ferric iron • Pourbaix (Eh-pH) diagrams • Mass flux calculations • Geochemical and groundwater modeling • Batch or column tests using site soil and groundwater

Notes:

DO: dissolved oxygen

K_d: solid-water partition coefficient

ORP: oxidation reduction potential

SEM: scanning electron microscopy

XRD: x-ray diffraction

Very little has been published on determining the rate of attenuation for inorganic constituents. For chemical attenuation, an estimate of attenuation rates could include a calculation of the time for the apparent transfer of mass from the aqueous (groundwater) to the solid (aquifer) phase. Empirical methods for estimating rates include concentration-time trends or concentration-distance trends (e.g., between two wells along a flow path). Concentration-distance estimates require quantification of groundwater velocity. Empirical methods are more consistent with USEPA's guidance to use site data rather than modeling (USEPA 2007a). For physical attenuation, dilution calculations or groundwater modeling that quantifies dilution, dispersion, or flushing could be performed.

If historical groundwater data are available, then the rate of attenuation may be directly calculated by dividing the decrease in constituent mass in groundwater in the impacted area by the period of time, provided the plume is stable or receding. Laboratory-based methods to estimate the rate of attenuation include batch and bench-scale tests, where rates of constituent sorption onto solids, for example, are measured directly (Fuller and Harvey 2000). Other methods include point decay calculations (DOE 2014), and mass flux calculations based on site-specific data.

Geochemical and groundwater modeling may be used as part of all four MNA investigation tiers, and both have been used as part of Tier 2 (mechanisms and rates of attenuation) and Tier 3 (long-term capacity for attenuation and stability of the immobilized constituents). Despite the widespread use of models, USEPA (2007a) recommends that estimates of attenuation rates be based as much as possible on field measurements rather than model predictions.

Tier 3 – Determine the Capacity of the Aquifer and Stability of the Immobilized Constituents

The third tier of MNA analysis consists of two parts: 1) determine the capacity of the aquifer to attenuate the mass of constituents of concern; and 2) determine the stability of the immobilized constituents. Two possible factors that could create an insufficient capacity for attenuation are 1) changes in groundwater chemistry (e.g., pH or redox potential) that result in slower rates of attenuation or remobilization of contaminants; and 2) insufficient mass of adsorbing solids in the aquifer media (USEPA, 2015).

Determining the long-term capacity for attenuation requires a comparison of the mass of constituents to be attenuated (mass flux) to the aquifer's ability to attenuate the constituents. Calculations based on bench-scale sorption tests and geochemical and groundwater modeling have been used to demonstrate the long-term capacity for attenuation. One Tier 3 approach using sorption tests is as follows:

- Measure the attenuation capacity (mass expressed in grams or moles) of a small representative sample of aquifer solids by determining sorption isotherms (e.g., Langmuir or Freundlich) in the laboratory.
- Multiply the unit mass attenuated in the small sample (e.g., grams of constituent per volume of soil or aquifer solids) by the volume of the aquifer within the plume.
- Use concentrations of constituents in groundwater and mass flux to determine the mass to be attenuated.
- Compare the constituent mass in groundwater to the aquifer's attenuation capacity.

This approach is based on sorption as the primary attenuating mechanism. If precipitation or coprecipitation is a significant attenuating mechanism, groundwater would need to supply the reactants in sufficient quantity, and the geochemical conditions would need to remain suitable for precipitation and/or coprecipitation of the constituents of interest. Geochemical modeling could be used to evaluate precipitation and coprecipitation.

Geochemical analysis can also be used to determine if the attenuation mechanisms are stable under ambient and expected future site conditions. For example, Eh-pH stability diagrams may be used to show mineral stability under an expected site-specific range of pH and redox conditions. Stability of attenuated constituents may also be determined by sequential chemical extraction, batch or column leaching tests provided they reasonably simulate aquifer conditions, established (published) stability characteristics of the attenuating solid phase(s), and geochemical modeling.

As previously noted, many of the Tier 3 tests are also useful under Tier 2. With advance planning, Tier 2 testing, particularly the laboratory bench scale tests, can provide data to inform stability determinations.

