11 Rhizospheric Treatment of Hydrocarbons Containing Wastewater

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Abstract

Hydrocarbons have a global attention as some of harmful contaminants due to their potential in causing fatal disease to mankind. In India, their usage is being continuously increasing to meet the needs of growing population from last few decades. Hydrocarbon discharge from various anthropogenic activities (viz., petrochemical industries, gasification, incineration) are primarily causing the detrimental effect onto the soil health and groundwater. Therefore, several methodologies and hybrid technologies are being developed for the remediation of these hydrocarbons, including physical, chemical, and biological processes. But, the remediation processes employing microorganisms and plants have been considered as environmental friendly as well as cost-effective techniques. Moreover, several efforts have been made in improving the effectiveness of these technologies. This chapter provides an understanding of remediation techniques

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by highlighting the multidisciplinary aspects. These approaches can be effectively deployed for the soil and groundwater remediation. Further, an approach of exploring the experimental outcomes in combination with the numerical modeling has been discussed which is a beneficial tool for making the technology transfer feasible from laboratory to field scale applications effectively as well as in cost-effective manner.

Keywords
Hydrocarbon pollutants · Plant-assisted bioremediation · Wastewater · Concurrent treatment

11.1 Introduction

With holding the second largest populated country in the world, still India is among the fast-growing economics of the globe. In India, agriculture is the primary supportive sector followed by the industry as secondary supportive sector for economic growth. With over a half of the country's population is living underneath the poverty level and lacking access to the basic facilities (Gupta and Sharma 2018), the policy makers and stakeholders are simultaneously adopting various schemes and policies to affirm the basic needs such as food, health, education, and livelihood to uphold the rising demand. Therefore, governments continuously encourage foreign direct investment (FDI) to boost their economy so that the provision of basic facilities could be ensured (Kuntluru et al. 2012). This scenario attracts the global manufacturer to relocate their industries in the land of the country. Currently, India imports 84% of the petroleum products, and in accordance with the Directorate General of Commercial Intelligence and Statistics (2015), during the financial year April 2018 to March 2019, the country imported around 46.6 million tons of crude oil. Several chemical industries have been established efficaciously all over the country that may release of numerous chemicals in the environment via transportation, processing, and storage. These contaminants generated via the leakage of petrochemicals further lead to the groundwater and soil pollution (Goswami et al. 2019a, b; Kushwaha et al. 2017; Seeger et al. 2011; Gupta 2020).

11.2 Hydrocarbon in Environment

Hydrocarbon contamination of soil and water is a ubiquitous problem all over the world, and remediation of these polluted resources is needed to eliminate risk to human and to the environment. Common anthropogenic sources of hydrocarbon contamination include the transportation and mishandling of petrochemical products and disposal and land application of petrochemicals from different sources and industrial sites (Goswami et al. 2017a, b; Sathe et al. 2020). According to Directorate General of Commercial Intelligence and Statistics (DGCIS), Government of India, there is increasing growth of the petrochemical industries and the exports and imports of the petrochemical products. Many researchers and agencies categorized hydrocarbon as a toxic and hazardous chemical for the ecosystem and human (Goswami et al. 2019c). United States Environmental Protection Agency (USEPA) refers hydrocarbons as toxic chemicals and are recommended for the bioremediation of all polluted sites (USEPA 1995). Similarly, USGS studies a long-term and interdisciplinary projects for the hydrocarbon-polluted sites. For examples, USGS sponsored a research project for crude oil-contaminated soil-water site near Bemidji, Minnesota (Delin et al. 1998).

When released to land, these contaminants can migrate downward through unsaturated zone, and consequently, light phase aqueous hydrocarbons float and move on top of the water table, while dense phase move downward through the water table and penetrate into the saturated zone (Dobson et al. 2007). The variable environment conditions like temperatures, soil moisture, nutrient supply, and water table fluctuation pose distribution pattern of the hydrocarbon plumes in the soil-water system. Therefore, the hydrocarbon spills present a significant threat to environment as they can result in extensive pollution from small spillages.

