Chemosphere 210 (2018) 941–948

Contents lists available at ScienceDirect

Chemosphere

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

Comparative evaluation of influence of aging, soil properties and structural characteristics on bioaccessibility of polychlorinated biphenyls in soil

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HIGHLIGHTS

- Bioaccessibility was susceptible to aging, soil properties and congener specificity.
- Bioaccessibility decayed in first-order kinetics with aging after stabilization.
- Larger soil particle size, soil moisture and PCBs polarity improved bioaccessibility.
- Greater dependency on aging and SOM was highlighted for bioaccessibility by PLS.
- Mechanistic links were shown with formulated relationship for PCBs bioaccessibility.

ARTICLE INFO

Article history:
Received 26 March 2018
Received in revised form
19 July 2018
Accepted 19 July 2018
Available online 20 July 2018

Handling Editor: T Cutright

Keywords:
Polychlorinated biphenyls
Bioaccessibility
Biomimetic extraction
Influential factors
PLS analysis

ABSTRACT/PURPOSE

Though bioaccessibility commonly recognized as a guideline for risk assessment is closely related with pollution occurrence and chemical species of compounds, the mechanistic links are barely evaluated particularly for widespread polychlorinated biphenyls (PCBs) in soil. With the biomimetic extraction of hydroxypropyl-β-cyclodextrin (β-HPCD), the temporal and spatial influences of soil properties, aging and structural characteristics, e.g. polarity of PCB congeners on bioaccessibility were investigated for PCBs. Sensitive variation of bioaccessibility with aging, soil organic matter (SOM), particle size and soil moisture were clearly evidenced for different PCB congeners. Due to aging, the bioaccessibility decreased in the long term after stabilization for 36 h. In concert with the first-order kinetics, the decay rates of bioaccessibility were shown with congener-specificity and were well correlated with dipoles of PCBs. The increment of SOM diminished the bioaccessibility for the strengthened adsorption while the increased particle size and soil moisture elevated it possibly due to the less adsorption on soil particles and more accommodation of PCBs in soil pore water. Except the positive correlations with particle size, soil moisture and dipole moment, the greater dependency on aging and SOM was highlighted for bioaccessibility by partial least squares (PLS) analysis. The mutual relationship with influential factors was quantitatively formulated for accelerative prediction of bioaccessibility, and the comparative evaluation and detailed insights into the mechanistic links would thus help enhance the precise determination of bioaccessibility and risk assessment of PCBs in soil.

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1. Introduction

Due to the thermal stability, non-flammability and dielectric properties, polychlorinated biphenyls (PCBs), structurally diversified with one or/to ten chlorine substitution (s) on biphenyl was purposefully produced on a large scale for varieties of industrial

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https://doi.org/10.1016/j.chemosphere.2018.07.111
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applications, e.g. as heat exchange fluids in electric transformers and capacitors or as additives in plastic processing and printing (Wang et al., 2012; Stella et al., 2017; Tombesi et al., 2017). With massive industrial use, it was roughly estimated that over 370,000 tons of PCBs have been released into environmental media (Tanabe, 1988) including air (Sohail et al., 2018), soil (Ma et al., 2018), sediment and sewage sludge (Loyola-Sepúlveda et al., 2018) as well as biological samples such as pork (Vaccher et al., 2018), fish (Figueiredo et al., 2017), human blood (Kraft et al., 2018) and breast milk (Asamoah et al., 2018). PCBs were found even in the arctic areas (Ubl et al., 2012). In addition to the wide distribution, PCBs are also known for the bioaccumulation in organisms (El Majidi et al., 2013) together with teratogenicity, carcinogenicity and endocrine-disrupting features (Rostami and Juhasz, 2011; Kramer et al., 2012; Wimmerová et al., 2015). Several studies have connected the exposure to PCBs with the adverse neurodevelopment of newborns and neuropsychological impairments like a possible deficit of memory (Obaid et al., 2000; Elnar et al., 2012). Though the strict regulations and prohibitions for the handling of PCBs have been carried out in most countries since they were listed in the protocol of the Stockholm Convention (2009), PCBs are still one of the major environmental concerns (Wang et al., 2017).

