Transport and retention of polybrominated diphenyl ether in soil from e-waste dump in Ghana and landfill site in South Africa: A laboratory-scale column soil flushing approach

Eric Akortia[1&2], Mlindelwa Lupankwa[2], Jonathan O. Okonkwo[2]

[1]Environmental Chemistry Research Group, Department of Environmental, Water and Earth Sciences, Tshwane University of Technology, Private Bag X680, Pretoria, South Africa; [2]Ghana Atomic Energy Commission, Radiation Protection Institute, P. O. Box LG 80, Legon, Accra, Ghana. Email (OkonkwoOJ@tut.ac.za).

Introduction

The movement of contaminants through subsurface is usually a complex phenomenon and as a result, contaminant-soil interactions are important for assessing the fate and transport of contaminants through the soil as well as groundwater flow systems[1]. Therefore, an understanding of the mechanisms that control the behaviour of organic contaminants in soil is essential for the development of practices and cost-effective remediation measures[2]. To date, understanding of how the sorption and transport dynamics of organic contaminants influence the mass transfer of contaminants subsurface is unclear. This is especially important to assess the possible contamination risk of shallow groundwater resources and to increase the efficiency of soil remediation. Meanwhile, most researchers have utilised mathematical models and multiple approaches to predict and help understand the transport behaviour of contaminants in soil[3]. To date, understanding of how the sorption and the transport dynamics of PBDEs in soil is unclear. In this study, we evaluated the transport behaviour and the flushing potential of PBDEs congener contaminated soils in two laboratory-scale columns as an optional method for soil remediation.

Materials and methods

Soil samples were collected from an e-waste dump and a landfill site in Ghana and South Africa respectively. The samples were taken from the upper 10 cm of each contaminated site using stainless steel spade after debris and large pebbles removal. The samples were subsequently air-dried and processed through a 2 mm brass sieve, homogenized thoroughly prior to use. All solutions were prepared using analytical-reagent grade chemicals and ultrapure deionized water dispensed from Labostar™ ultrapure water equipment. Stock standards solutions of PBDEs ^{13}C-BDE-77, -139, -209 were used. Three columns with a length of 47 cm, an internal diameter of 5.87 cm and external diameter of 6.51 cm were used for the flushing experiment. One column, packed with glass beads served as a blank; while the other two columns were packed with the homogenized soil samples in increments of about 1–2 cm with gently tapping and completely saturated with 0.01 M solution of pre-dissolved $CaCl_2$, using a peristaltic pump. Prior to packing of soil samples in the columns, the bottom of the glass columns was filled with layers of glass wool followed by about 5 cm layer of glass beads in order to prevent leaching of the soil grains and to obtain clean effluents. The packed columns were then tested for flow rate, and a flow from bottom to top was maintained in all columns to prevent air being trapped in the columns. The flushing experiment was conducted in all columns under the saturated steady-state flow conditions. The column beds were, prior to the flushing period, fortified with a known concentration of PBDE surrogate standards and left to equilibrate in the columns for 24 h. Thereafter, downward percolation of influent solution was applied in order to simulate different rainfall speed patterns which may enhance the leachability of pollutants through the soil. Pump speeds applied were 33 mL min-1, 66 mL min^{-1} and 99 mL min^{-1}. The PBDE compounds were subsequently flushed with about 10 L of deionized water (influent solution) during the entire period of 42 days with the peristaltic pump at a flow rate of 1.2 mL min^{-1}. After termination of the experiment, the soil samples were allowed to drain completely, removed, air-dried away from direct source of light, extracted and analysed for PBDEs retained using the GC-MS (Shimadzu Model 2010 Plus GC). The effluents collected were also analysed using the Shimadzu GC-MS after extraction. Total organic carbon and bulk density were also determined. Several quality control procedures were employed.

Results

The transport of PBDEs through the soil columns was dependent on sorption and the physicochemical characteristics of both soil and PBDEs. BDE-209 in the applied compound mixture exhibited the highest levels, 263.73 ng/L and 431.02 ng/L in the effluent and 11.1 ng g^{-1} and 12.0 ng g^{-1} retained for landfill and e-waste soils respectively (Table 1). This was followed by BDE-139 and finally BDE-77. The estimated equilibrium partition coefficient (K_d) values for the applied compound mixtures in both columns were 0.062 mg kg^{-1} and 0.023 mg kg^{-1} for landfill and e-waste soils respectively.

Table 1 Average concentration of the applied PBDE congeners in both the soil columns and effluents.

Congeners	Flushed (ng/L)		Retained (ng/g)	
	SA Column effluent	GH Column effluent	lfs-SA Column soil	ews-GH Column Soil
BDE-77	4.12	7.66	0.298	0.688
BDE-139	93.53	421.18	11.1	6.98
BDE-209	264.73	431.02	11.1	12.0
∑PBDEs	362.38	859.86	22.5	19.7

SA = South Africa; GH = Ghana; Lfs-SA = landfill soil from South Africa; ews-GH = e-waste from Ghana

Mass balance and ideal mass recovery of the PBDE compounds showed that a significant fraction of the initial contaminant mass (over 95%) was retained in both soils, which articulates the hypothesis that soils act as a sink for organic pollutants (Table 2). However, approximately 1.16% of the contaminant mass was presumably lost through degradation or sorbed onto the pore walls and packing materials other than the soil. This suggests that only a small fraction was available for diffusion.

Table 2 Mass Balance of PBDEs during the column experiment

Column name	Lfs-SA column	ews-GH column
Contaminant	PBDEs	PBDEs
Mass Balance (%)	98.8	96.2
Actual mass flushed (PBDEs) from the soil (%)	0.042	0.097

In addition, the slow movement of pore water through the landfill soil column may have been affected by its high bulk density, 1.33 g cm^{-3}, compared to the e-waste soil with a bulk density of 1.03 g cm^{-3} (Table 3).

Table 3 Physicochemical property of the experimental soils and effluents.

Parameters	SA column	GH column
pH	7.77 (6.66)^a	7.60 (6.4)^a
Conductivity (µS/cm)	140.1 (223.7)^a	96.7 (124.9)^a
Bulk density (g/cm3)	1.33	1.03
Particle density (g/cm3)	2.26	1.93
TOC (%)	5.29	12.4
Porosity (%)	41.2	46.4
Moisture content (%)	4.22	3.77
Partition coefficient (K_d)(mg/kg)	0.062	0.023
Soil mass (g)	174	180

(-)^a = average values determined for the column effluents.

The results of this study demonstrated the strong hydrophobicity of PBDEs and the limit of advection transport of PBDEs in two different soils. Sorption and physicochemical characteristics of the PBDE compounds thus played a key role in controlling its transport in the studied soils. Mass balance estimation of the applied PBDEs revealed that PBDEs were only partly flushed from the soil columns with a significant fraction being retained in the soil. The current study also confirms the poor flushing efficiency of organic contaminants by water as well as the poor solubility of PBDE compounds. This may also be due to other factors such as pH and natural organic carbon. Meanwhile, low pH may configure a higher mass transfer of PBDEs, however, despite the fact that the pH of the effluents was between 6.3–6.8, a significant fraction of the PBDEs remained in the column adsorbent. This may mean that the reduced potential for increase in pH in the soils could result in incomplete dissolution of PBDEs, and also the reason appreciable mass of PBDEs was not mobilized in the transfer zone.

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