REMEDIATION OF CONTAMINATED SOILS WITH GREEN PLANTS: AN OVERVIEW¹

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SUMMARY

Billions of dollars each year are spent on the remediation of contaminated soils in the United States alone. Contaminated soils represent an economic liability as well as a technical challenge. New technologies are needed to address numerous contaminants, especially those that are neither volatile nor mobile in soil solutions. One emerging technology, "phytore-mediation", employs green plants in the remediation process. The technique is relatively new, with few field demonstrations; however, it represents an ever-growing area of research built on a sound technical basis. This technology draws heavily from a wide range of agronomic, biological, and engineering disciplines. Exploiting all plant-influenced biological, microbial, chemical, and physical processes to remediate contaminated sites is the goal of much research in this area. In certain situations, sites remediated with a plant- based technology are expected to have significant economic, aesthetic, and technical advantages over traditional engineering solutions. This paper provides an overview of the phytoremediation area with an emphasis on providing background information and research avenues to plant biologists.

Key words: hydrocarbons; heavy metals; pollution; metabolism; detoxification.

INTRODUCTION

The remediation of contaminated soils in the United States is a multibillion dollar a year industry. Most often sites are remediated through a wide variety of engineering-based technologies that have evolved over the last 3 decades. These technologies can be grouped into two broad categories: a) isolation and containment techniques, and b) decontamination techniques.

Isolation and containment techniques exploit physical, chemical, and hydraulic barriers to isolate the pollutant and prevent its escape. This remediation strategy offers no actual reduction in the quantity of pollutant on a particular site, but the risk of the pollutant causing further environmental damage is reduced. Examples of containment techniques include vaults, caps, and hydraulic isolation curtains, as well as physical absorption or entrapment of the pollutant into a stable matrix (e.g., cement). In contrast, decontamination techniques reduce the total quantity of the contaminant at the site. Site decontamination permits increased flexibility in future land use decisions. Examples of decontamination techniques include soil washing, vapor extraction, and microbial bioremediation. One common site decontamination technique is the excavation of the hazardous material and disposal to a secure landfill. This strategy, despite its unsophistication and relatively high cost is often favored by the remediation engineer for smaller sites. It is dependable, leaves a clean site, and has definitive starting and end points. On the downside, it represents a transfer of the pollutant to a second location, and questions of residual liability linger. In addition, the siting of new secured and hazardous waste landfill operations is becoming increasingly more difficult.

The development of a site remediation strategy involves balancing legal, physical, chemical, biological, and economic considerations. Sites are often complex, with numerous environmental concerns. Philosophically, many site managers would prefer that the treatment process leave their site in as pristine a condition as possible. This is not always technically feasible. The chemical and physical properties of certain hazardous wastes sometimes preclude all current site decontamination techniques except for excavation and subsequent reburial. Seeming plausible solutions on closer examination result in transferring the pollutant from one medium to another with resulting increases in volume and complexity of the material to be treated.

There is often a legal, economic, and aesthetic advantage to remediating a site with minimal surface disruption. With certain contaminants and site conditions (e.g., underground storage tanks for gasoline) this "in situ" remediation is common. Soils with relatively immobile contaminants and tight soil structure, however, pose a technical challenge to all in situ techniques. Engineering technologies are being explored (e.g., electroosmosis, soil fracturing, thermal decomposition, and surfactant washing), but in many cases will be cumbersome and costly. Plant-based systems seem to be an interesting, cost-effective alternative that poses an exciting technical challenge to the research community.

Phytoremediation is defined as the use of green plants to remove, contain, or render harmless environmental contaminants. This definition applies to all plant- influenced biological, chemical, and physical processes that aid in remediation of contaminated substrates. The concept itself is not new. The use of plants in waste water treatment schemes is over 300 yr old (16). Plant-based remediation

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is also evolving in the control of both indoor air pollution (25), and urban smog (34). This paper, however, will confine itself to the latest media to be targeted by plant-based remediation methods: contaminated soils, sludges, and sediments.

These authors have explored the vegetation on dozens of sites where soil has been degraded by manufacturing, mining, and disposal activities. The ability of plants to survive in soils declared legally "hazardous" has proven to be impressive. Many people unfamiliar with hazardous waste sites imagine such sites to be barren. In truth, when they are devoid of vegetation, most have active "vegetation management" strategies (herbicide applications, stone mulch, etc.) to control vegetation. Many soils legally classified as "hazardous" revegetate rapidly when removed from these vegetation management schemes. Not all sites, however, revegetate as rapidly. Certain sites have pH, texture, ionic, and nutrient limitations that need to be altered before the establishment of a vegetative cover. In most cases, common agronomic practices can be utilized.

