



Review

Polycyclic aromatic hydrocarbons in deep sea sediments: Microbe–pollutant interactions in a remote environment

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HIGHLIGHTS

- PAH tend to accumulate in DSS
- High hydrostatic pressure and low temperature adversely affect PAH biodegradation in DSS
- DSS bacterial communities house distinct among strains, catabolic genes, enzymes and pathways
- We review the impact of DSS bacteria on PAH degradation under current and future climate scenarios.
- Future climate scenarios may indirectly affect PAH biodegradation in DSS.

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ABSTRACT

Recalcitrant polycyclic aromatic hydrocarbons (PAHs) released into seawater end up in the deep sea sediments (DSSs). However, their fate here is often oversimplified by theoretical models. Biodegradation of PAHs in DSSs, is assumed to be similar to biodegradation in surface habitats, despite high hydrostatic pressures and low temperatures that should significantly limit PAH biodegradation. Bacteria residing in the DSSs (related mainly to α - and γ -Proteobacteria) have been shown to or predicted to possess distinct genes, enzymes and metabolic pathways, indicating an adaptation of these bacterial communities to the psychro-peizophilic conditions of the DSSs. This work summarizes some of the most recent research on DSS hydrocarbonoclastic populations and mechanisms of PAH degradation and discusses the challenges posed by future high CO₂ and UV climate scenarios on biodegradation of PAHs in DSSs.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are hydrophobic molecules consisting of two or more fused aromatic rings arranged in a variety of structural configurations. PAHs bioaccumulate through trophic webs (Clements et al., 1994; Kanaly and Harayama, 2000) and exert acute, toxic, mutagenic, teratogenic and carcinogenic effects on living organisms (Menzie et al., 1992; Kanaly and Harayama, 2000; Neff, 2002). PAHs with two and three aromatic rings are considered low molecular weight (LMW) PAHs, while compounds with four or more aromatic rings are known as high molecular weight (HMW) PAHs. In Table 1, we listed the 16 priority PAHs as classified by the US Environmental Protection Agency (EPA) along with measured and estimated physico-chemical data. Overall, organic carbon-water partition coefficient (K_{oc}), octanol-water partition coefficient (K_{ow}) and half-life values increased with increasing numbers of rings, whereas vapor pressure decreased. Thus, an increasing number of rings reduces volatility, solubility and biodegradation rates, and increases hydrophobicity, adsorption to particulate matter, toxicity and recalcitrance (Seo et al., 2009).

Bioavailability refers to the forms and quantity of chemicals that biota can take up during their lives and thus determines how and to what extent these chemicals enter the food chain. In the biodegradation of hydrophobic pollutants, bioavailability is important and is expressed as the relative fraction of a specific pollutant that is available to microbes. Solubility and sorption are important parameters when considering bioavailability. The first is directly correlated with bioavailability, while the second is affected by various external factors. All PAHs are hydrophobic and lipophilic, and will adhere to available soil particles in a seawater environment. The 'strength' of this association varies and is among other things related to soil and PAHs physico-chemical properties (Semple et al., 2003). K_{oc} (Table 1) directly reflects the sorption capacity of PAHs and it increases with increasing number of rings. Sorption also increases with increasing soil organic carbon content (Wang et al., 2001) and decreasing pore size (Semple et al., 2003). In soot, a carbon-rich matrix, pyrolytic PAHs are highly sorbed and occluded, which reduces their degradation in comparison to other surfaces (Kim et al., 2009). At DSS organic carbon content is lower than at coastal marine sediments (Nagata et al., 2010) thus PAHs are more bioavailable. However, the rate of desorption of organic pollutants from the matrix decreases exponentially over time (aging). Aging functionally resumes to the shift from weaker hydrogen and van der Waals to covalent bonds. By aging, PAH-soil interaction becomes more stable and irreversible (Semple et al., 2003).

PAHs are introduced into the marine environments sporadically through marine oil spills and continuously through urban run-off, industrial and domestic wastewater/sludge discharge, atmospheric deposition, ship ballast cleaning, offshore oil exploration and natural seepage (Notar et al., 2001; Guitart et al., 2007, 2010). PAH inputs into marine environments can be classified into two groups: petrogenic and pyrolytic (Ke et al., 2002a). Petrogenic sources derive directly from petroleum and derivatives, while pyrolytic sources derive from the combustion of fossil fuel. Higher PAH concentrations in seawater, are normally associated with anthropogenic coastal activities, particularly shipping harbors (Cincinelli et al., 2001; Wurl and Obbard, 2004; Fang et al., 2008; Coelho et al., 2010) and major rivers (Lipiatou et al., 1997b). The highly urbanized coastal waters are reported to be 1.5 and 4.5 times more enriched in dissolved and particulate PAHs respectively than pristine sites (Guitart et al., 2007).