11.3 Decontamination Techniques

Over the past few decades, the hydrocarbon pollution is among the major problem, globally (Nedwell 1999; Goswami et al. 2018a). The current practice for remediating hydrocarbon-polluted sites relies heavily on encapsulation or isolation (capping, barriers); neither of which addresses the issue of decontamination. Cleaning these sites via immobilization or extraction by physiochemical techniques can be prohibitively expensive and is often appropriate only for small sites where rapid and complete removal is required (Kumar et al. 2019; Kushwaha et al. 2019; Bind et al. 2018; Goswami et al. 2017c). Costly methods, such as ex situ treatment and soil washing, have an adverse effect on the biological diversity (Gupta and Joshi 2017; Gupta and Yadav 2017c; Gupta et al. 2018d), soil structure, and fertility (Yadav and Hassanizadeh 2011).

For the safe drinking water production and the equilibrium of the natural resources with better ecosystem services, many technological approaches are applied in the last few decades (Gupt et al. 2018; Kumar et al. 2016; Kushwaha et al. 2015). The research studies are applied for the remediation of hydrocarbon-contaminated site by different process or integration of process such as pump-and-treat, in situ biodegradation, phytoremediation, soil washing, surfactant and co-solvent flushing, air stripping, and thermal entrapments (Bento et al. 2005). The physicochemical and other relative techniques are very economic and not feasible to its cleanup. Hence, amidst all the remediation techniques, bioremediation is the cost-efficient and sustainable technique for the eradicating the hydrocarbon contamination (Goswami et al. 2020). However, the devoid of operational facilities and interdisciplinary knowledge gaps on the research topics, literature seriously lacks the information of contamination sites particularly in India (Yadav et al. 2019; Gupta et al. 2020).

The other promising treatment options are through biological processes like bioremediation (Gupta et al. 2017), phytoremediation (Kushwaha et al. 2018; Susarla et al. 2002), and wetlands (Farhadian et al. 2008). Bioremediation is a developing cost-effective technique and causes no harm to the contaminated ecosystem as compared to the above-mentioned traditional chemical and physical methods since the biodegradation of hydrocarbons depends on the indigenous microorganisms stimulated by the pollutant (Borah et al. 2019; Goswami et al. 2018b). Various bioremediation techniques are developed to clean up residual BTEX from polluted soils, marine shorelines, and surface and groundwater systems under a broad range of environmental conditions (Gupta et al. 2018b). These techniques are readily utilized as a complementary polishing method after deploying the established techniques for the substantial removal of pure phase contamination. BTEX compounds get biodegraded in their aqueous phase by naturally occurring microorganisms in the subsurface environment, but the process is quite slow (Gupta et al. 2019). Therefore, engineered/enhanced bioremediation is practiced using additives to the natural environmental media. This involves the addition of seeded cultures, bioaugmentation or addition of nutrients, and biostimulation. The key role in the success of bioremediation in contaminated soil-water systems is played by microorganisms and various site-specific environmental parameters (Abhishek et al. 2018a, b). Use of plants may provide a multi-synchronous environment favorable for metabolism of microorganisms by increasing O_2 diffusion and root exudates, subsequently enhancing the rate of biodegradation in contaminated root zone (Gupta and Yadav 2017a, b; Gupta et al. 2018a, b, c; Gupta et al. 2019). Therefore, many researchers strongly recommended the urgent needs for knowledge development on the advance and interdisciplinary approaches of the remediation technology specially rhizoremediation/concurrent treatments (Goswami et al. 2018b; Ouyang 2002). To clean up by remediation using biological agents, three main strategies have been used: (a) stimulation of microorganism by providing the addition of substrate, (b) incubation of active organisms, and (c) integration with plant species. Rhizoremediation of petroleum contaminants is a phytoremediation process that depends on interactions among plants, microbes, and soils (Basu et al. 2015). During the rhizoremediation/plant-assisted biostimulation, some processes promote the remediation of a wide range of chemical at toxic site. Such processes are (1) modification of the physical and chemical properties of sites, (2) uses of nutrient organic carbon by releasing root exudates, (3) the aeration by transferring the oxygen to root zones, (4) retardation of the movements of chemicals by PRBs, (5) enhancement of the plant enzymatic transformation (Susarla et al. 2002).