PLANTS AS ENGINEERING STRUCTURES

Most site remediation personnel view vegetation as "debris", both in a legal and technical sense. Many industrial site managers have engineering backgrounds and are unfamiliar with the physics and chemistry of plants. We have found that redefining green plants in engineering parlance has been useful in describing the potential use of plants in remediation to this audience. A green plant is a "solar-driven, pumping, and filtering system that has measurable loading, degradative, and fouling capacity" (11). Roots are "exploratory, liquid-phase extractors that can find, alter and/or translocate elements and compounds against large chemical gradients". The internal and external surface of many plant parts are also home to microbial communities that can be exploited. Root surfaces maintain active microbial biofilms. These and a root's mycorrhizal extensions into the soil significantly augment soil-surface contact and increase the plants own somewhat meager metabolic capacities. The exploitation of these rhizosphere communities to remediate soil contaminants is an active area of research at numerous laboratories. Parallels between the rhizosphere and current engineering practices are numerous and the concept is familiar to many in the field. Degradation rates of certain xenobiotics can often be increased by the addition of exogenous carbon sources and encouraging microbial growth (composting and bioaugmentation). Additionally, many remediation managers are aware of fungal inoculants (e.g., whiterot fungi) being marketed for the in situ destruction of relatively immobile, soil-bound organics such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs). When plants are redefined in such terms, both the biologist and remediation engineer can contribute to research in the area. Engineering modeling studies have provided research goals in terms of rooting structure, patterns, water use, transpiration, and metabolism. Meeting these goals by the selection, creation, and cultivation of plants and their associated microflora is an exciting technical challenge.

Phytoremediation is a relatively new concept; however, techniques, skills, and theories from well-established fields are easily transferable. The concept requires a new paradigm. Traditionally we think of crops as productive, a source of food, fiber, and fuel. Plants as destructive or absorptive entities requires rethinking their agronomy, toxicology, biochemistry, microbiology, and molecular biology. The process is both an interesting exercise as well as a source of research leads. Certain microbial and plant engineering strategies that have been considered commercial failures in the past due to yield decreases of as little as 5% (e.g., xylem endophytes), may now have renewed promise. A yield decrease of 50% or more may be acceptable if it were to achieve maximum site decontamination.

PLANTS IN CONTAINMENT STRATEGIES

The capacity of plants to transpire large quantities of water is significant and has been exploited in the dewatering of sludge (27), prevention of downward water flux through landfill caps (22), and containment of contaminated water down gradient of a problem site (29). Certain sites, although polluted, do not pose an obvious environmental risk unless there is off-site migration into a waterway. Under authority of the Clean Water Act, these sites may then be of regulatory concern by both state and federal agencies. Pollutants slowing leaching from these soils into a shallow aquifer and then subsequently into a small stream have been targeted by this root-intercept strategy.

Plants have been integrated into engineering technologies in erosion control and are used commonly to maintain the integrity of caps, ditches, and berms. In numerous remediations, as the operator moves toward "closure", a site erosion plan often specifies vegetative covers. Particularly hardy grasses are also sold for revegetating the banks of some polluted ditches as an interim "corrective action" measure. On at least one site plants have been used to stabilize the actual contaminated soil matrix after the metal contaminates had been chemically stabilized into the matrix by the addition of soil amendments (24). Plant roots scavange available metals but there is little movement from root to shoot. The site is visually impressive and the "before" and "after" results are remarkable.

The ability of many wetland plants to alter the pH around their roots, provide oxygen into the anaerobic zone, and prevent stream bed erosion is being explored for the remediation of seeps from old mining and landfill operations (33). These mine and landfill seeps are often extremely acidic and carry significant heavy-metal concentrations. In the wetland environment where the biotic activity increases alkalinity, sulfide ion concentration, and organic residues, the metal ions precipitate out and the water is released to the stream outflows.

PLANTS IN DECONTAMINATION STRATEGIES

Plants absorb both organic and inorganic contaminants from soils. Absorption, sequestration, and metabolic transformations of these pollutants are possible and potentially exploitable to clean contaminated soil. Not all pollutants and matrices are possible. Due to the perceived limitations in rooting depth and time requirements, most researchers in the field currently target relatively non-leachable contaminants that pose little eminent risk to health or the environment. These restrictions are not as formidable nor exclusive as they may first sound; however they should be remembered when considering the usefulness of a given demonstration system to field application.

