

Review of Creosote Pollution Toxicity and Possibilities of Bioremediation

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Abstract. Creosote oil is a complex mixture of hydrocarbon compounds obtained from high temperature distillation of coal tar. It is used for over 100 years as a fungicide, insecticide, miticide, and sporicide to protect wood and is applied by pressure methods to wood products, primary utility poles and railroad ties. This treated wood is intended for exterior or outdoor uses only. Its commercial uses include railroad ties 70 %, utility poles 15-20 %, and other miscellaneous commercial uses 10-15 %. Composition of the creosote depends on the source and it has typically 85 % polycyclic aromatic hydrocarbons (PAHs), 10 % phenolic compounds, and 5 % heterocyclic. Between 20 and 40 % of the total weight of typical creosote can be attributed to the 16 PAHs defined as priority pollutants by the United States Environmental Protection Agency (EPA). The production of creosote in the European Union (EU) has been estimated to be approximately 60.000-100.000 t per year. The presence of the toxic PAHs and phenolic compounds make creosote treated wood harmful for the environment at the end of its service life and direct or indirect human exposure to creosote treated wood may cause carcinogenic affect to kidney, liver, bladder, eyes and skin. In this presentation we review creosote environmental pollution toxicity and possibilities of remediation

Keywords – creosote, PAH, pollution, remediation, toxicity.

I INTRODUCTION

Wood-preserving industry uses more pesticides than any other industry worldwide. The one of major chemical employed for this purpose is creosote used in the wood treating industry for over 100 years. Wood treated with creosote is widely used, e.g., as poles for electric power or telephone lines and as railroad sleepers [1]. The production of creosote in the EU has been estimated to be approximately 60,000-100,000 t per year, preservation of railway uses 54 % of creosote. [2]. When properly used and disposed of, creosote does not appear to significantly threaten human health. However, misuse, accidental spillage, and improper disposal of creosote results in contaminated environments with serious potential health risk. Creosote contamination is generally associated with surface soils, waters in treatment lagoons or evaporation areas, and groundwater contaminated with leachate from the above sources [3]. Creosote oil is a complex mixture of hydrocarbon compounds obtained from high temperature distillation of coal tar. Coal tar creosote is composed of approximately 85 % polycyclic aromatic hydrocarbons (approx. 50 % two-ring, 39 % three-ring, 9 % four-ring and 2 % five-ring); 10 % phenolic compounds; and 5 % N-, S-, and O- heterocyclic [3-10]. Between 20 and 40 % of the total weight of typical creosote can be attributed to the 16 PAHs defined as priority pollutants by EPA and EU (Fig.1), many of the

PAHs are recalcitrant compounds and have toxic properties.

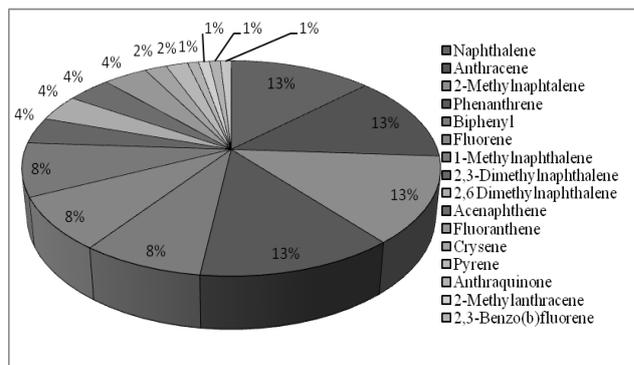


Fig.1. Predominant PAHs constituent in Creosote [3],[8],[11]

II CREOSOTE TOXICITY

Creosote's qualities as a wood preservative are excellent; however accidental spillage, improper disposal results in contaminated environments with serious potential health risks. Creosote compounds are commonly occurring industrial pollutants and are often found as co-contaminants in the environment. Most of creosote consisting compounds are known as carcinogenic, teratogenic, genotoxic and immunotoxic [8, 12-16]. The effects on human health will depend mainly on the length