The zone or electron acceptor-based application of the microbe diversity can result to more effective and efficiency rhizodegradation. The sub-surface zones represent the unique ecological niches due to their specific environment condition like separation of nutrient contents in different zones. Generally, the microbes are associated with thermodynamically favorable electron acceptors. Therefore, each zone process of electron acceptors inhibits the specific adaptive microorganisms. Various molecular surveys of microbial communities in various anoxic environmental, characteristic degrader ecotypes become evident for the particular environment and electron acceptor process (Kleinsteuber et al. 2008). The rhizosphere soil has 10–100 times more microbes than unvegetated (Imfeld et al. 2009). Plants influence xenobiotic biodegradation by increasing in microbial cell numbers and microbial activation that occurs in rhizospheres as a result of growth on carbon substrate provided by rhizodegradation. Plant microbes' interaction increased the mineralization process and immobilization process resulting in enzymatic enhancements. The deep fibrous root systems of plants may improve the aeration in soil by removing water through transports and by alternation of soil structure through agglomeration. The decay of dead root hair and fine root serves as an important source of the carbon for growth of rhizospheric microorganisms (Susarla et al. 2002). Plants also secrete surfactants which reduce the surface tension and solubilize contaminants in soil water. Therefore, bioavailability in different zones is increasing due to reduction in the toxicity. The constructed wetlands are the examples to stimulate the combined effects of the biostimulation, bioaugmentation, and phytoremediation. Some specialized plant species play a very important role in phytostimulation under wetland condition, which removes almost 100% of pollutants from soil-water system. Microbial growth kinetics meets to mass transfer kinetics and enzymatic kinetics which results as the ultimate biodegradation of substrate. Many researchers investigate batch experiment and column experiment using different plant species and reported different kinetics models. These are zero order kinetics models, first-order kinetics models, Monod's kinetics, etc. Mathematical modeling of plant-assisted bioremediation is helpful for the bioremediation technology, proposed schemes, policy, and managements of contaminant site (Narayanan et al. 1998a, b). Therefore, a better understanding is needed for plant-assisted bioremediation of hydrocarbon-contaminated soil-water system that is presented here with special emphasis on the rhizoremediation strategies and their kinetics and mathematical approaches for two-dimensional and three-dimensional modeling. That will help in the policy framework and recommendations for the cost-effective remediation technologies. It is also powerful for the common platform to address and respond to dialogue, priority setting, and policy formulation for the better managements of cleanup technology. Models in the rhizoremediation during fate and transports of hydrocarbons are summarized as follows with governing equation and mechanisms in Fig. 11.1.

11.4 Concurrent Treatment Facilities

This low-tech in situ approach of concurrent treatments is more attractive for bioremediation of hydrocarbon-polluted soils as it offers site restoration, partial decontamination, and maintenance of the biological activity, which is visually unobtrusive, and there is the possibility of clean water production (Van Gestel et al. 2003). Due to the enormous potential for its cost and environmental savings (Gupta et al. 2017), there is a significant interest in this technology that is in its early stage of development, and very little information is available related to site cleanup from start to finish. Effective implementation of concurrent treatments requires a thorough understanding of the soil-plant-atmospheric continuum processes which is currently

1. Moisture based geo-chemical mechanisms of the hydrocarbons transports

$$\frac{\partial \theta}{\partial t} - \nabla . D(\theta)\nabla\theta - \frac{\partial K}{\partial z} = 0$$

4. Plants Uptakesmechanisms

$$\frac{\partial C}{\partial t} = -q\, T_{scf} C_s$$

5. Biodegradation Kinetics mechanisms

$$\frac{\partial C}{\partial t} = \mu_{max} C \frac{(C_o + X_o - C)}{(K_s + C)}$$

2. Multi-dimensional advective-dispersive mechanisms

$$D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} - V_x \frac{\partial C}{\partial x} - \frac{r}{n} = \frac{\partial C}{\partial t}$$

