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Human exposure to PCBs, PBDEs and HBCDs in Ghana: Temporal variation, sources of exposure and estimation of daily intakes by infants

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ABSTRACT

Human exposure to polychlorinated biphenyls (PCBs) and brominated flame retardants (BFRs) such as polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) was evaluated in Ghana using breast milk samples collected in 2004 and 2009. Mean levels and ranges of PBDEs (4.5; 0.86–18 ng/g lw) and PCBs (62; 15–160 ng/g lw) observed in the present study were unexpectedly high, in spite of the fact that Ghana is a non-industrialized country when compared with many of the Asian and European countries. Significant increases were found in the concentrations of PCBs and PBDEs over the years, while no significant increase was observed for HBCDs. Estimated hazard quotient (HQ) showed that all the mothers had HQ values exceeding the threshold of 1 for PCBs, indicating potential health risk for their children. PCBs in dirty oils and obsolete equipment should be of concern as potential sources in Ghana, and e-waste recycling with little or no experience in safe handling could be a threat to this sub-region noted for unregulated disposal of e-waste. The results may point towards an increase in trends in human milk in Ghana, especially in the larger cities but further analysis would be required to confirm this upward trend in levels. This is the first study to report BFRs in human breast milk from Africa, and undoubtedly from Ghana.

1. Introduction

Human milk is, beyond doubt, the best source of nutrition for infants. Breast milk contains the optimal balance of fats, carbohydrates, and proteins for developing babies, and it provides a range of benefits for growth, immunity, and development (Solomon and Weiss, 2002; Pronczuk et al., 2004). It also contains essential immune factors that help infants fight infections (Oddy, 2001). Breast-feeding builds an essential bond between a mother and her child, and this bond enhances health and well-being across generations. Unfortunately, breast milk is not pristine at present. There is concern about the potential risks posed by the presence of chemical contaminants including the globally distributed polychlorinated biphenyls (PCBs) and brominated flame retardants (BFRs) in human breast milk. Human beings, like other mammals, have a lipid-rich tissue (adipose) that efficiently retains and accumulates lipophilic contaminants (Travis et al., 1988), and they transfer much of the contaminant loads to an offspring during breast feeding, resulting in trans-generational transfer of contaminants (Munoz-de-Toro et al., 2006; Kanja et al., 1992).

PCBs are environmental contaminants which have been used for decades in different applications such as plasticizers, surface coatings, inks, adhesives, paints and flame retardants. Because of their general chemical inertness and heat stability, they have been used in dielectric fluids in transformers and capacitors (ATSDR, 2000). The commercial production of PCBs started in 1929 and increased until 1976, when production and sales were banned in the United States. PCBs were banned in the first half of the 1970s in many countries and have been listed in the Stockholm Convention since 2001 (UNEP, 2009a).

BFRs constitute a diverse group of compounds mainly used to inhibit fire. The representative additive flame retardants, polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) are widely used in plastics, furniture, textiles and electronics such as computers, DVDs and TV-sets. As a result of their environmental stability, persistence and high production volume, PBDEs and HBCDs are among the most abundant BFRs detected in the environment, wildlife and humans (Alaee et al., 2003). BFRs, like PCBs and other persistent organic pollutants (POPs) are lipophilic and have the propensity to bioaccumulate and biomagnify in a food web. It is reported that the main sources of human exposure to PBDEs are through floor dust and food consumption (Sjoedin et al., 2008). Previous studies have shown that food is a major route of human

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exposure for PCBs and other legacy POPs, via fish, meat and dairy products (Duarte-Davidson and Jones, 1994; Fuerst et al., 1990). Unlike PCBs, concentrations of which have steadily decreased in the environment, food and people since their production stopped in 1977, the human body burdens of PBDEs have increased substantially over the last 30 years (Zhu et al., 2009). Due to their relatively low reactivity and high hydrophobicity, PBDEs are persistent environmental contaminants, and certain congeners are bioaccumulative. The tetra- to hexa-brominated congeners exhibit the greatest bioaccumulation and toxicological potentials (De Wit, 2002; McDonald, 2002) and recently, such BDE congeners have been listed as new POPs in the Stockholm Convention.