and route of exposure, the amount or concentration of creosote compounds. Human exposure to creosote compounds occurs through 3 routes: respiratory tract, gastrointestinal tract and skin contact coming into direct contact with creosote and breathing in airborne creosote particles increase the chances for creosote exposure inside the body. Through the lungs and skin creosote will slowly pass into the bloodstream. Common external effects include rashes, either severe or mild, chemical burns around the eye area, sensitivity to light, eye damage and blistering or peeling skin. Internal damage can also occur from exposure to creosote from eating, drinking or breathing in the creosote chemicals [17]. Internal toxic damages can include unconsciousness, cancer, convulsions, mental impairment, kidney and liver problems and several other internal issues [18]. However, the industrialization of creosote has increased the amount of the chemical present in groundwater and soil. The creosote forms a tar-like substance in the water, where it takes many years to completely break down. One or two per cent of creosote used to treat wood ends up in the air [19]. Any plant or animal that is near the creosote in the water, air or soil absorbs the chemicals. Plants absorb less of the creosote than animals. The main danger of creosote exposure to the environment is then that the chemicals absorbed by the animals or plants move on to humans (Fig.2.).

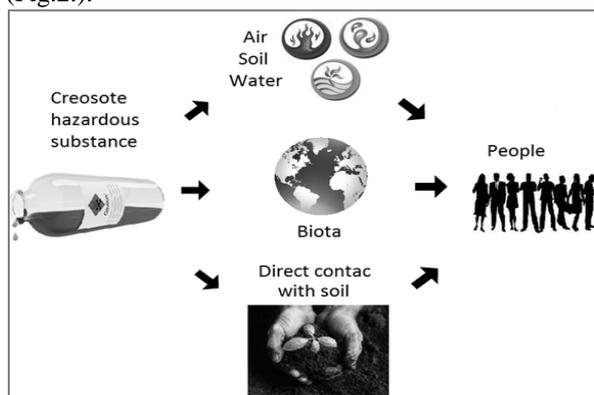


Fig. 2. Exposure pathways of creosote pollution.

III BIOREMEDIATION OF CREOSOTE

Biological degradation is the primary dissipation mechanism for the most of organic pollutants in the environment, however the activity of degrading microorganisms depends upon many factors, including contaminant uptake and bioavailability, concentration, toxicity, mobility access to other nutrients and activated enzymes [11]. The rate and extent of biodegradation of creosote in soils and sediments is affected by multiple factors, such as physico-chemical (physical/chemical properties of PAHs, phenols, heterocycles, organic content of soil, structure/particle size of soil, presence of contaminants), biological factors (characteristics of the microbial population: diversity, genetic/catabolic potential) and environmental factors (temperature, moisture, pH, sorption, degree of contamination).

Two-ringed PAH as naphthalene is broken down easily. The three-ring PAHs, such as acenaphthalene,

fluorene, phenanthrene, anthracene and the four-ring PAHs such as fluoranthrene, pyrene, benzo(a)anthracene and crysene have been shown to be metabolized by microorganisms and degraded in creosote bioremediation although at slower rates as the molecular weight increases. However, the high-molecular weight five, six, seven ring PAHs, such as benzo(b)fluoranthene, indeno(1,2,3-cd)pyrene and coronene, have not been shown to significantly biodegrade even over long periods of time [7].

Because of its toxicity, creosote-treated wood does not degrade easily in the environment and requires special disposal methods. The detoxification of creosote treated wood has become more popular since this material has been classified as dangerous waste on the basis of the existing law dispositions (EU Directive 2001/90/CE).

It is generally accepted that creosote is a major wood preservative. Creosote-treated wood has commonly been used for railroad crossties and landscape facilities, as a result, creosote contamination of surface soils, segments, and groundwater is commonly observed in areas in which creosote-treated wood was used. The combustion of creosote treated wood is the main industrial treatment actually in use, however up to nowadays the great majority of bioremediation experiments concerned with creosote regards polluted soil, where different microorganisms are involved in the oxidation and subsequent mineralization of creosote components: soil bacteria or fungi, are described (Fig.3.).