3. Sorptionmechanisms

$$\frac{\partial \phi C^i}{\partial t} = -{r_{ads}}^i + {r_{des}}^i$$

6. Contamination Evaporation (Volatilization) mechanisms

$$C_g^i = H^i C_{lig}^i$$

Fig. 11.1 Describes the summery of governing equation and the mechanisms of the rhizospheric treatments of the hydrocarbons polluted soil-water

poorly understood and makes this technology expensive and inefficient despite the tremendous potential mentioned above. Most of the current research deals with the effect of microorganisms on soil and water, wherein the hydrocarbon-contaminated effluent or sewage sludge is applied to the cropped soil and the plants grown on such sites are analyzed experimentally to determine their capacity to remove hydrocarbons from the root zone. These studies simply correlate the hydrocarbon contaminant concentration in growing media with its presence in the plant biomass for a particular soil-water-plant system without hypothesizing how the fate and transports actually take place. This lack of understanding hinders the efforts of researchers in their quest to develop concurrent treatments from contaminated soil-water system. Techniques are used separately for treating wastewater and contaminated water. Therefore, innovative concurrent method for treating both the resources in symbiosis way is urgently needed. Moreover, isolated experimental and modeling works are mostly performed for both experimental and numerical methods, which are needed to be studied together. Similarly, there is limited scientific information on the impacts of the rhizospheric treatment during the fate and transports of the hydrocarbon in the soil-water system. Other than this, one remaining hurdle for commercial implementation of such treatment has been the disposal of the produced contaminated biomass, which is addressed rarely by the researchers so far. Therefore, the focus of this chapter is to generate the interdisciplinary and multidisciplinary aspects of effectiveness and the mechanisms of the highly complex soil-water-plant-atmospheric continuum processes during the concurrent treatments of wastewater and hydrocarbon-polluted soil-water resources from start (laboratory investigation) to finish (modeling approaches and field application). The specific research topics include interdisciplinary aspects as listed in Box 11.1.

Box 11.1 Interdisciplinary and multidisciplinary aspects of treatment facilities

A. Contaminant hydrology and biochemical engineering.
- A1—the mechanisms governing the concurrent treatment process.
- A2—plants and chelating agent's for hydrocarbon removal from polluted sites.

B. Mass transfer (analytical tool) and mass balance (modeling tool).
- B1—gas chromatography-mass spectrometry (GC-MS) for quantitative measurement of organic matter and metal dynamics in plant biomass and root zone.
- B2—magnetic resonance imaging (MRI) for in situ measurement of plant growth and dynamic root density distribution.
- B3—metal distribution in the soil solution and plant biomass using flame atomic absorption spectrophotometer.

C. Use of mathematical modeling in vadose zone processes.
- C1—simulating water and contaminant dynamics in vadose zone and their uptake by plant.
- Biomass using a realistic approach.
- C2—validating of the developed model using experimental data obtained in B1, B2, and B3.

D. Technology comparison in cleanup processing.
- D1—valorization of plant-enhanced decontamination during rhizospheric treatment for different techniques.
- D2—economic evaluation of rhizospheric treatment against traditional techniques.

11.5 Numerical Modeling

The modeling involves the simultaneous movement of soil water and hydrocarbons through soil-water-plant system by coupling of the moisture flow equation with the contaminant transport equation in the presence of sink terms as mentioned in Eqs. (1) and (2). Similarly, the degradation pattern of the hydrocarbons can be used to numerically simulate the movement of water and hydrocarbon transport through the heterogeneous variably saturated zone (Gupta and Yadav 2019). The modeling involves the simultaneous movement of soil water and pollutant movement through soil-water-plant system by coupling of the moisture flow equation with the contaminant transport equation in the presence of sink terms. The transient moisture dynamics in variably saturated porous media is expressed by a parabolic partial differential equation popularly known as Richards' equation which is derived by integrating the Darcy's law with the equation of continuity. This equation is in its three-dimensional mixed form which is coupled by non-uniform sink function for water by plants:

