

The Need for Sustainable Soil Remediation

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Humanity requires healthy soil in order to flourish. Soil is central to food production, the regulation of greenhouse gases, recreational areas such as parks and sports fields and the creation of an environment pleasing to the eye. But soil is fragile and easily damaged by uninformed management or accidents. One type of damage is contamination by chemicals that provide the lifestyles to which the developed world has become accustomed. Traditional soil “clean-up” has entailed either simple disposal or isolation of contaminated soil. Clearly this is not sustainable. Modern remedial techniques apply mineralogical and geochemical knowledge to clean up contaminated soil and make it good for reuse, rather than simply discarding this precious and finite resource.

KEYWORDS: remediation, contamination, soil, bioremediation, nanoparticles, phytoremediation, adsorption, precipitation

INTRODUCTION

Perhaps uniquely in the animal kingdom, humans have the tendency to damage or destroy the environment upon which they are reliant for survival. One of the many good examples of this is our past attitude to soil. Early, pre-industrial approaches to managing waste materials were mixed. Useful “waste materials” were carefully recycled; for example, animal wastes were added to soil as fertiliser. Other wastes were simply discarded and left to rot or be preserved for the delectation and delight of future archaeologists. As society developed and became industrialised, so our capacity to damage our natural resources increased, and we generated more and more wastes which were not recycled as useful materials. Careless discarding of “rubbish” allied to accidental release of industrial products has left a legacy of contaminated land.

Back in the 1970s a series of high-profile cases across the globe led to an increasing awareness of the issues related to soil contamination. The Love Canal neighbourhood in New York State, USA, was used as a chemical dumping ground and subsequently built on (Engelhaupt 2008). After discovery of the contamination problem the local community was evacuated. Contaminated material was removed from the site and incinerated. A specialised drainage system was installed to deal with contaminated water draining through the site. The site was lined along its base to stop this drainage from spreading further, the top of the site was covered by a protective layer or cap to isolate the site and the whole area (about 40 acres or 16 hectares of land) was fenced off. Parts of the site remain fenced off today (Fig. 1). At Times Beach in Missouri, USA, dioxin-contaminated oil was sprayed on roads to control dust (Yanders 1986; US EPA 2010a). After identification of the problem

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FIGURE 1 More than 30 years after the evacuation of the community, parts of the Love Canal site are still fenced off due to potential hazards. Image taken from the cover of *Environmental Science & Technology*, November 2008, volume 42, issue 22, USED WITH PERMISSION

the community was evacuated and about 19,600 m³ of soil were incinerated. Parts of the town of Lekkerkerk in the Netherlands were built over ditches containing, amongst other contaminants, aromatic hydrocarbons (US EPA 1992). Almost 94,000 m³ of soil were removed and incinerated. In all these cases the costs of remediation were in the order of tens to hundreds of millions of US dollars.

These cases and others led to an increased awareness of health issues related to the presence of contaminants in soils. As a result, legislation has been adopted that seeks to protect both humans and the environment. Inevitably legislation varies among jurisdictions, but some common strands exist.

1. Initial legislation often focussed on getting rid of contaminants completely or reducing concentrations to a fixed value.
2. Over time legislation has become more sophisticated, due in part to cost issues related to soil clean-up but also to a better appreciation of how to quantify the risks associated with contamination; permissible concentrations of contaminants in soil below which no remediation is deemed necessary now vary depending on land use.
3. There is a requirement at some administrative level to identify contaminated sites, as well as those responsible for the contamination; it is these people who should pay for the clean-up.

WHAT IS CONTAMINATED LAND AND HOW MUCH OF IT IS THERE?

Formal definitions of what constitutes contaminated land have been developed as part of the legislative process in many countries. The definition used in the UK is typical of many and defines contaminated land as “any land which appears to the local authority in whose area it is situated to be in such a condition, by reason of substances in, on or under the land that:

- significant harm is being caused or there is a significant possibility of such harm being caused; or
- significant pollution of the water environment is being caused or there is a significant possibility of such pollution being caused.”