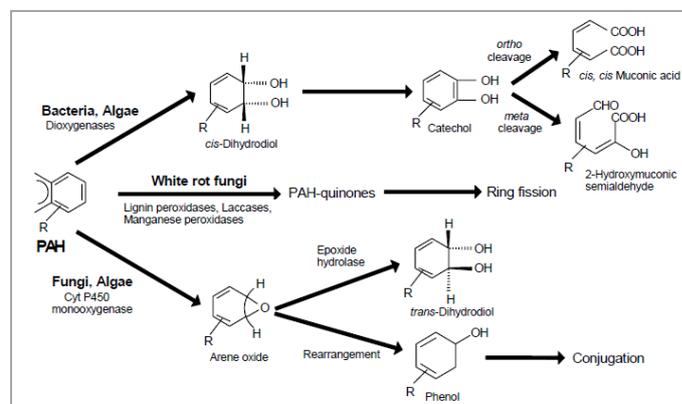


Fig. 3. Major pathways involved in the metabolism of polycyclic aromatic hydrocarbons by bacteria, fungi and algae [20].

There are a lot of articles related with bioremediation experiments concerned with creosote regards polluted soil, however, information related to wood impregnated with creosote bioremediation is not much compared to bioremediation of soil.

Creosote degradation by bacteria

Creosote is not easily degraded by microorganisms. Bacteria involved in degradation of creosote components (some PAHs, HACs, phenolic compounds) have been isolated and identified as belonging mostly to the genera *Pseudomonas* [21-24], *Spingomonas* [25], *Bacillus* [26] and *Mycobacterium* [27],[28]. Creosote biodegradation by bacteria strains limited the high- molecular-weight PAHs continued to be recalcitrant in treated soil [29].

Despite the removal of a majority of creosote contaminants from groundwater through biotreatment, only a slight decrease in toxicity and teratogenicity of biotreated groundwater was observed. Recent studies demonstrated that several treatments have been successful in reducing, for example, the total PAH concentration in contaminated soils, however increased the microbial mutagenicity of these soils [30]. Creosote compounds biodegradation by bacteria are used more frequently than biodegradation by fungi or removal with phytoremediation.

Degradation of creosote by fungi

The pollutants contained in the creosote-treated wood waste are also barriers to its use as landfill cover due to potential migration of contaminants into ground water. The life expectancies of creosote-treated railway ties

depend on rail traffic, tie placement and environmental hazards. The service life of creosote-treated railway ties is from 30 to 50 years. Therefore, a proper disposal method should include an effective degradation of the pollutants in creosote treated wood waste. Positive results were obtained with fungi belonging to different genus are presented in Table 1 [31],[32]. High degradation was obtained with white-rot fungi *Phanaerochaete cryosporium* [33], *Pleurotus ostreatus* [34-36], *Cladosporium sp.* [31], *Polyporus sp.* [37], filamentous fungus *Fusarium sp.* (strain E0330) [38], ascomycetous fungus *Thermoascus aurantiacus* [43]. White-rot fungal species could be interesting for the treatment of wood waste because of their ability to degrade phenols and PAH and to grow on ligninocellulosic materials [39-41].

TABLE 1.

STUDIES ON CREOSOTE CONTAMINATED SOIL AND CREOSOTE TREATED WOOD BIOREMEDIATION BY FUNGI

Fungi	Research findings	References
<i>Creosote-contaminated soil</i>		
<i>Pleurotus ostreatus</i> , <i>Irpex lacteus</i>	Fungus degraded PAH with 4–6 aromatic rings more efficiently than the microbial community present initially in the soil. PAH removal was higher in <i>P. ostreatus</i> treatments than in <i>I. lacteus</i> . Indicates a potential for synergistic interaction between the fungus and actinobacteria in bioremediation.	[35]
<i>Cladosporium</i> , <i>Fusarium</i> , <i>Penicillium</i> , <i>Aspergillus</i> , <i>Pleurotus</i>	White-rot fungus belonging to the genus <i>Pleurotus</i> was better able to degrade creosote than other organisms studied, and that mixed fungal cultures more effectively degraded creosote than did pure cultures.	[31]
<i>Phanerochaete sordida</i>	Demonstrate the potential of lignin-degrading fungi in the solid-phase bioremediation of creosote-contaminated soils, but, the persistence of highly toxic PAHs containing > 5 rings represents a significant challenge which must be overcome before the technology can be considered a viable treatment option for creosote-contaminated sites.	[42]
<i>Pleurotus ostreatus</i>	<i>P. ostreatus</i> has an overall positive effect on the degradation of aged creosote in soil and that degradation is enhanced by increased temperature and pre-treatment.	[34]
<i>Creosote-treated wood</i>		
The fungal inoculums comprise at least one creosote-tolerant fungus (e.g. <i>Antrodia radiculosa</i> , <i>Neolentinus lepideus</i> , etc.)	Waste wood containing creosote is remediated or degraded by inoculating it with the fungal inoculums. The inoculums and waste wood are combined until an end product is achieved that is at least partially remediated or of a reduced volume.	[32]
<i>Pleurotus ostreatus</i>	Phenols are completely degraded. Some more dangerous compounds, such as methyl-PAH and 5–6 rings PAHs are generally well degraded. Almost complete degradation of creosote oil components, also confirmed by a significant reduction of ecotoxicity.	[41]
A total of 179 fungal isolates was recovered from 240 sampling points	There was a large difference in the fungal diversity between the surface and the inner area of crosstie wastes. <i>Heterobasidion annosum</i> and <i>Schizophyllum commune</i> showed very high resistance to PAHs, which indicates that these species may play a role in the degradation of PAHs.	[9]
<i>Agrocybe</i> , <i>Armillaria</i> , <i>Auricularia</i> , <i>Daedalea</i> , <i>Pleurotus</i> , <i>Trametes</i>	The most tolerant strain, <i>Pleurotus ostreatus</i> SMR 684, extracellular laccase and peroxidase specific activities were monitored during growth in the presence of creosote-treated wood.	[40]
<i>Thermoascus aurantiacus</i>	The inoculation of cellulolytic and thermophilic fungus <i>Thermoascus aurantiacus</i> did not accelerate the bioremediation process in degrading phenolic compounds and may have inhibited the metabolic activity of composting organisms.	[43]