$$\frac{\partial \theta}{\partial h}\left(\frac{\partial h}{\partial t}\right) = \frac{\partial}{\partial x} K_x(h)\frac{\partial h}{\partial x} + \frac{\partial}{\partial y} K_y(h)\frac{\partial h}{\partial y} + \frac{\partial}{\partial z} K_z(h)\frac{\partial h}{\partial z} + K(h) - S(t,h) \qquad (1)$$

where θ is the volumetric water content defined in the volume of water per unit volume of soil and h is the pressure head. $S(t, h)$ is a sink function that represents the water extraction by surface vegetation, z is the depth of root zone measured positive upwards, K is the hydraulic conductivity of the soil, and t is the time. This equation is highly nonlinear for unsaturated flow, since hydraulic conductivity K and the volumetric water content are nonlinear functions of the dependent variable h, the soil moisture pressure head. To solve this equation, explicit expressions for the soil constructive relationship between the dependent variable h and the nonlinear terms K and θ are required.

The classical convection dispersion equation is used for contaminant transport in multidimensions taking the contaminant extraction term (Kumari et al. 2019). The Fick's law coupled with the mass balance equation yields a modified form of advective-dispersive equation.

$$\frac{\partial(\rho_s S_D)}{\partial t}+\frac{\partial(\theta C)}{\partial t}=\frac{\partial}{\partial x}\left[D_{xx}\theta\frac{\partial C}{\partial x}+D_{xy}\theta\frac{\partial C}{\partial y}+D_{xz}\theta\frac{\partial C}{\partial z}\right]+ \frac{\partial}{\partial y}\left[D_{yx}\theta\frac{\partial C}{\partial x}+D_{yy}\theta\frac{\partial C}{\partial y}+D_{yz}\theta\frac{\partial C}{\partial z}\right]+\frac{\partial}{\partial z}\left[D_{zx}\theta\frac{\partial C}{\partial x}+D_{zy}\theta\frac{\partial C}{\partial y}+D_{zz}\theta\frac{\partial C}{\partial z}\right] -q_xC-q_yC-q_zC+S(t,C) \tag{2}$$

Yadav and Hassanizadeh (2011) described the general expression for the solute biodegradation in soil-water system, in which only microbial densities and the contaminant concentration determine the degradation kinetics.

$$\frac{\partial C}{\partial t}=\mu_{\max}C\frac{(C_0+X_0-C)}{(K_s+C)} \tag{3}$$

where $\mu_{\max}$ is the maximum growth rate, C is the contamination concentration at time t, C_0 is the initial contamination concentration, X_0 is the contamination required to produce initial microbial density, and K_s is the half saturation constant.

The equilibrium adsorption isotherms founded in the case of hydrocarbons in soil-water system is mostly Langmuir equilibrium adsorption isotherms. The Langmuir equation is

$$S_{eq}=\frac{S_{\max}K_L C_{eq}}{1+K_L C_{eq}} \tag{4}$$

where S_{eq} is the concentration of adsorbed viruses and C_{eq} is the concentration of free viruses after apparent equilibrium has been reached. $S_{\max}$ is the maximum adsorbed concentration when all active surface sites are occupied; K_L is a constant related to the bonding energy. The movement of water and hydrocarbons in soils is generally better described with multidimensional non-equilibrium models than with more commonly used one-dimensional and/or equilibrium models. Furthermore, such equations are solved for the validation of the different field data set. Therefore, it plays very important role for the contamination fate and transport modeling including hydrocarbon contamination in the vadose zone and/or also saturated zone.

