



# Fate and behaviour of nitrogen-containing polycyclic aromatic hydrocarbons in soil



Ihuoma N. Anyanwu, Kirk T. Semple\*

Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, United Kingdom

## HIGHLIGHTS

- N-PAHs are present in hydrocarbon polluted soils, often in the presence of PAHs.
- There is a lack of information on the presence and behaviour of N-PAHs in soil.
- N-PAHs are typically more toxic and more mobile than homocyclic PAHs.
- N-PAHs are typically less biodegradable and more persistent than homocyclic PAHs.

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## ABSTRACT

Nitrogen-containing polycyclic aromatic hydrocarbons (N-PAHs) are analogues of the homologous polycyclic aromatic hydrocarbons (PAHs) in which one or more carbon atom(s) is replaced with nitrogen atom(s). They do not only exist in conjunction with the homocyclic PAHs in polluted systems, but unlike PAHs, they are intentionally produced. It has also been reported that heterocyclic PAHs contribute to the toxic effects identified in many environmental samples, containing PAH/N-PAH mixtures. N-PAHs are toxic, often displaying greater biological effects than their homocyclic analogues. As these chemicals are suspected of being carcinogens in humans and other biota, there is a need to know their fate, behaviour, bioavailability, ecotoxicity, and biodegradation for risk assessments of contaminated soils. The aim of this review is to consider the presence and behaviour of N-PAHs, focusing on their bioavailability, ecotoxicity and biodegradation in the soil environment.

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## 1. Introduction

It is now accepted that polycyclic aromatic hydrocarbons (PAHs) are a key group of contaminants present at many contaminated sites, particularly at refinery, timber treatment, manufactured gas plant and coke work sites. What is less well described is that

\* Corresponding author.

E-mail address: [k.semple@lancaster.ac.uk](mailto:k.semple@lancaster.ac.uk) (K.T. Semple).

nitrogen-containing polycyclic aromatic hydrocarbons (N-PAHs) can also be present, often at high concentrations (Brumley et al., 1991; Webber, 1994; Švábenský et al., 2009). Many of these chemicals are carcinogenic, mutagenic, genotoxic and reproductive toxins and are of concern to both biotic and human exposure (vo-Dinh et al., 1998; National Toxicology Program, 2001; US Environmental Protection Agency, 2001; Bleeker et al., 2002; Sutherland et al., 2005; Hazardous Substance Data Bank, 2010; Environment Canada, 2011; Brar et al., 2010; IARC, 2012; IPCS Environmental Health Criteria, 2003). The fate, behaviour and impacts of PAHs in soil have been extensively studied, and reviewed because of their environmental persistence and implications for their putative toxic and carcinogenic properties (Agency of Toxic Substances and Disease Registry, 1995; U S Environment Protection Agency, 1997; Santodonato, 1997; Wilcke, 2000; US Department of Health and Human Services, 1995; Villeneuve et al., 2002; Semple et al., 2003; IPCS Environmental Health Criteria, 2003). The impact of oxygenated PAHs in soil has also been reviewed (Lundstedt et al., 2007). An overview of the sources, toxicity profile and some effluent treatment methods of N-PAHs which focused mainly on the monocyclic compounds, such as pyridine and its derivatives have been reported (Padoley et al., 2008). However, investigations into the behaviour of N-PAHs in soil and their impacts on the environment have not been reported as extensively.

N-PAH contamination, resulting from petrogenic, pyrogenic and biogenic activities, represents major pollution and toxicological problems within the environment (vo-Dinh et al., 1998; Environment Canada, 2011; IARC, 2012). Despite the putative toxicological properties of N-PAHs, there is a paucity of information on the fate and behaviour of N-PAHs in the soil environment. Due to their presence in the environment coupled with their physico-chemical properties, mobility and toxicity, it is imperative that the behaviour of N-PAHs is investigated more fully, particularly with regard to residence-time, bioavailability, toxicity and biodegradation in soils, and the relevance of these factors to the risk assessment and remediation of contaminated sites.

Therefore, the aims of this review are to provide a critical review of the fate and behaviour of N-PAHs in soil considering bioavailability, ecotoxicity and biodegradation in soil. Following on from this to consider these factors in relation to risk assessment and remediation of contaminated systems.