Since the low bioavailability of PAHs by bacteria, the increased bioavailability of PAHs metabolites, produced by white rot fungi, can increase their mineralization by bacteria [44]. In contrast to bacteria, fungi generally do not utilize PAHs as sole carbon and energy source, but transform them comethabolically to detoxified metabolites. The degradation process in soil proceeds as a cooperation between white rot fungi (catalyse PAHs

oxidation) and bacteria (mineralise PAHs with higher water solubility and oxidise metabolites of recalcitrant). In soil treated with *Pleurotus ostreatus*, where the soil bacteria were almost eliminated by the fungus, mineralization of PAHs was incomplete due to accumulation of intermediary metabolites that could not be oxidised by the fungus [45], but can be mineralised by bacteria.

Phytoremediation

Phytoremediation describes the treatment of environmental problems through the use of plants that mitigate the environmental problem without the need to excavate the contaminant material and dispose of it elsewhere. This cleaning method consists of mitigating pollutant concentrations in contaminated environment with plants able to accumulate, degrade, or eliminate various contaminants from media that contain them. There are several basic phytoremediation techniques: rizofiltration, phytoextraction, phytotransformation, phytostimulation, phytostabilization. The main advantages phytoremediation compared with other approaches are the preserve the natural structure and texture of the cleaned soil; it is low in cost and has potential to be rapid. One serious limitation is that many plant species are sensitive to contaminants [46].

The role of plants in remediation creosote and PAHs contaminations (which are the main part in creosote contaminants) presented in Table 2. PAHs can be degraded in the rhizosphere however may also interact with vegetation by accumulation in plant tissues or adsorption on root surface. Contaminant bioavailability and toxicity are critical factors in phytoremediation and bioavailability depends on lipophilicity and age, soil

characteristics, such as pH, clay and organic matter content. The PAHs are widely distributed hydrophobic organic contaminants in soil usually exhibit low bioavailability to both microorganisms and plants due to their strong affinity to the soil matrix, especially to soil organic matter which limits the application of phytoremediation [11],[45],[47].

According the literature data there are selected number of plant species which have been found to be promising candidates for phytoremediation of such persistent pollutants as PAHs and could be used as an alternative technique to reduce creosote (PAHs) levels in soils [46-48] (Table 2). Plants provide root exudates with carbon, energy, nutrients, enzymes and sometimes oxygen to microbial populations in the rhizosphere. Root exudates of sugars, alcohols, and acids are annually from 10 to 20 % of plant photosynthesis [49] and provide sufficient amount of carbon and energy to support the large numbers of microbes. Root exudates are link between plants and microbes that causes the rhizosphere effect. This environmental cleaning method is effective, however evidence exists for the enhanced dissipation of PAHs in the rhizosphere when compared to nonrhizosphere soil [50-52].

TABLE 2.