Plants have a long history of use as reed bed (14), wetlands (31), and overland flow (30) for the polishing of waste water. Their use in remediation of soils is more recent; however, reports of pesticide spill clean-up (12), degradation of polycyclic aromatic hydrocarbons (PAHs) (5), chlorinated solvents (1), DDT (23), dioxanes (6),

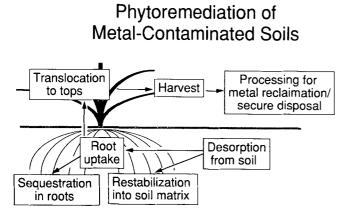


FIG. 1. Scheme for the application of phytoremediation technology to metal-contaminated soils.

phenols (7), and aiding in vapor extraction are proliferating. Reports of successful metal remediation are more rare (2), but its suggestion is more than a decade old (32) and progress is being made in a number of different laboratories (10).

PLANTS AND INORGANIC CONTAMINANTS

Most sites legally classified as "hazardous" due to their metal ion contaminants contain concentrations and compositions of metals duplicated in natural settings. Soils formed on the outcroppings of many ore bodies can greatly exceed the legal "hazardous" limits. Many of these natural soils, like their anthropogenic equivalents, support a wide variety of flora. In most cases, plants growing on these sites do not accumulate significant quantities of metal in the above-ground biomass. Some plants, however, accumulate small amounts of metals in their tissues, indicative of the soil concentrations below. These plants have been used by the mining community for prospecting for at least 70 yr (19). These relatively minor amounts, however, are not sufficient to decontaminate a soil by harvesting the tissue and subsequent processing. To provide an effective soil decontamination solution, we believe we will have to find, breed, or engineer plants that absorb, translocate, and tolerate levels of heavy metals in the 1 to 2% range. We have limited our thinking to the harvest of the above-ground biomass due to the ease of harvesting and our desire to minimize worker and community exposure to dust and debris. Other possible solutions may exist in the harvest of roots where these constraints are not relevant. Such a scheme is shown in Fig. 1.

These metal-removal goals are ambitious, paralleling removal rates of plant nutrients such as nitrogen, potassium, and calcium. Heavy metals are often toxic because they interfere with normal cell processes. The discovery or development of plants capable of containing greater than 2% metal might be assumed to be infeasible if it were not for the existence of a small group of plants called hyperaccumulators (3). These naturally occurring plants can have spectacular metal-uptake capacities. The sap of one tropical tree growing on a nickel outcropping has been reported to have concentrations of Ni in excess of 25% dry weight (4). Other plants have been reported to have concentrations in excess of 1% Cu and Co and 3% Zn, Ni, and Mn on a dry weight basis (28). Lead (Pb) levels, although lower, have been reported as high as 8200 ppm in one plant (28). Hyperaccumulators are being examined for potential remediation uses; however, due to their generally low growth habits and small biomass they are agronomically unsuited for phytoremediation. Nevertheless, these plants are a valuable store of genetic and physiologic material and data. They also indicate the prioritization of metals most likely to be remediated with this technology. Transferring and extending these metal-accumulating, translocating, and tolerance capacities to a plant with better agronomic characteristics may provide the ideal solution to the clean-up of metal contaminated soils. Modern molecular biology may help, but there is much exploratory biology needed in this area in general.

This accumulation of massive amounts of metal into plant tissue has been referred to as "biomining". In some instances, if successful, it might eventually provide not only solutions to remediation problems but new dimensions to the mining of certain metals. The "bio-ore" product contains both metal and the fuel for its own smelting (Fig. 2).

Lead is perhaps the metal contaminant of largest environmental concern and largest area/volume of impact. Unfortunately Pb is one of the most difficult cations to be accumulated by plants. Parallel to our efforts with hyperaccumulators, we have explored lead (Pb)-contaminated superfund, mining, and other industrial sites for plants that accumulate Pb. Our goal in this case is to find, manipulate, and extend the Pb-uptake limits of these plants. We have analyzed many plants from these sites in search of appropriate germplasm. Of the plants we have analyzed to date, two have shown significant and unexpected abilities to accumulate lead: hemp dogbane (*Apocynum* sp.) and common ragweed (*Ambrosia* sp.). Their lead accumulation abilities, however, are dependent on the chemistry of the soil in which they are growing. Most metals, and lead in particular, have numerous forms in the soil, not all of which are equally available for plant uptake.