Harm is defined as “harm to the health of living organisms or other interference with the ecological systems of which they form a part, and in the case of man includes harm to his property” (DEFRA 2006).

The important point in this and similar definitions is that contamination is not defined by the presence of contaminants but by the potential of the contaminants to cause harm. The definition encapsulates the “source–pathway–receptor” concept that is widely used to determine whether a site is contaminated or not (Fig. 2) (Nathanail and Bardos 2004). In this framework the contaminated soil represents the source of contamination, but this is only regarded as a problem if the contaminant can reach a target (the receptor). The means by which the contaminant reaches the receptor is the pathway.

Within this framework it becomes clear that to resolve the contamination issues at any particular site it need not be necessary to remove the contaminant source. Rather it is necessary to break one of the linkages in the source–pathway–receptor model. The adoption of the source–pathway–receptor method of assessing risk and deriving remedial solutions, linked with the sustainability agenda, has led to the development of the remedial methods discussed in this issue of *Elements*.

Following political acceptance that contaminated soil poses a threat to humans and the environment, and with the development of legislation, many countries have tried to quantify the extent of the contaminated land legacy with which they will have to deal. This has been no easy task as by their very nature sites with harmful levels of contaminants due to accidents or careless management are often poorly documented.

In the UK contaminated land is regulated by the Environmental Protection Act 1990, part 2A (DEFRA 2006). The Environment Agency (EA) assessed industries that



FIGURE 2 A typical example of source–pathway–receptor linkages: soil at a contaminated site (the source) reaches humans (the receptor) via hand-to-mouth incidents (pathway) when people put their dirty hands in their mouths. Another example is the uptake of contaminants by roots (pathway) into vegetables (receptor) grown in contaminated soil (source) and the subsequent consumption (pathway) of the vegetables (source) by humans (receptor). PHOTO: iSTOCK

could potentially generate contaminants. According to their calculations, around 325,000 sites (~300,000 ha) have had some form of current or previous use that could have led to contamination (EA 2009). To put this in context, the area of England, Wales and Northern Ireland that falls within the remit of the Environment Agency is approximately 165,000 km², so about 2% is potentially contaminated. However, until all these sites are investigated, the true extent of contamination will not be known. To date the EA calculates that about 33,500 sites have been identified as contaminated and of these about 21,000 have required treatment. Overall, inorganic contaminants are found far more commonly than organic contaminants, possibly due to the potential for organic contaminants to degrade naturally over time (Fig. 3).

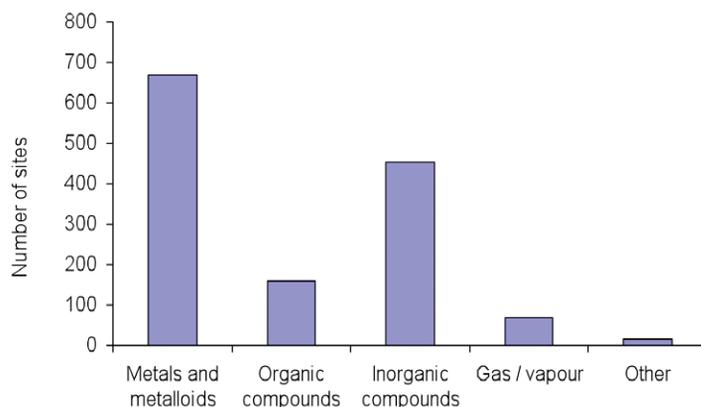


FIGURE 3 Main contaminants found at reported contaminated land sites in England and Wales, 2007. More than one contaminant can occur at an individual site. Data from the Environment Agency (www.environment-agency.gov.uk/research/library/data/58782.aspx, accessed December 11, 2009)