Tier 4 – MNA Performance Monitoring Program and Contingency Remedies

An MNA performance monitoring program may include sampling for both groundwater and aquifer solids, and begins with a monitoring plan that specifies items such as locations and frequency of sample collection, sample collection methods, field and laboratory parameters for analysis, analytical procedures, and data analysis techniques.

According to the USEPA (2007a), the monitoring program should have a network of wells that provides adequate areal and vertical coverage to verify that the area of groundwater impacts remains static or is shrinking, and the ability to monitor groundwater chemistry in areas where attenuation is occurring.

The MNA effectiveness monitoring program will likely be different from, and perhaps larger than, the detection and assessment monitoring program. Figure 3 shows a hypothetical, but realistic, example of an MNA effectiveness groundwater monitoring network, with the purpose of each well shown on the figure. In addition, periodic collection and analysis of aquifer solids may be considered to confirm the mechanisms, rates, and permanence of attenuation mechanisms as well as the capacity of the aquifer for continued attenuation (USEPA 2007a). Performance monitoring of solids is essentially additional iterations of the investigations performed in the first through third tiers of MNA analysis for confirmation purposes.

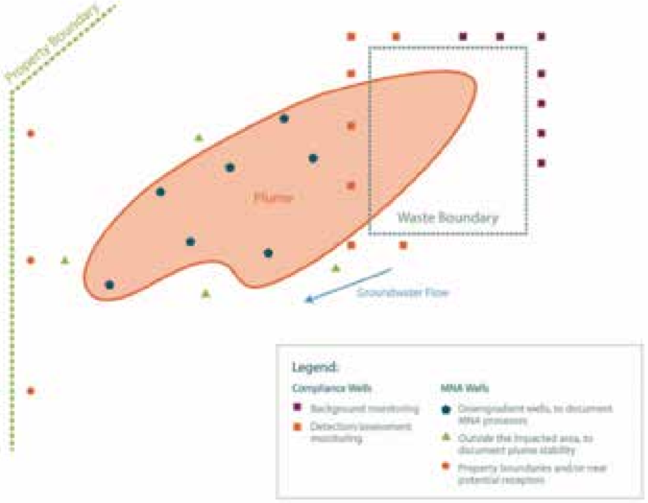

Figure 3 – Example Groundwater Monitoring System for MNA at a CCR Facility (Source: EPRI 2015a)

Developing contingency plans or alternative remedies would be similar to a feasibility study or assessment of corrective measures process, depending on site-specific regulatory requirements. Contingency plans could include additional source control and/or downgradient groundwater remediation in the event that MNA does not meet remedial objectives. The following are some possible corrective measures, which are further described in the EPRI report *Corrective Action for Closed and Closing Ash Ponds* (EPRI 2015b):

- Hydraulic control; i.e., groundwater extraction followed by appropriate management of the extracted groundwater
- Deep well injection (in states where allowed)
- Barrier walls (including slurry walls)
- Grouting
- ISS
- PRBs
- Geochemical manipulations (in situ injections)
  - Sequestration in sulfides under reducing conditions
  - Adsorption to oxyhydroxides under oxidizing conditions
  - Redox manipulation
- Phytoremediation
- Combinations of these, and possibly other technologies

Application of a contingency does not necessarily eliminate MNA from the corrective action plan. The contingency may address one constituent, and/or a small area of the plume, where MNA performance does not meet remedial objectives, while MNA continues for other constituents and/or areas of the plume where performance meets remedial objectives.

OTHER CONSIDERATIONS

MNA of Boron and Arsenic

Boron and arsenic are highlighted here because they represent opposite ends of the MNA spectrum for inorganic constituents in groundwater.

Boron has limited potential for chemical attenuation and is mobile in groundwater under normal aquifer conditions. Additionally, the oxidation state of boron does not change as a function of redox potential in aqueous solutions (EPRI 2004, 2005). Because boron is poorly chemically attenuated in natural groundwater conditions, it can be used as a conservative tracer by which to gauge the attenuation of other constituents of interest (Repert et al. 2006).