11.6 Rhizospheric Treatment Facilities: Solar/Wind-Based Design

Rhizosphere deliberates as the "ecological remediation unit" for treating contaminated soils, possessing huge amount of microbes particularly bacteria, fungi, and rhizobacteria (symbiont with the plant roots). This approach gives how and in what extent the extreme environmental variations of soil moisture content, temperature, and water table dynamics could affect the biodegradation of hydrocarbon contaminants in variably saturated soils. Direct practical importance for remediating hydrocarbon-polluted natural resources is very high for Indian climatic conditions. A successful transformation of this cost-effective technology of bioremediation from laboratory to the field would have a significant impact on science and industrial application, not only in India but also for countries having the similar environmental conditions. An improved understanding of bioremediation processes that control biodegradation of organic contaminants is required to effectively implement this environmental-friendly technology for decontaminating the polluted sites (Mustapha et al. 2018). Such remediation technologies are convenient for the polluted site where the handling of petrochemical substances is established such as oil refineries and port and costal area. The produced database and knowledge gained in this chapter can be used to encourage petrochemical and hydrocarbon production industries and other environmental agencies for remediating hydrocarbon-contaminated soil-water systems. If the application of bioremediation from the lab to the field proves to be efficient, this would have a positive impact on sustainability and the marketing of petrochemical of the country. At the same time, soil-water systems are referring under vulnerability due to such activities. Therefore, the technological supports to the commercial or industrial activities become the millstones

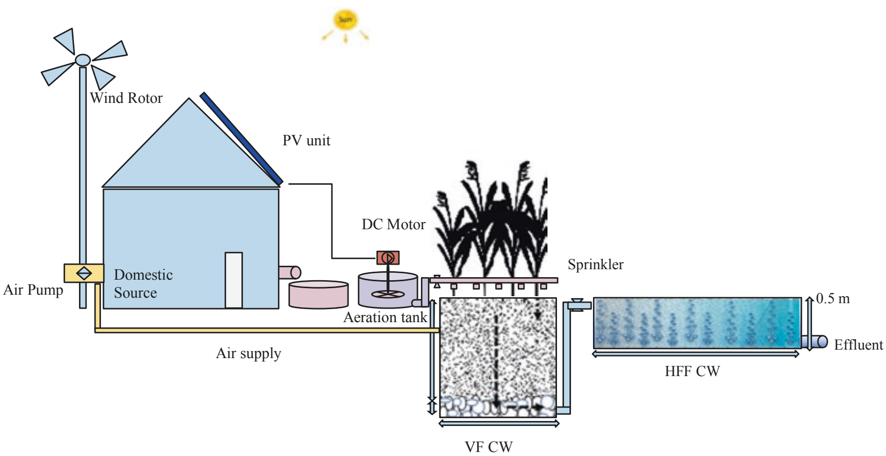

Fig. 11.2 Rhizospheric treatment: solar/wind-driven aeration of hybrid CW (VF + HFF)

for the sustainable developments. In this regard, a new treatment facility (Fig. 11.2) has been designed for treatment of wastewater containing hydrocarbon pollutants. The treatment system consisted of influent tank having a capacity of 200 L made of high-density polyethylene plastic containers located between source and aeration tank. The aeration tank having the capacity of 400 L is located next to influent tank and peristaltic pumps used to pump the influent into the hybrid CWs. The hybrid CW was composed of two parts: (1) a vertical flow (VF) CW planted with *Canna generalis* and (2) a horizontal flow filter (HFF) CW. Aeration of root zone enhances the biodegradation of hydrocarbon in VF chamber where motor of aeration can be driven by solar/wind system.

11.7 Summary and Recommendations

In India, the increase in the demand of hydrocarbons and its utilization cause the devastating effects to the ecosystems due to occurrence of mishandling episodes and lack of infrastructure, which in turn requires the development of engineered technologies in their remediation. Rhizospheric treatment alone with the conventional methods seems to be effective and reliable in this respect. However, appropriate innovations are needed to upgrade the literature for direct practical implication of the technique according to Indian climatic conditions. Some recommendations are as follows:

1. Use of nano-biomaterials and biochar to enhance the rhizospheric degradation of hydrocarbon is a new direction of research and application (Ranjan et al. 2018).
2. Solar/wind-driven aeration may accelerate the aerobic biodegradation of petrochemical in root zone; however to maintain optimal aeration, it is important to investigate the other operational parameters.
3. In situ aerobic heating, i.e., providing optimal heated water using PV system, can be an effective approach.
4. Modeling of root zone mechanisms is needed to understand the accurate root uptake and pollutant distribution in subsurface.

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