In the USA two key pieces of legislation cover contaminated soils (LaGrega et al. 2001). Sites contaminated by hazardous waste due to past activities are covered by the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA), passed in 1980, and the Superfund Amendments and Reauthorization Act (SARA) of 1986. Sites currently being contaminated or contaminated in the past by still-current activities are covered by the Resource Conservation and Recovery Act (RCRA) of 1976 and subsequent amendments. The United States Environmental Protection Agency (US EPA) is responsible for enforcing these acts. The US EPA states that the total number or extent of contaminated sites is unknown but lists 3746 sites expected to require action under the RCRA legislation by 2020 (US EPA 2010b). As of November 2010, 347 sites had remedial actions completed under the Superfund programme, another 1280 are listed as requiring remediation or being subject to ongoing remediation, and another 62 are being considered for addition to the Superfund list (US EPA 2010c).

In Germany contaminated land is covered by the Bundesbodenschutzgesetz (the BbodSchG, or Federal Soil Protection Act) and the Bundes-Bodenschutz- und Altlastenverordnung (the BBodSchV, or Federal Soil Protection and Contaminated Sites Ordinance), which became law in 1999 and require the government to locate and remediate contaminated soil. By 2000 the German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety had identified about 360,000 sites that were potentially contaminated, but stressed that these sites may not be contaminated and that the area of each site was not known (BMU 2002).

The Dutch Ministry of Housing, Spatial Planning and the Environment (VROM) (now part of the Ministry of Infrastructure and Environment) estimates that “quite a significant part of Dutch soil is polluted” (VROM 2009). It states that some 60,000 sites could be in urgent need of clean-up and in 1997 estimated that costs incurred during remediation of all Dutch contaminated sites would be about 100 billion Dutch guilders (about 500 million euro at current hypothetical exchange rates). The Danish Ministry of the Environment estimates that about 40,000 sites are contaminated in Denmark due to former or current industrial activities (Danish EPA 2009).

Clearly contaminated land is an international problem, with significant areas affected in many different countries. The following methods of remediation are among the most commonly used internationally.

CURRENTLY POPULAR REMEDIATION METHODS

Remediation methods are carried out either *in situ*, with the contaminated soil remaining in place, or *ex situ*, where the contaminated soil is excavated. Once the soil is excavated, remediation may occur either on or off site. There is always the possibility of returning remediated soil back to its site of origin. Alternatively, once excavated, material may be removed off site and disposed of (most commonly in a landfill). All these methods remove the source from the source–pathway–receptor linkage. An additional method of remediation is simply to isolate the contaminant source, thereby breaking the pathway from the contaminant source to the receptor (Nathanail and Bardos 2004). Although the contaminant is still present, under the UK definition for example, the site would no longer be termed “contaminated” as there is now no threat of harm to humans or the environment.

Perhaps the most common and popular remedial method is that known as “dig and dump”. In simple terms the contaminated soil is dug up, removed from the site, and stored elsewhere, usually in a landfill site (Fig. 4). Depending



FIGURE 4 A landfill site under construction near Quito, Ecuador, where waste materials, including contaminated soil, can be disposed of. Landfills have to be sited sufficiently close to industry and residential areas that it is economic to transport waste to them, and sufficiently far away that local residents do not complain about noise, smell, visual appearance of the landfill and excess traffic. Other considerations also apply; for example it is good practise to exploit existing holes in the ground rather than create new ones and, in order to reduce potential leakage of liquids from the landfill and subsequent pollution events, the landfill should be underlain by impermeable rocks or sediments (such as clays) and not sited over an aquifer. PHOTO: iStock

on development needs, the excavated site is either left as a hole or is filled in with clean material, for example, demolition debris.