As soil is an important reservoir for hydrophobic organic pollutants, the gradual introduction of PCBs certainly aggravates the pollution burden of soil matrix. Both the significant detection and the environmental persistence seem to imply the low removal of PCBs from the soil. The degradation of PCBs by aerobic (Čvančarová et al., 2012) or anaerobic bacteria (Field and Sierra-Alvarez, 2008) suggested the retention may, instead, be connected with some specific interaction in soil, rendering them relatively inaccessible to organisms and further reducing the bioaccessibility. As well known, bioaccessibility (Semple et al., 2005) could be served as the reference for the accurate assessment of the potential hazard imposed on the ecosystem and has attracted more attention of researchers. Lots of work have been carried out especially for the validation of bioaccessibility assessing techniques (Riding et al., 2013; Yan et al., 2017). Considering the time-consuming and costly nature of animal experiments, some physicochemical methods designed as the biomimetic extraction to simulate the interphase transfer and the bioaccumulation have been developed to accelerate the prediction of bioaccessibility in recent years (Lal et al., 2015). Among them, the non-exhaustive extraction using hydroxypropyl-β-cyclodextrin (β-HPCD) appeared to be one of the promising approaches, which showed a good correlation with the most bioaccessible amount of earthworms or bacteria (Umeh et al., 2017). Wong and Bidleman (2010) reported that β-HPCD, as a non-exhaustive extractant for PCBs could distinguish the loosely and strongly bound residues and therefore infer their bioaccessibility in soil. Given that the bioaccessibility was governed by physicochemical and biological properties of ambient environment where pollutants were anchored, for example, the different soils or organisms may show different bioaccessibility for one compound (Hamelink et al., 1994; Kelsey and Alexander, 1997), the use of biomimetic approaches to predict bioaccessibility was strongly recommended by Sijm and coworkers (Sijm et al., 2000). Hypothetically, if the texture and composition of soil and the corresponding temporal and spatial variance that affects the bioaccessibility are considered, it would be better to enhance the applicability of physicochemical methods, such as using β-HPCD for the accelerative prediction of bioaccessibility.

The physicochemical characteristics of soil such as soil organic matter (SOM) content, are mostly responsible for the efficiency of transport process of contaminants in soil and further affect their bioaccessibility (Helena et al., 2015). Irrespective of different SOM characteristics or species, the content of SOM could be used as a standard for the extrapolation of bioaccessibility due to the close relationship between them, as suggested in the European Technical Guideline on Risk Assessment (2003). Except the generally-recognized linearity, the non-linear relationship between the decline in bioaccessibility and the increment of SOM was also observed by Weston (1990) and Cuypers et al. (2002) respectively. However, no detailed explanation was given for the intriguing insight into the correlation of bioaccessibility with SOM. Besides, the influence of other soil essential physicochemical properties which are distinctively depicted by particle size and soil moisture etc. should not be neglected when they are associated with the interfacial transfer and bioaccessibility. Apparently, much research work are still demanded to be done for clarifying the mutual relationship between the multiple soil physicochemical properties and PCBs bioaccessibility, so as to reduce the uncertainties in evaluating the behaviors of PCBs in soil (Terzaghi et al., 2018).

When the bioaccessibility is used as a standard for risk assessment, the duration of soil-contaminant contact that is referred to as aging should be preferentially considered. It has been demonstrated that the aging resulted in a substantial decrease in mobility of pollutants and significantly influenced the bioaccumulation in earthworms (Vlčková and Hofman, 2012). With the aging time experimentally varied from 7 days to 5 months, the relative bioaccessibility of PCB-180 to mouse model sharply decreased from 84.7% to 45.0%, which was due to the gradual reinforcement of interaction between PCBs and soil constituents with the prolonged aging time (Hatzinger and Alexander, 1995; Li et al., 2017). Until now, the influence of aging on bioaccessibility was commonly recognized for hydrophobic pollutants. Nevertheless, the extent to which the bioaccessible fraction of PCBs in soil decays with the aging time or the decay kinetics to finally reach the equilibrium is seldom discussed in the literature. For accurate and rapid characterization of bioaccessibility, however, it is necessary to sufficiently clarify the effects of aging on bioaccessible species of PCBs in soil.

The present study was conducted with the aim to evaluate the effects of aging, soil physicochemical properties and structural descriptors of PCBs on their bioaccessibility in soil. The variance of bioaccessible fraction was distinctively measured with β-HPCD when the soil inherent properties like SOM content, particle size and soil moisture were artificially changed to different gradient levels. The influence of aging on bioaccessibility was also studied with β-HPCD extraction in a fixed ambient soil environment. To distinguish the relative importance of various soil properties, aging effect and PCBs structural characteristics, partial least squares (PLS) analysis was applied for the capability to summarize the correlations between observed features and target variable after dimension reduction (Šilarová et al., 2017). Satisfactorily, the analytic quantitative relationship with bioaccessibility was successfully developed, and it was anticipatorily used as an alternative for predicting bioaccessibility of PCBs in soil. The full evaluation of diverse influential factors would not only help well understand the variation mechanism of bioaccessibility, but also facilitate enhancing the accurate characterization of bioaccessibility of PCBs in soil.

2. Materials and methods

2.1. Chemicals

In consideration that the highly-chlorinated biphenyls possess strong potential of adsorption on the soil constituents and are more sensitive to the changes along with the surrounding conditions (Zhang et al., 1998; Fava and Ciccotosto, 2002; Hu et al., 2016), a

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