Physical attenuation processes (dilution, dispersion, and flushing) will reduce boron concentrations in groundwater after the source is controlled. These processes can be modeled and reductions of boron quantified. Repert et al. (2006) report boron reductions from greater than seven times background to background concentrations due to advection (flushing) in a sand-and-gravel aquifer contaminated with municipal wastewater. There are also examples of physical attenuation due to dilution, dispersion, and flushing for boron at CCR sites, although not described as natural attenuation in the documentation (EPRI 2002, 2001a, 2001b).

Based on a comprehensive literature review and unpublished electric utility experience, more case histories for arsenic attenuation exist than for any of the other trace constituents studied. Iron and arsenic chemistries are closely related, so the iron-attenuating processes generally attenuate arsenic as well. Attenuating mechanisms frequently reported for arsenic include adsorption to, or coprecipitation with, iron oxyhydroxides and iron sulfides (USEPA 2007b). Adsorption to iron oxyhydroxides (e.g., ferrihydrite) generally occurs under oxidizing conditions, and sequestration in sulfides (e.g., iron pyrite) generally occurs under sulfate-reducing conditions.

The Pourbaix (Eh-pH) or stability diagram, such as the one shown for iron in Figure 4, may be useful in determining attenuating mechanisms for arsenic and other constituents that are similarly attenuated by reactions with iron. Geochemical modeling programs, such as PHREEQC and The Geochemist's Workbench®, can create Eh-pH diagrams as well as predict saturation indices for various mineral phases involved in attenuation. In Figure 4, the blue areas show the Eh-pH conditions under which specific iron solids are stable. These iron solids are capable of attenuating arsenic, selenium, and several other trace constituents.

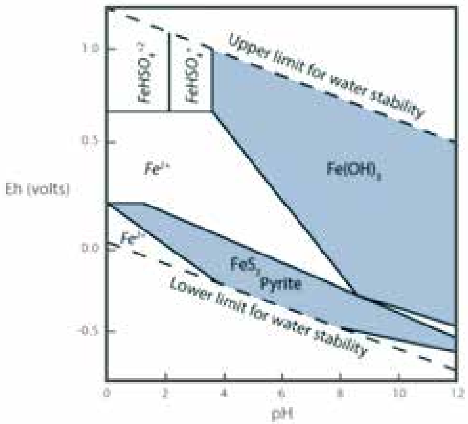

Figure 4 – Eh-pH Stability Diagram for Fe-S-H_2O (Source: EPRI 2015a)

MNA in the Vadose Zone and at the Groundwater-Surface Water Interface

The vadose zone is the unsaturated zone between the land surface and the water table, and it may be extensive in arid climates. Much less work has been performed on natural attenuation in the vadose zone than in the saturated zone (below the water table). In a recent study, Hay et al. (2016) demonstrated that reductive precipitation of selenium was a key mechanism controlling the attenuation of selenium within the vadose zone. Truex and Carroll (2013) point out that attenuation in the vadose zone can significantly decrease impacts to groundwater; they have prepared a framework document for the vadose zone based on the USEPA's four-tier MNA protocol.

Natural attenuation can also occur in the zone of groundwater-surface water mixing, i.e., where groundwater discharges into surface waterbodies (EPRI 2015a). Rapid geochemical changes in this zone can cause precipitation of major elements (e.g., iron and manganese), which then capture trace constituents—such as arsenic, cadmium, chromium, cobalt, copper, nickel, radium, uranium, and zinc—by sorption and/or coprecipitation. Redox potential, pH, and microbial processes often control attenuation in the zone of groundwater-surface water mixing (Gandy et al. 2007; Winde and Van der Walt 2004; Fuller and Harvey 2000; Benner et al. 1995).

Cost Considerations

EPRI (2015a, 2018b) estimated MNA cost based on implementation of all four tiers of the USEPA's protocol. Specific tasks that formed the basis of the cost estimate were determined from a literature review, regulatory guidance documents, and electric utility experience with MNA projects. Many of the MNA cost elements are linked to the number of monitoring wells and number of groundwater and aquifer solids samples. To facilitate a cost estimate, a typical disposal site size, including the number of monitoring wells, was developed based on EPRI (2014).

