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A review of non-exhaustive chemical and bioavailability methods for the assessment of polycyclic aromatic hydrocarbons in soil

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HIGHLIGHTS

- Small range of *in vivo* animal models for quantifying PAH bioavailability in soil.
- Absence of guidance documents for PAH *in vivo* models.
- *In vivo* to *in vitro* relationship studies on PAHs is limited.
- There is no certified reference material for assessment of bioavailability or bioaccessibility.
- Absence of PAH and metal or metalloid mixture effect on bioavailability or bioaccessibility.

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ABSTRACT

Current environmental regulations relating to risk assessment for contaminated sites are often set assuming that the contaminant is 100% bioavailable. It is therefore not surprising that remediation of contaminated sites is very expensive and maybe it is unrealistic to achieve a "remediated state" under current regulatory guidelines using existing technologies. In fact the reality is that only a portion of the contaminant on site becomes available for absorption and goes into the systemic circulation. This article gives an overview of existing *in vivo* animal models that quantify oral bioavailability of polycyclic aromatic hydrocarbons (PAHs) in soil. It also provides a summary of *in vitro* gastrointestinal extraction methodologies and some of the key factors influencing absorption of PAHs in soil. For innovation, it highlights that bioaccessibility values derived from *in vitro* studies still require validation from *in vivo* animal models to gain regulatory acceptance. Additionally, this review highlights the use of non-exhaustive chemical methodologies as a valuable tool to understanding the bioavailability process and behaviour of PAHs in soil.

Contents

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1. Introduction

Soil is an important and limited resource and it is timely that the United Nations has declared 2015 as "The International Year of Soil" (United Nations General Assembly, 2014). The development of soil policy at the European Union level such as the introduction of thematic strategy for the protection of soils (EC, 2006a) and the proposed soil framework directive (EC, 2006b) is envisioned to reduce ecosystem and human health risk from possible exposure to contaminated soil. It is noteworthy that contamination of soil creates a significant risk to human health. Soil contamination is mainly associated to industrial and commercial land-use, waste land-fills, military camps and nuclear power plants (Panagos et al., 2013). In a recent paper by Naidu et al. (2015a) it is estimated that on a global scale, there are over 3 million potentially contaminated sites that may pose a threat to human health and the environment.

In the developed world the cost of remediating such contaminated sites is estimated to be in billions of dollars. In Australia remediation of contaminated sites is estimated to cost around 5–8 billion dollars (Australia, 1997), in the USA about 1 trillion dollars (Rao et al., 1996) and about 0.1%–1.5% of GDP per annum in Europe (Lanno et al., 2004). The high cost for remediating contaminated sites can be attributed to current legislations which are mainly based on the total contaminant concentrations in soil, sediment and groundwater and that the contaminant is 100% bioavailable (Ng et al., 2013). In reality, only a fraction of the contaminant may be bioavailable and as such the assumption that all of the ingested contaminant is solubilised in the gastrointestinal tract (i.e. 100% bioavailable) and is absorbed into systemic circulation may grossly overestimate the daily chemical intake thereby influencing risk assessment (Ng et al., 2010). Bioavailability is defined as the amount of contaminant that is absorbed into the body following skin contact, ingestion or inhalation (Ng et al., 2010). On the other hand, bioaccessibility is often specifically used when *in vitro* assessment models are used and is defined as the fraction of a contaminant that is soluble in the gastrointestinal tract and is therefore available for absorption (Ng et al., 2013; National Environment Protection Council, 2013). A critical review of the "bioavailability process" including the concept of a decision tree for the determination of contaminant bioavailability and bioaccessibility for health risk assessment is provided elsewhere (Ng et al., 2013; Oomen et al., 2006).

In the 1980s and 1990s most of the industrialised (G5) countries had environmental protection guidelines enacted with a focus on total chemical concentrations in soils (Naidu et al., 2015b). Notably, current environmental regulatory guidelines for site contamination assessment in Australia, National Environment Protection Measures (NEPM) and overseas are often set assuming the contaminant is 100% bioavailable (Ng et al., 2010; National Environment Protection Council, 2013). Such a conservative approach that in the absence of site-specific data, the bioavailability of a contaminant is assumed to be 100% could lead to unnecessary or expensive remediation options (Ng et al., 2013). It is therefore not surprising that a number of studies highlight the possibility that clean-up at most of the contaminated sites using existing technologies may not be sufficient to achieve the standards set under current environmental regulatory guidelines (Doick et al., 2005; Ng et al., 2015; Swindell and Reid, 2006). In Australia, NEPM recommends that contaminant bioavailability and bioaccessibility measurements be adopted as part of tier-two risk assessment (i.e. quantitative) for contaminated sites. The inclusion of bioavailability values is a key parameter which can be utilised for minimising the uncertainty associated with exposure in risk assessment (Ng et al., 2013).

In the absence of human studies or the availability of suitable epidemiological data, the relative bioavailability of soil-borne contaminants may be assessed using *in vivo* methods (Ng et al., 2013). The "gold" standard for *in vivo* studies is using animal models such as immature swine because of its similarity of anatomical gastrointestinal tract to infants, the most sensitive risk receptor in human populations (Ng et al., 2013; Bordelon et al., 2000; Duan et al., 2015). Nevertheless, animal studies can be very expensive with many ethical issues relating to animal welfare in research and testing. To address such issues several *in vitro* models have been developed to determined bioaccessibility as a surrogate model for the estimation of bioavailability (Ng et al., 2013). Despite this, few *in vitro* bioaccessibility studies have been calibrated against a reliable animal model, such as immature swine (Ng et al., 2013).

Further, different contaminants have different effects on human health and the environment depending on their properties (Swindell and Reid, 2006). PAHs cover a wide range of hydrophobic organic chemicals that are structurally similar and are formed during incomplete combustion or pyrolysis of organic matter resulting from natural or anthropogenic processes. PAHs are fairly persistent in the environment and are lipophilic with capacity to bioaccumulate in living organisms. As organic pollutants with known or potential carcinogenic and mutagenic properties, PAHs are of concern to human and environmental health. The United States Environmental Protection Agency (US-EPA) has listed 16 un-substituted PAHs as priority contaminants as these are most widely found in the environment (Jahin et al., 2009) and considered markers of urban activities (Peng et al., 2013). PAHs are ubiquitously present in soil, sediments, air, water and in living organisms. Consequently, humans are exposed to PAHs on a daily basis from their surroundings including incidental soil ingestion, dietary intake and inhalation from ambient and indoor air. It is understood that PAHs only become a risk if they are or become