Maximum lead removal requires balancing plant-nutritional requirements for biomass production and the bioavailability of lead for uptake by plants. Maximizing lead availability requires a lower pH and low levels of available phosphate and sulfate. Limiting the fertility of the soil in this manner directly impacts plant health and vigor. Some of our current research effort is directed at managing the plant nutritional status of the soil relative to the lead bioavailability status to maximize total metal removal.

Post-Harvest Processing of Metal-laden Biomass

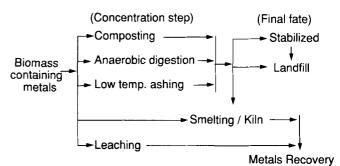


FIG. 2. Potential methods for handling metal-laden biomass.

Targeting Phytoremediation of Organics

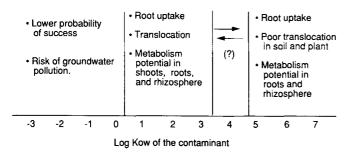


FIG. 3. The role of the octanol-water partitioning coefficient $(K_{\sf ow})$ in phytoremediation of soil organics.

PLANTS AND ORGANIC CONTAMINANTS

Much of our knowledge about the behavior of organic compounds in plant-soil system is derived from the study, development, and registration of soil-applied pesticides. Models have been developed for these systems based on measurable physical and chemical parameters (15). Using these models, we can make rough judgements as to the applicability of phytoremediation to specific organic contaminants and specific soil types. The most common parameter used in the pesticide industry to predict plant uptake from the soil is the octanol-water partitioning coefficient (K_{ow}). Contaminants with a log K_{ow} (≤ 1) are considered very water soluble and would be predicted to cause ground-water contamination under many climatic conditions (Fig. 3). Some sites or remediation projects may have hydrologic containment systems or rainfall:evapotranspiration ratios that might reduce the risk, but plant roots do not generally accumulate these water-soluble compounds at a rate surpassing passive influx in the transpiration stream. In some cases this can be substantial; however these factors suggest these compounds are not appropriate targets for phytoremediation. Compounds with low log K_{ow} (≤ 1) can be accumulated in plants and many are generally considered mobile in both plant xylem and phloem. Pollutants with intermediate log K_{ow}'s (approximately 1 to 4) are taken up by roots and are considered zylem mobile but generally phloem immobile unless chemically modified by the plant. Compounds in this range would be expected to be good targets for phytoremediation, and the list of priority pollutants that fall in this range is extensive. Again, some of these compounds would be expected to cause ground-water problems, and suitability is dependent on soil type, rainfall, and how the material entered the environment. Compounds that are denser than water, have lower log Kow, and came from point sources (e.g., leaking drums) would tend to have vertical concentration profiles in the soil and make them more difficult for plant root treatment without excavation. Compounds with log Kow's greater than 4 are greatly adsorbed to roots but are not substantially translocated to the shoot (8). Remediation technologies that either harvest roots (23) (a difficult task for large surface areas with a high worker-exposure potential) or rely on degradative capacities at root surfaces are being explored in connection with these compounds (5). Ionizable organic pollutants fall under slightly different mobility guidelines (8). Certain volatile compounds can travel in the soil void space to be absorbed by roots. The type of soil, including clays and organic matter, as well as the plant-rooting structure relative to the compounds location can greatly influence plant uptake of any compound. Certain generalizations to cover these cases can be made (20) but much work in the area is needed.

In addition to information on the flux of organics in soil and plant systems, the pesticide industry has given us an appreciation for plant metabolism of organics. Differences between the metabolic capacities of crop and weed plants are the backbone of the multibillion dollar selective-herbicide market (17). Most modern selective herbicides work on the principle that desirable plants rapidly metabolize the herbicide into a nontoxic compound, whereas undesirable weeds do not, and are consequently killed. These metabolic capacities of plants are both constitutive and inducible (18) and potentially exploitable in the remediation of contaminated soils. Under this remediation strategy, plants would absorb pollutants from the soil and metabolize them into nontoxic materials or incorporate them into stable cell constituents (e.g., lignin). This capacity of plants to detoxify xenobiotics is widely recognized, and plants have been referred to as "green livers" for this ability (26). In addition, in an effort to broaden the uses of currently registered herbicides, many researchers are further extending this degradative capacity by incorporating microbial or mammalian genes into the plant genome. Again, parallels to phytoremediation efforts are not difficult to draw and patent applications for this specific use have been filed (13).