Unit costs for the various cost elements were obtained by contacting vendors and suppliers, reviewing published literature, and considering unpublished electric utility industry experience. Costs were reported as ranges. Actual costs may vary considerably from this analysis based on site-specific conditions, such as size of the plume, and whether or not all four tiers of USEPA's protocol would be relevant and necessary. For example, EPRI (2018a) outlines a framework for MNA at CCR sites that indicates less testing in Tier 2 and no testing under Tier 3 as appropriate when the constituents subject to MNA are boron, chloride, lithium, and others for which the only applicable attenuation mechanism is physical.

Estimated costs for the initial four-tier analysis ranged from the low to mid hundreds of thousands of dollars (EPRI 2015a, 2018b). Ongoing monitoring and data analysis can drive costs to more than 1 million dollars for a large site over a 30-year period (EPRI 2018b). These numbers are in addition to routine detection and assessment groundwater monitoring costs and reflect that MNA is more costly than detection and assessment groundwater monitoring due to the additional sampling and analysis needed to satisfy Tiers 1 through 3 of the MNA protocol.

MNA Compared to Other Corrective Action Technologies

MNA is compatible with most, if not all, groundwater corrective actions at CCR management facilities. Some level of natural attenuation will likely be occurring at most sites for most constituents, even if it is not recognized and quantified. Therefore, MNA is a logical component of corrective action even if other corrective actions are implemented. At a minimum, MNA can serve as a polishing step (USEPA 2015).

If the constituent plume is static or shrinking, and the source has been abated, then other plume remediation technologies, such as pump and treat, may offer little advantage over MNA. For example, pump and treat for metals and metalloids may reach a point of diminishing returns relatively quickly (after a few months to a few years), as illustrated by the "tailing" or "tail-off" on concentration versus time graphs where the curve begins to flatten with time and often becomes asymptotic to a concentration that may not achieve remedial objectives (Figure 5).

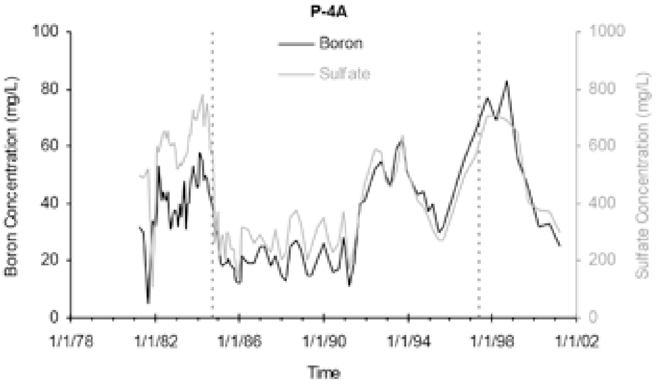

Figure 5 – Concentration in Groundwater During Pump and Treat Remediation, where Pump and Treat Occurs During the Period Between the Dashed Vertical Lines (1985–1997) (Source: EPRI 2001a)

Tailing is widely reported in the literature, and has been attributed to the slow desorption kinetics of constituents from soil or aquifer solids (Bethke and Brady 2000; USEPA 2000), although slow diffusion from strata with low hydraulic conductivity and high porosity may also be a factor (EPRI, 2017). Due to these slow kinetics, pump and treat, on its own, may take decades or may not achieve site remediation objectives and, therefore, offers limited long-term advantages over MNA after high-concentration zones are treated. This is not to say that pump and treat should not be considered; it may still be a valuable component of corrective action for hydraulic control and for removing mass from groundwater.

As discussed in EPRI (2015b), in many cases MNA will be a sustainable option, requiring fewer resources to implement and having a lower carbon footprint than other remedial technologies. Depending on the site, MNA may be the only groundwater remediation alternative needed, it may be used to complement other remediation options, or it may replace another remedial alternative as a polishing step after that alternative has treated most of the mass in groundwater. Regardless of how implemented, application of the tiered MNA protocol represents an increase in effort and cost over detection and assessment groundwater monitoring.

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