Although most African countries have ratified the Stockholm Convention, information on environmental pollution by POPs in Africa is scarce, which is not enough to evaluate population exposure levels and changes over time (Linderholm et al., 2010). The recent Global Monitoring Report prepared for the fourth meeting of the Stockholm Convention on POPs in 2009 underscores that there is a knowledge gap on levels and trends of POPs in ecosystems as well as in humans in most developing countries and countries in transition (UNEP, 2009b). Ghana signed and adopted the Stockholm Convention on 23rd of May, 2001 and ratified it on 30th of May, 2003. Preliminary inventories carried out in 2003 to ascertain the status of POPs in Ghana revealed that there is no production of PCBs in Ghana. However, there is illegal or uncontrolled usage of PCB oils in Ghana which have been imported from other countries. In addition, there is no information on PBDEs and HBCDs, albeit Ghana is one of the major dumping grounds for electrical and electronic wastes (e-waste) in Africa and these may be potential sources for BFRs. At Agbogbloshie in Accra, the capital of Ghana, heaps of old computers and their accessories that are unusable are continually being dumped there without any regard for the hazard that they pose to the environment and the people living within the environs of the dump site. In contrast to numerous studies available on environmental pollution by these persistent compounds in developed countries, less information is available in most developing countries, particularly in Africa. Therefore, it is important to start research on BFRs in Ghana. The objectives of this study were to elucidate the contamination status by measuring temporal and spatial variations of contaminant profiles of emerging and legacy POPs (PBDEs, HBCDs and PCBs) in human breast milk from Ghana, and assess the health risk associated with the intake of these contaminants by infants through breast milk.

2. Materials and methods

2.1. Study area and sample collection

Ghana is located on the west coast of Africa with a population of about 25 million. The main sources of income are from mining (gold) and agriculture (cocoa), and on 15th December 2010, the country officially began commercial production of oil having discovered oil in commercial quantities by the American explorers Kosmos Energy in June 2007. Sixty-seven human breast milk samples from primiparous mothers were used for this study. Forty-two samples were collected in 2009 from three geographical locations in Ghana; Accra (coastal) ($n=16$); Kumasi (forest zone) ($n=14$) and Tamale (savannah) ($n=12$). In each city, samples were taken from both urban and rural areas. Prior to this, 25 samples collected from Accra in 2004 were analyzed for assessing temporal variation. Accra is a fast growing large city followed by Kumasi in terms of industrial establishment and infrastructural development. Generally, people from Accra and Kumasi are known for their extravagant lifestyles while the inhabitants of Tamale live largely in poverty. All donors were from the general population who were non-smokers and appeared healthy. Breast milk was expressed by the donors themselves or with the help of midwives into solvent-precleaned glass containers with Teflon-lined screw caps prepared for every individual. The samples were kept frozen and airlifted to the Center for Marine Environmental Studies (CMES), Ehime University, Japan on dry ice and stored in the Environmental Specimen Bank (*es*-BANK) of Ehime University at −25 °C (Tanabe, 2006) until extraction and chemical analyses. The samples were used for analyzing PCBs, PBDEs and HBCDs.

2.2. Ethics, permission and questionnaire

The aim of the study was explained to the mothers and they gave consent to participating in the study prior to donating the samples. In view of the fact that some environmental and physiological factors can influence the levels of the contaminants in human breast milk, all participants answered for a detailed questionnaire on age, weight, height, and dietary habits (Table 1). Area of residence and other data were obtained from each donor as an informative record for subsequent data information. The diet of the participants did not seem to differ much among the studied areas. It was a mixed diet with consumption of meat, fish, milk and dairy products. The donor characteristics of the sampling in 2004 were also similar to 2009 (data not shown). The Ethics Review Committee of the Health Research Unit of the Ghana Health Service approved this study.

2.3. Chemical analysis

Approximately 50 g of the human milk sample was lyophilized and extracted with a high speed solvent extractor (SE-100, Mitsubishi Chemical Analytech) using 50% acetone in hexane. Fat content was determined gravimetrically from an aliquot of the extract. The remaining extract was spiked with $^{13}C_{12}$-labeled PCBs, $^{13}C_{12}$-labeled PBDEs (5 ng each) and 10 ng of $^{13}C_{12}$-labeled HBCDs as surrogates and then subjected to gel permeation chromatography (GPC) for fat removal and eluted with a mixture of hexane/dichloromethane (1:1). The lipid-removed GPC fraction containing organohalogen

Table 1
General demographic characteristics of breast milk donors from Accra (2004, 2009), Kumasi (2009) and Tamale (2009), Ghana.