## 2. The properties of N-PAHs

N-PAHs are analogues of PAHs in which one or more carbon atoms are replaced with the corresponding number of nitrogen atoms (Table 1). These compounds occur in nature and are also produced through anthropogenic activities (Research Institute for Fragrance Materials, 2003; Hazardous Substance Data Bank, 2010; Environment Canada, 2011). Compared to homocyclic PAHs, N-PAHs possess lower  $K_{ow}$  values, ranging from 2.03 to 6.05 (Table 1), and are more water soluble and therefore more mobile in the environment. Examples include pyridines, indoles, quinolines, methyl-quinolines, acridines, phenanthridines, benzo-quinolines, carbazoles, dibenz-acridines and pyrazines. Typically, N-PAHs tend to be present together with homologous PAHs in the environment, but often the former have greater acute toxicity (Pereira et al., 1988; Anyanwu et al., 2013); for example, the N-PAHs have been shown to be responsible for toxic and carcinogenic effects in many environmental samples (Wang and Meresz, 1982). From both toxicological and epidemiological studies, many heterocyclic aromatics are highly toxic and suspected to be carcinogenic agents in humans and other biota (van-Herwijnen et al., 2004; Hazardous Substance Data Bank, 2010; Environment Canada, 2011; IARC, 2012). Studies have shown that N-PAHs have greater toxicity when they are bio-transformed to dihydrodiols, hydroxyls and epoxides (Kummar et al., 1989; Warshawsky et al., 1992; Bleeker et al., 2002; Sutherland et al., 2005). Because of their high mobility, they can leach into water and contaminate both groundwater and drinking water supplies. Table 1 shows some PAH and N-PAH structures, and the key differences between their physico-chemical properties.

### 2.1. Sources and uses of N-PAHs

N-PAHs originate from coal, creosote, crude oil spills, vehicle exhaust, tobacco smoke, shale oil, fossils, pesticide, industrial effluents, incomplete combustion of fuels and organic materials (Kaiser et al., 1996; Fetzner, 1998). Besides PAHs and phenolic compounds, 3%–5% of heterocyclic compounds are present in coal tar creosote (Johansen et al., 1997). Approximately  $15 \times 10^6$  tonnes/year of coal tars are produced worldwide mainly from coking plants. Furthermore, several thousand tonnes of pure condensed heterocyclic compounds, such as carbazoles, quinolines, pyridines, indoles and phenols, are produced each year. Further,  $2 \times 10^6$  tonnes of aromatic oils containing N-PAHs used as wood preservatives, solvents and gas-washing oils, are produced annually > (Collin and Hoke, 1995). Unlike PAHs, N-PAHs are found in pharmaceutical and industrial wastes because of their use as solvent, chemical intermediate, corrosion inhibitor, fragrance mixtures and in the manufacture of pharmaceuticals (Research Institute for Fragrance Materials, 2003; Hazardous Substance Data Bank, 2010; Environment Canada, 2011).

Most of N-PAHs are used in the synthesis of dyes, antiseptics, drugs, flavouring agent, paints, pesticides, detergents, and as raw materials in industries. They are an intermediate in the manufacture of some pharmaceuticals (sulfadiazole, pyridoxine, vitamin B6, antihistamines, steroids), textile water-repellent agents (zelan), rubber chemicals (piperidine), bactericides (cetylpyridinium bromide), and herbicides (diquat and paraquat); as a denaturant for alcohol, as well as anti-foaming agents in petrochemical manufacturing (Padoley et al., 2008; Research Institute for Fragrance Materials, 2003; Environment Canada, 2011). In contrast, it is known that some N-PAHs are used as antioxidants; it has been reported that, as strong antioxidants, some synthesized N-PAHs may protect DNA from harmful chemical reactions (Naik et al., 2009).

## 3. Behaviour of N-PAHs in the environment

The fate and behaviour of N-PAHs in the environment has generated interest because of their putative impacts on function and toxicity on soil biota, such as microbes, earthworms and plants (Pereira et al., 1988; Willumsen et al., 2001; van-Herwijnen et al., 2004; Sverdrup et al., 2002; Pasková et al., 2006; Kobetičová et al., 2008, 2011; Anyanwu et al., 2013).

Soil represents a significant sink for anthropogenically-produced chemicals (Kwon and Choi, 2014) (Table 2). In soil, N-PAHs may be subjected to a variety of losses through either physical, chemical or biological processes, including volatilization, leaching, photo-oxidation or biodegradation. They may also be retained within the soil through chemical binding or physical sequestration processes, such as sorption

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