Despite the capacity of some plants to metabolize certain xenobiotics, plants generally are "constructive" and not "destructive" organisms. Microbial metabolic processes act on a wider range of substrates, do more difficult degradative steps, and generally take the xenobiotic to a molecularly simpler end point. Combining the engineering infrastructure and autotrophic nature of the plant with the degradative capacity of the microbe is receiving much attention at the moment. Researchers are exploring the rhizosphere as a biological active zone where degradation might occur. Research in this field is just beginning on the appropriate choice of plant host (1), necessary rooting patterns, and associated microflora. Research avenues include questions' over the composition of plantproduced root exudate, exudation of specific compounds to induce microbial metabolic pathways, inoculation of the rhizosphere with microbes capable of efficient xenobiotic degraders (21), and alternative ways of sequestering the metabolically more active microbes into the plant tissue and structure. The rhizosphere is a metabolically active zone where pollutants that are normally poor microbial substrates can be microbial degraded by cometabolism. (Cometabolism is the process by which a compound that cannot support the growth of microorganisms can be modified or degraded when another growth-supporting substrate is present.) The root zone, where up to 25% of all the crop biomass can be sloughed off seems to be the ideally suited environment for this to occur (9). The mechanism of degradation behind many of the contaminants listed in the paragraph on contaminant reduction above is believed to be rhizosphere degradation. An overall scheme for possible phytoremediation of organic- contaminated soils is shown in Fig. 4.

WHAT TISSUE CULTURE AND CELLULAR BIOLOGISTS MIGHT ADD

Research and development work is needed across the entire spectrum of basic to applied science. Additional understanding of

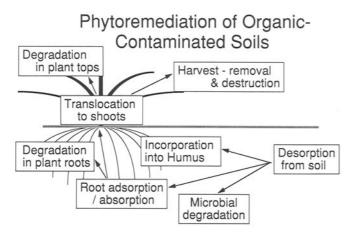


FIG. 4. Scheme for the application of phytoremediation technology to organic-contaminated soils.

the physiology, molecular biology, and chemistry of the heavy metal absorption, translocation, and tolerance by plants is needed. Our knowledge of the uptake and detoxification of organic pollutants is also rudimentary. This knowledge base is necessary as it provides the foundation on which both molecular and remediation engineers can build.

Overall, we would like to see the degradative capacity of plants increased. Possibilities in exploiting molecular genetics, plant pathology, and plant selection protocols abound. Degradative genes are regularly cloned from microbial and animal systems and have now begun to be introduced into plant systems. Many researchers involved in cell culture routinely select transformed tissues based on the introduction of new metabolic capacities into the transformed cell. Extending this concept to industrial pollutants is well within the grasp of hundreds of laboratories.

Plant microbiologists and pathologists, working with microbes both internal and external to the plant, may successfully exploit the plant as a self-sustaining bioreactor for pollutant degradation. The plant provides water, pollutant, and photosynthate flux. The microbe, mycoplasms, virus, or other provides the degradative capacity. The promise of manipulating plant-microbe systems to accomplish this is well supported by phenomenologic data from the field.

Plant structures are not optimized for remediation. Such concerns as rooting depth, structure, and density have not been altered for maximum efficiency. Molecular biology, physiology, and irrigation sciences can all directly add to the alteration of rooting structure and absorptive capacity with fairly established techniques that have not yet been brought to bear in phytoremediation.

Exploiting and expressing these capacities in the appropriate tissue at an appropriate time is a challenging goal. Setting a research strategy, much like setting a clean-up strategy, is a complex task perhaps best done by a multidisciplinary team familiar with the regulatory, physical, chemical, and biological constraints of contaminated soils. However, much remains to be done.

Last, cell and tissue culture may also provide a product in and of itself (see Paul Jackson's paper in this issue) or produce a tissue or product that can be used as an analytical tool to develop more appropriate risk-based environmental clean-up standards and tests. The risk-based prioritization of remediation projects is an expanding field which requires additional measurement tools and techniques to fully appreciate the relative risk posed by the many and varied contaminated soils that exist today.

CONCLUSION

Phytoremediation is an emerging technology built on a sound technical basis. We believe it has the potential to develop into a viable remediation option in cases where pollutants: a) are near the surface, b) are relatively non-leachable, and c) pose little imminent risk to health or the environment. Research in this area is expected to grow over the next decade as many of the current engineering technologies for cleaning surface soil of metals and non-volatile organics are clumsy, costly, and physically disruptive. The technology, when fully developed, could result in significant cost savings and result in the restoration of numerous sites by a relatively noninvasive method which in some forms can be potentially aesthetically pleasing. The technical, economic, and regulatory climate for continued research in this area is excellent.

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