The attraction of this technique lies in its simplicity, ease of costing, speed and finality. All contaminated soil can be quickly taken away from the site, leaving it “clean” with no need for future monitoring. This method was particularly attractive when assessment of contaminated soil was carried out by comparing contaminant concentrations to a fixed, legislatively permitted concentration in the soil, rather than on a risk-management basis. However there are important limitations to “dig and dump”. There are practical constraints to the depth of excavation, and the sides of excavations need to be shored up. Also excavation in saturated soils or below the water table presents engineering challenges, and, unless a hole is required at the remediated site, some form of fill has to be found to replace the excavated material. However, perhaps the biggest drawback to “dig and dump” is the requirement that a site has to be found for disposal of the contaminated soil. There is growing political pressure, for example in Europe through the European Union (EU) Landfill Directive (EU 1999), to reduce the amount of contaminated soil disposed of as waste. Not only are the costs of such disposal being increased to reduce the practise but also legislation is being enacted to control the forms of waste that landfills can accept, thereby reducing the number of landfills available for contaminated soil disposal. Whilst excavation remains a popular remedial measure, there is now far more political and economic pressure to treat the excavated material and return it to the original site or to use an alternative remediation technique.



FIGURE 5 Impermeable, black sheeting forming part of the basal lining of a landfill site designed to prevent leakage of contaminant-bearing fluids from the bottom of the landfill. The landfill is located in Hungary (South-Transdanubien region) between the villages of Görcsöny and Baksa; construction began in 2008. Typically the basal lining of a landfill site will comprise a series of layers. The uppermost layer of the lining, on which the waste rests, is usually some form of geotextile, i.e. a fabricated material, or graded sand that acts as a filter, permitting the drainage of leachate from the waste but preventing particle transport. This filter layer rests on top of a more porous, free-draining layer that houses a series of collection pipes designed to draw any leachate from the base of the landfill and carry it up to the surface for treatment. The bottom of the free-draining layer takes the form of an impermeable membrane of the type shown in the photo. There may then be a further free-draining layer with collection pipes, underlain by a second impermeable layer as a fail safe. These lining systems are expected to last for the lifetime of the landfill, from the time it receives its first wastes to a time after the landfill is full and no longer in operation. At this time, liquids draining through the contents of the landfill should have achieved a composition similar to that of “normal” water, so that any leakage from the landfill has no detectable impact on soil water or groundwater composition; this can take tens to hundreds of years. Underlying the lining systems, in the ideal situation, is a bed of naturally occurring, impermeable clay. PHOTO: ZSOLT BICZÓ, iStock

Another common and popular remedial method is containment. Containment can be applied off site to excavated materials (i.e. the “dump” end of “dig and dump”), on site to excavated materials, or in situ. The significant advantages of containment are that it is relatively cheap and rapid. The most common methods of containment are the installation of liner systems that form a coating or lining to the holes used for the storage or disposal of contaminated materials (FIG. 5), layers that cover or cap contaminated sites (FIG. 6) and in situ vertical and horizontal barriers (FIG. 7).

Linings typically comprise a series of layers each fulfilling a specific purpose. Examples are impermeable barriers to prevent fluid escape; sorptive layers to reduce contaminant



FIGURE 6 A natural cover layer over the Reichs Ford Road landfill site, Frederick, Maryland, USA (www.frederickcountymd.gov/index.aspx?NID=530, accessed November 11, 2010). The tap in the foreground is part of the system for monitoring and releasing any gas that builds up in the landfill. Once a landfill is “full”, the story is not yet over. A cap is placed on the landfill similar in design to the lining systems at the bottom of the landfill. Such caps are designed to isolate the waste in the landfill from the natural environment for visual and safety reasons. The cap is usually covered by a layer of topsoil that can be several metres thick, which is then planted with grass or other vegetation to improve the visual appearance of the landfill site. Although no further waste is being added to the site, the landfill is not “finished”. In particular bacteria will be actively degrading any organic waste present in the landfill. Bacterial degradation of the waste generates various gases, including carbon dioxide, methane and hydrogen. It is important that these latter two gases do not build up in landfills as they can be explosive. Thus after a landfill is “full” it is necessary to continually monitor the evolution of gas from the site and control its release. PHOTO: JOHN KIETH, iSTOCK

