Science of the Total Environment 478 (2014) 201–225

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Review

Removal of polycyclic aromatic hydrocarbons in aqueous environment by chemical treatments: A review

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HIGHLIGHTS

- PAHs are present in aquatic ecosystems and affect human health and living beings.
- Conventional water treatments can be unsuccessful in the PAH degradation.
- AOPs and their simultaneous combination generally result in high PAH conversions.
- AOPs coupled with biotreatments could be a cost-effective solution for PAH removal.

ARTICLE INFO

Article history:
Received 9 October 2013
Received in revised form 30 December 2013
Accepted 30 December 2013
Available online 16 February 2014

Keywords:
Polycyclic aromatic hydrocarbon
Wastewater treatment
Ozonation
Photolysis
Advanced oxidation process

ABSTRACT

Due to their carcinogenic, mutagenic and teratogenic potential, the removal of polycyclic aromatic hydrocarbons (PAHs) from aqueous environment using physical, biological and chemical processes has been studied by several researchers. This paper reviews the current state of knowledge concerning PAHs including their physico-chemical properties, input sources, occurrence, adverse effects and conventional and alternative chemical processes applied for their removal from water. The mechanisms and reactions involved in each treatment method are reported, and the effects of various variables on the PAH degradation rate as well as the extent of degradation are also discussed. Extensive literature analysis has shown that an effective way to perform the conversion and mineralization of this type of substances is the application of advanced oxidation processes (AOPs). Furthermore, combined processes, particularly AOPs coupled with biological treatments, seem to be one of the best solutions for the treatment of effluents containing PAHs.

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http://dx.doi.org/10.1016/j.scitotenv.2013.12.126
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1. Introduction

PAHs are a type of organic compounds that consist of two or more condensed benzene rings and/or pentacyclic molecules that are arranged in various chemical configurations. These compounds are formed primarily from the incomplete combustion or pyrolysis of organic material such as oil, petroleum, gas, coal, and wood (González et al., 2012; Manariotis et al., 2011). PAHs have recently attracted a lot of attention in studies on water, soil and air pollution because some of them are highly carcinogenic, mutagenic and teratogenic substances (Busetti et al., 2006; Manoli and Samara, 2008; Menzie et al., 1992; Reynaud and Deschaux, 2006). Additionally, PAHs are persistent organic pollutants due to their chemical stability and biodegradation resistance. For these reasons, they are strictly regulated by law in most industrialized countries. However, many of them are not regulated in developing countries. Several PAHs have been identified by the US-EPA and the European Union (WFD, 2000/60/EC) as priority pollutants in order to reduce the release of these compounds into the environment (Busetti et al., 2006; Manoli and Samara, 2008). In fact, a maximum admissible concentration for the most dangerous PAHs in the environment has been set (WFD, 2008/105/EC).

The removal of such substances from the environment can be performed through physical, biological and chemical processes (Fatone et al., 2011; Manoli and Samara, 2008; Tian et al., 2012; Veignie et al., 2009; Vela et al., 2012; Zeng et al., 2000a). In the case of removing PAHs from aqueous systems, physical processes, such as volatilization and adsorption, have an important role, greatly reducing the amount of PAHs in the water. Nevertheless, physical processes do not solve the problem of PAH pollution because of their inability to degrade these contaminants. For this reason, biological and chemical degradation processes are generally preferred (Veignie et al., 2009; Vela et al., 2012; Zeng et al., 2000a,b). However, due to the biorecalcitrant, toxic character and low aqueous solubility of PAHs, conventional biological systems exhibit limited contribution to PAH removal from water (Manoli and Samara, 2008; Mueller et al., 1989). Therefore, chemical processes may be the most efficient for the conversion of these substances. Among the chemical techniques, direct photolysis is one of the major transformation processes affecting the fate of PAHs in the aquatic environment (Vela et al., 2012). Furthermore, PAHs may undergo oxidation through ozonation (Beltrán et al., 1999; Bernal-Martinez et al., 2007, 2009; Ledakowicz et al., 2001; Zeng et al., 2000b) and chlorination (Manoli and Samara, 2008), among other processes. Advanced oxidation processes (AOPs) have also been broadly investigated. AOPs are techniques that use the highly oxidant and non-selective hydroxyl radical (°OH), which is able to react with almost all types of organic compounds, to lead to their total mineralization or the formation of more biodegradable intermediates (Glaze et al., 1987).

In recent years, the research interest in the removal of PAHs from the environment has been increased. However, to the authors' knowledge there is no a review article concerning chemical treatment systems for PAH degradation in aqueous environment. Therefore, the aim of this work is to review, evaluate, discuss and compare different conventional and alternative chemical processes, and their combination with biological ones for the degradation of PAHs and their removal from water.

2. Background

2.1. PAHs: definition, classification and physico-chemical properties

PAHs, also known as polynuclear aromatic hydrocarbons, or more simply as polyaromatics, are a group of over one hundred organic compounds known for their toxicity and environmental persistence. They are mainly made up of carbon and hydrogen assembled in two or more stable benzene rings. Their physical and chemical properties are determined by their conjugated π-electron systems, which are dependent on the number of aromatic rings and their molecular weight. They are classified as having a low molecular weight if they have two or three condensed aromatic rings, or a high molecular weight if they have four or more condensed benzene rings. PAHs can be also classified as alternant or non-alternant if their structure is composed entirely of benzene rings or if four, five, and six-member non-aromatic rings are included, respectively (Wick et al., 2011).

The solubility of PAHs in water is dependent upon temperature, pH, ionic strength, and water matrix components (i.e. dissolved organic carbon) (Dabestani and Ivanov, 1999; Vela et al., 2012). However, in general PAHs have a relatively low solubility in water (ranging from 31 g m^{-3} for NA to 0.26 mg m^{-3} for BghiPY) (Dabestani and Ivanov, 1999), but dissolve easily in fats and oils, thus, they have a tendency to accumulate in the fatty tissue of living organisms. PAHs have variable vapor pressures (ranging from 10.4 Pa for NA to 0.37 nPa for DahA) (Dabestani and Ivanov, 1999). Those PAHs with lower vapor pressures are associated with particles, whereas those with higher vapor pressures are found as vapor at ambient temperature in air. Generally, the solubility of PAHs decreases and hydrophobicity increases with an increase in the number of condensed benzene rings. In addition, volatility decreases as the number of condensed aromatic rings increases.

2.2. Emission sources and occurrence in the environment

PAHs are widely spread throughout the natural environment and are found in soil, sediments, water, air, plants and animals, as a result of both natural and anthropogenic processes (Chen et al., 2004; Guo et al., 2007). In nature they are generated by natural forest fires, reactions in living beings, volcanic eruptions and natural oil seeps. However, PAHs are more commonly generated by anthropogenic activities, mainly as a result of combustion processes, especially the incomplete burning of organic materials during industrial and other human activities (e.g. industrial discharge, transportation, cooking, biomass burning, tobacco smoking, coal, petrol, gas and wood combustion, and waste incineration) (Ravindra et al., 2008).

PAHs are released into the atmosphere mainly via gaseous emissions, but they can also be discharged from soil and water compartments through evaporation or resuspension of particles. Once in the atmosphere, PAHs are subject to short and long-range transport, and they are removed by wet and dry deposition onto water, soil and vegetation. On the other hand, polyaromatics can reach aquatic systems through natural oil seeps, by atmospheric deposition and/or through accidental or intentional discharges mainly from oil extraction, transportation and refining. PAHs in water can bind to suspended particles or sediments, or bioaccumulate in aquatic organisms through the

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