STUDIES ON PHYTO-REMEDICATION WITH CREOSOTE (PAHS, PHENOLS) CONTAMINANTS IN SOIL

Common name of plant (Scientific name of plant)	Research findings	References
Alfalfa (<i>Medicago sativa</i> L.).	The residual concentrations of pyrene in the rhizosphere soil were lower than those in the non-rhizosphere soil.	[51]
	All vegetated treatments resulted in higher phytoremediation efficiency.	[53]
	Arbuscular mycorrhizal inoculation and addition of the surfactant consistently promoted phenanthrene dissipation in the soil	[54]
Alfalfa (<i>Medicago sativa</i> L.), tall fescue (<i>Festuca arundinacea</i> Schreb.).	Cropping promoted the dissipation of soil PAHs. Tall fescue gave greater removal of soil PAHs than alfalfa, intercropping was more effective than monoculture. Stimulates PAH degrading bacterial population.	[44]
Alfalfa (<i>Medicago sativa</i>), ryegrass (<i>Lolium perenne</i>).	Are potential plants for phytoremediation. The interactions among the consortia of plant root exudates, microorganisms, and amended compost in rhizosphere soils could facilitate bioavailability of pyrene and subsequently enhance its dissipation	[52]
Ryegrass (<i>Lolium perenne</i> L.).	Enhances dissipation or biotransformation of a large range of PAHs including 5 and 6-ring PAHs. The major part of PAHs dissipation in rhizosphere soil was due to biodegradation or biotransformation.	[50],[55]
Willows (<i>Salix viminalis</i>).	Enhanced the degradation of all PAHs, most likely by the pronounced rhizosphere effect on the activity of active microorganisms, including PAH degraders.	[56]
Tall fescue (<i>Festuca arundinacea</i>), kentucky bluegrass (<i>Poa pratensis</i>), wild rye (<i>Elymus canadensis</i>).	Tall fescue is more suitable for use in phytoremediation. Plant growth promoting rhizobacteria were evaluated for plant growth promotion and protection of plants from contaminant toxicity.	[46]
Tall fescue (<i>Festuca arundinacea</i>), red fescue (<i>Festuca rubra</i>), ryegrass (<i>Lolium perenne</i>), red clover (<i>Trifolium pratense</i>), white clover (<i>Trifolium repens</i>).	In soils spiked with pure PAHs or coal tar the rate of loss of PAHs is much greater than in historically contaminated soils. There were clear differences in the losses of the PAHs depending on ring size amongst the 16 PAHs tested.	[57]
Bean (<i>Vicia faba</i>), corn (<i>Zea mays</i>), wheat (<i>Triticum aestivum</i>).	All vegetated treatments resulted in higher phytoremediation efficiency.	[58]
Vetch (<i>Vicia sativa</i> L.).	Vetch could tolerate and remove high phenol concentrations, avoiding serious phytotoxic effects. Thus, vetch could be considered an interesting tool in the field of phytoremediation.	[59]

Addition of surfactants has been explored in the cleanup of contaminated soils [60]; there are few reports of their application in phytoremediation, they found that the presence of the surfactant enhanced PAH degradation by a rhizosphere effect on the microbial activity and by increased bioavailability [54],[56].

IV CONCLUSION

Creosote, which is a common preservative used in wood treating processes, consists of different polycyclic aromatic, phenolic hydrocarbons and heterocyclic compounds. Because of their toxicity and their mutagenic and carcinogenic properties polycyclic aromatic hydrocarbons are classified as ubiquitous contaminants of highest concern in the environment. Toxic, environmental-persistent chemicals are resistant to degradation in the natural environment. Creosote contaminants do not degrade easily in the environment and require special disposal methods as there are not so many creosote-tolerant microorganisms, fungi and plant species.

Although many studies have been conducted to develop useful methods of bioremediation using microbes, fungi or plants, there is a limited number of species that can be used. Therefore, predictions of creosote dissipation from laboratory amended soil or treated creosote wood do not reflect the true situation in the field conditions. The further studies must be conducted to identify various species and fungal communities, microbial and plant species capable of bioremediation not only in lab conditions, but must show good results in field testing. Forthcoming studies should be the basis for the development of efficient technology, suitable for both in-situ and ex-situ processing of creosote-contaminated soil or large amounts of creosote-treated wood.

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