Increasing energy demand will lead to an increase in the transportation, storage and use of fossil fuels thereby increasing the risk of oil spill. Remediation of oil pollution response relies on physical, biological and chemical processes. Biological methods mainly focus on enhancing natural biodegradation through the supplementation of nutrients, surfactants (biostimulation) or microorganisms (bioaugmentation). Nutrient biostimulation approaches rely on N and P supplementation

in the form of inorganic salts to contaminated areas. Autochthonous microbial biodegradation of oil pollutants is often limited by the low relative abundance of bioavailable nitrogen in comparison to carbon. Supplementation of N and P increases microbial degradation, but is only effective when these elements are limiting. This approach is more effective in soil, sand and embayments. In the open ocean, nutrient dispersion significantly reduces its effectiveness and requires continuous supplementation which raises costs (Nikolopoulou et al., 2007). Oil degradation is also limited by the reduced oil-water interface, where biodegradation occurs. Application of surfactants reduces the interfacial tension and increases oil-water emulsification, enhancing biodegradation (Hazen et al., 2010). Bioaugmentation (addition of exogenous or endogenous bacteria) is an overlooked tool in oil spill response and has been deemed ineffective and relatively expensive (Megharaj et al., 2011). Maladaptation of the inoculated microorganisms to the new environment constitutes a major barrier to successful bioaugmentation (Churchill et al., 1995). However, the use of autochthonous strains could help to overcome this limitation and improve bioremediation efficiency (Hosokawa et al., 2009) and bioaugmentation with PAH-degrading bacteria, immediately after spill, can minimize persistent PAH afterwards (Ron and Rosenberg, 2014).

The extension of oil exploration towards deeper ocean introduces novel technological challenges and environmental concerns. Therefore, in depth scientific knowledge and new technological need to be developed. Here we focus on the particular features of the deep sea that make it a relevant bioremediation site, the origin and fate of PAHs in deep sea sediments and the microbial players, pathways and genes involved in PAH biodegradation.

2. Bacterial life in the deep sea

In the present review, deep sea sediments refer to the sea floor below 1000 m depth. This environment is considered extreme because of the challenging conditions to microbial life in terms of nutrient supply, electron donors and acceptors, and exposure to physical factors such as temperature and pressure.

The DSS environment is characterized by high pressure (10–50 MPa), low temperatures (2–3 °C; except in hydrothermal vents) and low concentration of labile organic carbon. Salinity varies between 34.3 and 35.1 g L⁻¹ and pH between 7.5 and 8.0 (Nagata et al., 2010). Collectively, the physico-chemical conditions of the deep sea environment are more stable than at the ocean surface (Nagata et al., 2010). Although the abyssal plains form the major part of the deep sea, other habitats are present and include hydrothermal vents, cold seeps, mud volcanoes, Fe(III)–Mn(IV) nodules, trenches, seamounts and canyons (Nagata et al., 2000).

Marine sediments, in general, are covered with biofilms of dense and diverse microbial communities (Chipman et al., 2010). They are efficient players in the biogeochemical cycles of carbon, nitrogen and phosphorus (Silva et al., 2003; Shao et al., 2010; Kostka et al., 2011). Life in the deep sea mainly depends on the input of organic carbon from the ocean above. Dissolved organic carbon in the superficial deep sea sediments varies from 509 to 1038 $\mu\text{mol L}^{-1}$ in the Atlantic Ocean and 672 to 1529 $\mu\text{mol L}^{-1}$ in the more eutrophic Arabian Sea (Lahajnar et al., 2005). Exogenous organic carbon inputs into the deep sea derive from lateral advection from slopes and shelves, diffusion and sinking. Resuspension from bottom sediments by turbulence mixing, chemosynthetic activity and hydrocarbon seeps also contributes to the microbial food web.

Organic matter is continuously degraded by microbes while sinking, leaving behind a refractory remnant (Nagata et al., 2010). Bacteria in the deep sea are adapted to degrade these recalcitrant compounds. Specific adaptations to oligotrophic environments, such as the production of bioflocculants, have been reported in deep sea bacteria and assist in nutrient and carbon sequestration (Wu et al., 2013). Deep sea microbial communities have been reported to decompose biodegradable

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