Parameter	Accra ($n=16$): 2009	Kumasi ($n=14$): 2009	Tamale ($n=12$): 2009	Accra ($n=25$): 2004
Age (years)	23 (18–34)	23 (17–32)	22 (18–26)	28 (21–37)
Weight (kg)	56 (45–75)	55 (45–81)	55 (45–64.2)	64 (50–81)
Height (m)	1.5 (1.49–1.64)	1.6 (1.46–1.66)	1.6 (1.37–1.68)	1.6 (1.47–1.75)
Body mass index (kg/m^2)	24 (19.6–33.3)	23 (19.5–29.4)	22 (17.6–25.6)	25 (21.1–33.3)
*Diet	Mixed	Mixed	Mixed	Mixed
Sex of infant (M/F)	7M/9F	6M/8F	8M/4F	11M/14F
Geographic	Coastal	Forest	Savannah	Coastal
Industrialization	High	Moderate	Low	High

Note: values are means and ranges in parentheses; mixed = fish, meat, egg, milk and dairy products etc.
*Diet.
Accra: Fish (both sea and freshwater) and fish products, meat and poultry-derived products, eggs, milk and milk products.
Kumasi: Fish (both sea and freshwater) and fish products, meat and poultry-derived products, eggs, milk and milk products.
Tamale: Fish (mainly freshwater) and fish products, milk and milk products, meat and poultry-derived products, eggs.

compounds was concentrated and passed through 4 g of activated silica gel packed in a glass column. The first fraction containing PBDEs and PCBs was eluted with 5% dichloromethane in hexane and the second fraction containing HBCDs with 25% dichloromethane in hexane. $^{13}C_{12}$-labeled BDE-126, −139 and −205 were added to the PCB/PBDE fraction and deuterated HBCDs added to the HBCDs fraction as internal standards.

Quantification of PBDEs and PCBs was performed using a gas chromatograph coupled with a mass spectrometer (GC–MS). All the congeners were quantified using the isotope dilution method to the corresponding $^{13}C_{12}$-labeled congeners. Forty-two BDE congeners from mono- to deca-BDE and sixty-two PCB congeners were analyzed in this study based on the methods published elsewhere (Malarvannan et al., 2009; Tue et al., 2010) with slight modification. HBCD isomers (α-, β-, and γ-HBCDs) were quantified using a liquid chromatograph coupled with a tandem mass spectrometer (LC–MS/MS) (Isobe et al., 2007). Concentrations of all the targeted BDE and PCB congeners, and HBCD isomers were summed to obtain the values of $\sum$PBDEs, $\sum$PCBs and $\sum$HBCDs, respectively. Concentrations of PBDEs, PCBs and HBCDs were expressed as nanogram per gram lipid weight (ng/g lw). For quality assurance/quality control (QA/QC), procedural blanks were analyzed simultaneously with every batch of 7 samples to check for any interference or contamination from solvents and glass wares during sample processing. Detection limits for the target compounds were calculated as three times the procedural blank and the mean value was 0.01 ng/g lw for each compound. Recoveries of ^{13}C-labeled surrogates were in the range of 65–110% for PCBs, 78–110% for PBDEs and 103–112% for HBCDs.

2.4. Statistical analysis

Statistical analysis was performed using SPSS (version 12.0, SPSS Inc., Chicago, IL, USA) for Windows. One half of the value of the respective limit of detection was substituted for those values below the limit of detection and applied in statistical analysis. The Mann–Whitney *U*-test was used to compare concentrations of the contaminants among the three locations and between 2004 and 2009 for the Accra samples. The Spearman's rank correlation was used to examine the strength of associations between parameters (concentrations, age and body mass index). A probability value of $p<0.05$ was considered as statistically significant in this study.

2.5. Health risk assessment

The health risk of infants exposed to contaminants in mother's milk was assessed using hazard quotients (HQs). A HQ is defined as the ratio of the estimated daily intake dose (DI) of the compound through breastfeeding to the corresponding maximum acceptable oral dose for human, or reference dose (RfD). A HQ value higher than 1, indicates potential risk. The DIs were calculated based on the assumption that an infant on average weighs 5 kg and consumes 700 g of their mother's milk daily (Oostdam et al., 1999). HQs of BDE-47, BDE-99, BDE-153, BDE-209 and $\sum$HBCDs were calculated using RfD values of 0.1, 0.1, 0.2, 7 (EPA, 2008) and 0.2 μg/kg/day (European Chemicals Bureau, 2007), respectively, while HQs of $\sum$PCBs were calculated using RfD values of 1 (Oostdam et al., 1999) and 0.03 μg/kg/day (ATSDR, 2000).

3. Results and discussion

3.1. Contamination status and spatial distribution

Concentrations of PCBs, which have a longer history and larger amount of usage than BFRs were the highest among the three contaminants in the human breast milk from Ghana (Table 2) followed by PBDEs. PCBs were found in all the analyzed samples from 2009, suggesting wide environmental contamination and human exposure to this contaminant in Ghana.

