



Review

Advances in in vitro methods to evaluate oral bioaccessibility of PAHs and PBDEs in environmental matrices



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HIGHLIGHTS

- In vitro methods simulate human GI tract to measure POP bioaccessibility.
- They can potentially predict POP bioavailability after correlation with in vivo animal data.
- Reviewed five in vitro methods using PAHs and PBDEs as traditional and emerging POPs.
- Discussed their applications and limitations plus method improvements and challenges.
- Shed light for research to assess human exposure to POPs via oral ingestion pathway.

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ABSTRACT

Cleanup goals for sites contaminated with persistent organic pollutants (POPs) are often established based on total contaminant concentrations. However, mounting evidence suggests that understanding contaminant bioavailability in soils is necessary for accurate assessment of contaminant exposure to humans via oral ingestion pathway. Animal-based in vivo tests have been used to assess contaminant bioavailability in soils; however, due to ethical issues and cost, it is desirable to use in vitro assays as alternatives. Various in vitro methods have been developed, which simulate human gastrointestinal (GI) tract using different digestion fluids. These methods can be used to predict POP bioavailability in soils, foods, and indoor dust after showing good correlation with in vivo animal data. Here, five common in vitro methods are evaluated and compared using PAHs and PBDEs as an example of traditional and emerging POPs. Their applications and limitations are discussed while focusing on method improvements and future challenges to predict POP bioavailability in different matrices. The discussions should shed light for future research to accurately assess human exposure to POPs via oral ingestion pathway.

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1. Introduction

Persistent organic pollutants (POPs), such as most polycyclic aromatic hydrocarbons (PAHs) and polybrominated diphenyl ethers (PBDEs), are characterized by low aqueous solubility, low vapor pressure, lipophilic properties and long half lives in soils (Juhász and Naidu, 2000). Due to their adverse impacts on human health (Därnerud et al., 2001), it is important to determine their exposure to humans from different environmental matrices including soil, dust and food. Human exposure to POPs mainly occurs from three pathways including inhalation, dermal absorption and oral ingestion, with the oral ingestion pathway being the most important, which includes consumption of food and incidental ingestion of soil and dust.

Oral ingestion is a major exposure route for the most susceptible sub-group of the population, i.e., young children with high frequency of mouth-hand behavior. When quantifying exposure to ingested contaminants, 100% bioavailability is usually assumed, i.e., all ingested contaminants are soluble in gastrointestinal (GI) tract and absorbed into systemic circulation. However, there is growing evidence to indicate that total concentration may not represent the fraction of the contaminant that is absorbed into the body (i.e., the bioavailable fraction), which exerts a negative effect on human health. As a result, understanding POP bioavailability is important to accurately quantify the risk of contaminant exposure to humans (Cui et al., 2013; Rostami and Juhász, 2011). In the context of human health, contaminant bioavailability can be described in absolute or relative terms. Absolute bioavailability (ABA) can be defined as the fraction of a dose reaching the systemic circulation (Oomen et al., 2003). Relative bioavailability (RBA) is the comparative bioavailability of different forms or exposure media containing the contaminant, which is expressed as a fractional relative absorption factor (Ruby et al., 1999). For example, Smith et al. (2012) used DDT-spiked sand as the dosing vehicle to measure the RBA of DDT in contaminated soils. It was calculated by comparing the adipose tissue concentration of DDT in soil-dosed mice to that of sand-dosed mice. Related to bioavailability is the concept of bioaccessibility, referring to the fraction of contaminant dissolved in simulated GI solution, which can be potentially taken up by intestinal cells, and can be measured by physiologically based in vitro methods.

In vivo assays are preferred methods as they can directly determine the bioavailability of POPs in different matrices. However, they are expensive and impractical for large scale testing. Simple and inexpensive in vitro methods, which simulate digestion processes in human GI tract, have been developed as surrogates to predict POP bioavailability (Dean and Ma, 2007). The underlying principle of in vitro assays is that the amounts of a contaminant that are extracted by digestive fluids are correlated to the amounts that are potentially absorbed following ingestion of a contaminated matrix. Although in vitro assays offer an attractive alternative to in vivo assays, their application to refine POP exposure is still in the developmental phase.

This paper reviews the current status of in vitro methods to

predict POP bioavailability in different matrices including soil, food and indoor dust. Emphasis is placed on recent developments to overcome method limitations, including the use of a sorption sink to overcome solubility constraints associated with hydrophobic organic contaminants, and the use of epithelial Caco-2 cell lines to simulate human sorption of contaminants. While the principles described here applicable to all POPs, this review focuses primarily on two contaminant classes namely PAHs and PBDEs. These contaminants represent both traditional and emerging POPs with many compounds within each class having a range of physicochemical and toxicological properties.

2. Presence of PAHs and PBDEs in the environment

PAHs refer to hydrocarbons containing >2 fused benzene rings in different arrangements. There are several hundreds of PAHs, but only 16 PAH compounds are listed as priority contaminants by the US Environmental Protection Agency (Gan et al., 2009). PBDEs are a group of brominated compounds comprising 209 congeners, which have been used as flame retardants to reduce the flammability of furniture, textiles and electronic equipment.

Soil is the primary reservoir for PAHs compared with other matrices (Wild and Jones, 1995). Urban soil impacted by nearby industrial and human activity or soil irrigated with wastewater usually contains elevated concentrations of PAHs (Tao et al., 2002; Duan et al., 2015). The total concentrations of 16 PAHs in urban soils from three European cities are 1.49–51.8 in Glasgow, UK, 0.22–4.49 in Ljubljana, Italy, and 0.15–3.41 mg/kg in Torino, Slovenia, with the higher value for UK samples resulting from vehicle exhausts (Table 1; Morillo et al., 2007). PAH concentrations in foods varied depending on local conditions, such as wastewater irrigation or industrial plants. For example, total concentrations of 16 PAHs in edible vegetables from soil irrigated with wastewater are 158–995 µg/kg (Table 1; Wang et al., 2011a). For the aquatic biota, low molecular weight PAHs with 3- or 4-rings are generally more bioaccumulative than those PAHs with 5- or 6-rings (Bordajandi et al., 2004; Palma-Fleming et al., 2004). For example, 16 PAHs were detected at 8.22–71.4 ng/g of fresh weight in Spanish marine biota (sea bream, bivalves, and prawns) with pyrene as the dominant congener, accounting for >80% of the 16 PAHs. While 5- and 6-ringed PAHs (e.g., benzo[a]pyrene (BaP), chrysene, and dibenz[a,h]anthracene) were below the detection limit for all samples (Bordajandi et al., 2004). The bioaccumulation of PAHs mainly depends on biota feeding preference, PAH origination, and trophic level of biota. In addition, higher molecular PAHs can be metabolized to different extents by different species of aquatic organisms (Wang et al., 2010). Compared with flame retardants, lower PAHs concentrations have been detected in indoor dust (Hoh et al., 2012; Pieters et al., 2013; Qi et al., 2014). Indoor dust from houses using coal for heating usually contain elevated concentrations of PAHs. For example, PAH concentrations in indoor dust from 20 coal-burning houses in China are 8.45–121 mg/kg (Wang et al., 2013a), which are higher than those from other countries (Table 1).

PBDEs may enter the environment via production facilities, e-

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