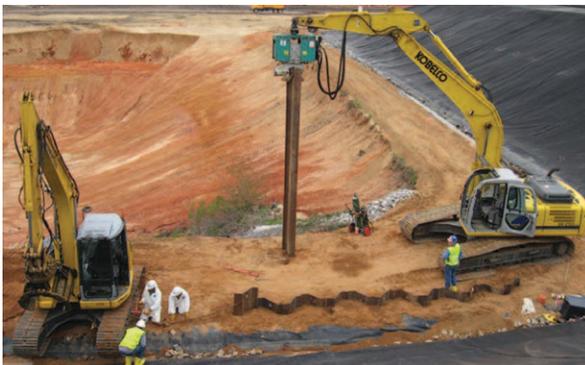


FIGURE 7 Vertical sheet piling being inserted at a contaminated site to create a containment cell that isolates contaminated soil. The Escambia Wood Treating Company was located in Pensacola, USA. Prior to going into bankruptcy in 1991, this site had been used for 40 years for treating wood products with creosote and pentachlorophenol. The site was placed on the US EPA National Priorities List in 1995. A variety of organic contaminants were detected at the site. As part of the remedial process, a “containment cell” was constructed for the disposal of about 500,000 m³ of treated, contaminated soil (US EPA 2010c). PHOTO: ESCAMBIA TREATING COMPANY CLEANUP (www.etccleanup.org/, accessed November 10, 2010)

movement; coarse, porous layers to promote fluid collection and removal; and strengthening layers to prevent damage to the lining. As well as synthetic materials, natural materials such as bentonites and zeolites may be used in such layers. Cover layers are usually composite constructions, comprising impermeable layers to isolate the contaminated soil from percolating rainwater from above, drainage layers to direct infiltrating rainwater away from the contaminated site, and vegetated soil layers. In situ, vertical and horizontal barriers may be produced by inserting sheeting – for example, overlapping lengths of steel sheet piling; by excavating and infilling, for example, with a slurry that subsequently solidifies; or by manipulation of the soil – for example, by freezing to produce a cryogenic barrier. Vertical containment systems are more common than horizontal ones due primarily to the relative costs of the associated engineering issues related to their construction.

WHY BOTHER TO PROTECT THE SOIL?

If a suite of contamination-remediation techniques exists, why is there a need for new methods? The reason is that the above methods are not sustainable. The concept of sustainability has gained widespread acceptance since its use in the Brundtland report from the United Nations’ World Commission on Environment and Development (WCED), which defined sustainable development as “development that meets the needs of the present without compromising the ability of future generations to meet their own needs” (WCED 1987). Sustainability is commonly stated to have “three pillars”: environmental, social and economic demands (FIG. 8).

Applied to remediation, this means that any remedial treatment should achieve a balance between protecting the environment now and not limiting use of the environment in the future, acceptance by the general population, and not being too expensive. None of the methods outlined above could truly be labelled sustainable. For example, “dig and dump” relies on an infinite provision of holes to put contaminated material in and clean material to fill up voids. In many senses this is not remediation but merely moving contamination from one site to another. Similarly, although containment breaks pathways, the contaminants remain in place and the soil is still not useable by future generations. This is important because soil is a finite resource. The world’s population is growing and much of the food that feeds this population is grown in soil. Soil also stores a vast amount of carbon and sustains the majority of the planet’s animal and plant life. Just as importantly for people’s quality of life, soil supports parks and other recreational areas. However, we are losing soil at a rate estimated at approximately 11.6 ton ha⁻¹ y⁻¹, equivalent to a reduction in soil thickness of about 0.38 mm y⁻¹ (Yang et al. 2003). Estimating the production rate of soil is diffi-

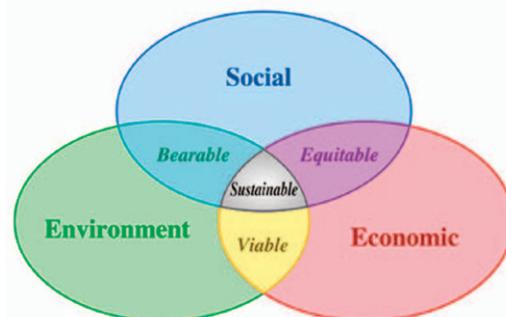


FIGURE 8 The three pillars of sustainability. FIGURE BY JOHANN DRÉO (http://en.wikipedia.org/wiki/File:Sustainable_development.svg#file)

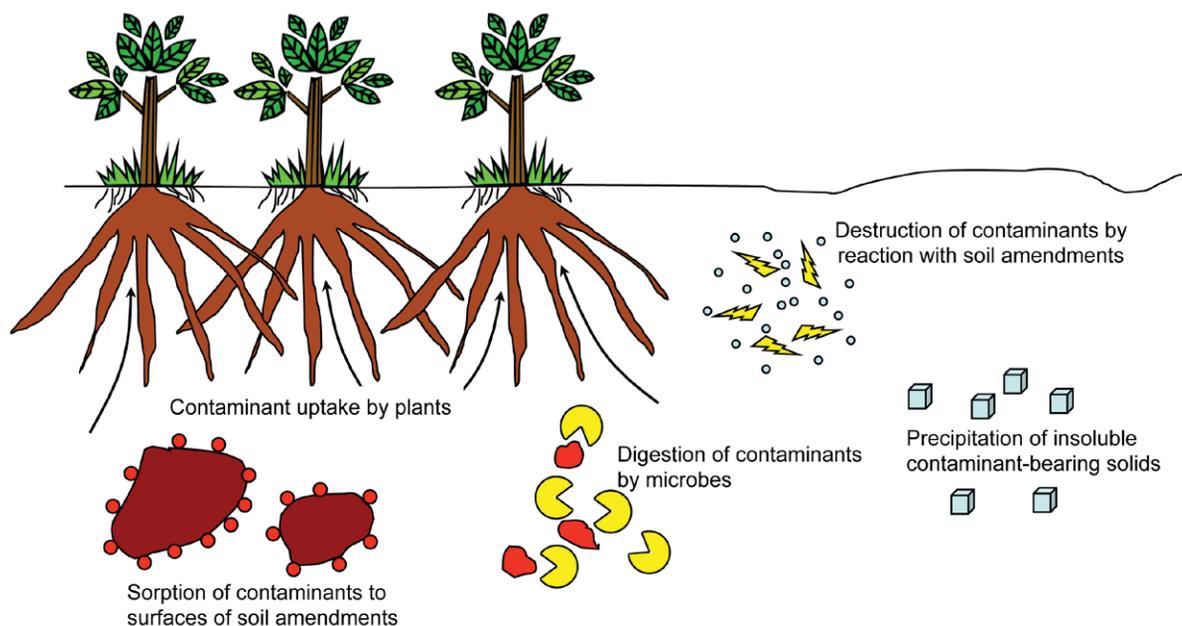


FIGURE 9 A selection of methods employed in the sustainable remediation of contaminated soil

cult. However, as it is thought that about 60% of soil erosion is induced by human activity and if we assume that, without human intervention, soil production and erosion would be in some sort of steady state, at least on human timescales, it seems likely that we are depleting the soil resource. Not only should we protect the soil we have but we should bring soil we have previously damaged back into beneficial use, thereby repairing the damage caused by society's previous activities. Disposing of contaminated soil in a hole and sealing it off, or just covering up contaminated soil to isolate it from the environment, does not fulfill this aim.

SUSTAINABLE REMEDIATION

The remedial methods detailed in this issue demonstrate how mineralogical and geochemical principles are allowing soils to be remediated and reused in a sustainable fashion (Fig. 9).

The approaches of these methods to inorganic and organic contaminants are somewhat different. The sustainable remediation of inorganic contaminants usually involves breaking the pathway between the source and the receptor. Soil chemistry is manipulated so that, despite the contaminants remaining in the soil, they become immobilised and no longer pose a threat. The chemical processes that these techniques rely on are precipitation and sorption reactions (Jones and Healey 2010 this issue; O'Day and Vlassopoulos 2010 this issue). Either contaminants precipitate out of solution (after, for example, changes in soil pH, oxidation state or the concentration of potential reactants), or they are removed from solution by attaching to the surface of materials, such as clays, zeolites and organic material, via adsorption.

Alternatively, the contaminants can be taken out of the soil altogether, i.e. the source is removed. One means of source removal is phytoextraction – the use of plants to extract contaminant metals from soils. In the ideal end scenario, metal-laden plants become an exploitable metal source, though the usual result is that the plants are just used to concentrate the contaminants for ease of disposal. The problem with phytoextraction is getting a sufficiently high concentration of contaminants into plants of sufficient biomass. High concentrations of contaminants are one thing. However, the plants that typically accumulate

high concentrations of metals (i.e. hyperaccumulator plants) are so tiny and so slow growing that they do not remove large quantities of contaminants. Large, fast-growing plants that have a high concentration of metals are required. This has led to the development of assisted phytoextraction, in which soil chemistry is manipulated to help fast-growing plants extract high concentrations of contaminants (Tack and Meers 2010 this issue; Hodson and Donner 2011).

Although the above methods can also be applied to organic contaminants, an important difference between inorganic and organic contaminants is that the latter can often be degraded or broken down, particularly by bacteria, to simple oxides such as water and carbon dioxide. This occurs naturally over time and is accelerated in bioremediation by manipulating conditions to make them favourable for bacterial digestion (Antizar-Ladislao 2010 this issue). Another way to remove organic contaminants is to oxidise them. In many ways this is the abiotic equivalent of bioremediation. The nanoparticle revolution has made possible the production of particles both sufficiently small to mix well with contaminated soil and reactive enough to degrade organic contaminants. Although still in its infancy, the use of nanoparticles in remedial treatments is now being reported (Mueller and Nowack 2010 this issue). Thus sustainable remediation of organic contamination works through removing the contaminant source.

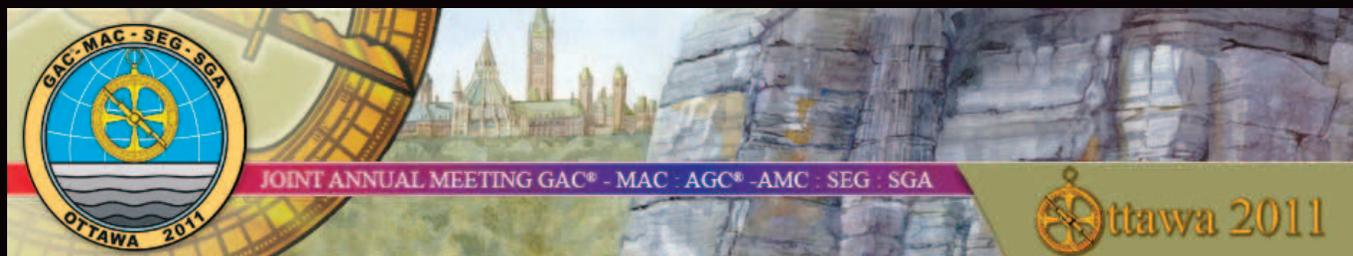
In all cases the end result of sustainable remediation is either the removal of the contaminant source or the immobilisation of the contaminants, so that the soil can be used once again for the benefit of society.

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