Total PCB concentrations (sum of 62 congeners) in all the samples varied between 15 and 160 ng/g lipid weight (ng/g lw) with a mean of 62 ng/g lw. Location wise, the means were: Accra (82 ng/g lw), Kumasi (65 ng/g lw) and Tamale (30 ng/g lw). Statistically significant differences were found between Accra and Tamale ($p<0.001$) as well as Kumasi and Tamale ($p=0.002$). The relatively higher concentrations obtained from mothers in Accra are suggestive that inhabitants of Accra are more exposed. The data obtained from all the samples collected from the outskirts of the three cities; Accra, Kumasi and Tamale were pooled together (as rural) and compared with those collected in the three cities (as urban). The comparison between urban and rural areas leads to the understanding of the influence of urbanization on the human contamination by these persistent compounds. Lack of statistical difference between urban and rural locations (Fig. 1) suggests that mothers are uniformly exposed to PCBs regardless of location.

To understand the magnitude of contamination, the mean concentration of total PCBs in human breast milk from Ghana was compared with those reported in different studies in different countries (Table 3). There are only few studies reporting PCB levels in breast milk from Africa. Our mean level (62 ng/g lw) was higher than the value observed in South Africa (10 ng/g lw; Darnerud et al., 2006) and Zimbabwe (26 ng/g lw; Chikuni et al., 1997) but lower than in Tunisia (196 ng/g lw; Ennaceur et al., 2008). Beyond the shores of Africa, our level was higher than some studies in Asian countries but lower than in China, Japan and Europe (Table 3). This indicates the necessity to investigate PCB pollution and the potential sources, and to eliminate PCBs as an environmental contaminant even in Ghana and other African countries where no production of PCBs was reported.

The concentrations of the sum of the 17 congeners ($\sum$PBDEs) in all the breast milk samples from the three locations in 2009 ranged from 0.86 to 18 ng/g lw with an overall mean value of 4.5 ng/g lw. The ranges of concentrations were Accra (1.3–12 ng/g lw, mean of 4.8 ng/g lw); Kumasi (0.89–18 ng/g lw, mean of 5.8 ng/g lw) and Tamale (0.86–4.9 ng/g lw, mean of 2.5 ng/g lw) (Table 2). Statistical difference was found between Accra and Tamale ($p=0.011$), and Kumasi and Tamale ($p=0.01$) indicating that nursing mothers from Accra and Kumasi are more exposed to PBDEs; this could be due to the more significant sources (use of computers, cars, TVs, DVDs, phones, carpets, furniture, etc.) and the high preference for food (dietary habits) (Fraser et al., 2009; Johnson-Restrepo and Kannan, 2009) of people in those two cities. Concentrations of PBDEs in urban milk samples were higher than in rural samples and the difference was statistically significant ($p=0.047$), buttressing the lifestyle of people from the urban cities as stated.

The diets of Ghanaians are mixed and comprise meat (beef, chicken, pork etc.) and fish, among others. Furthermore, Ghana is a coastal country where the consumption of marine fish is high and could be one of the reasons for the relatively high levels of PBDEs. Recent studies have shown that house dust contributes significantly to the exposure of PBDEs in the general population (Stapleton et al., 2008). For example, Lober (2008) showed that about 66% of the overall estimated intake of PBDEs in US was through house dust. Probably, exposure via dust may explain the high PBDE levels found in this study. Coupled with that, the style of eating in Ghana and Africa (generally by hand), could also contribute to the levels if the hands are not properly washed (Stapleton et al., 2008). Concentrations of PCBs and PBDEs were statistically different ($p<0.001$) and the two contaminants were not correlated, suggesting different exposure sources. This may indicate that the two contaminants get into people's bodies through different pathways. As already stated, the principal source of PCB contamination in people seems to be food, particularly fish (Duarte-Davidson and Jones, 1994; Fuerst et al., 1990). For PBDEs, ordinary house dust, containing minute quantities of PBDEs sloughed off from furniture and other household goods may be the principal route of exposure (Lober, 2008).

Interestingly, the mean concentration of total PBDEs in this study was higher than some studies in Asian and European countries, but lower than in Canada and USA (Table 3), a region which consumed much of technical penta-BDE. Despite the fact that Ghana is a non-industrialized country when compared with many of the Asian and European countries, the mean concentration of PBDEs observed in the present study was unexpectedly high.

The sum concentrations of the three isomers (α-, β-, and γ-HBCD) found in the human breast milk samples varied between 0.01 and 3.2 ng/g lw with an overall mean of 0.54 ng/g lw in the present study. The concentrations of HBCDs from the three locations were not statistically different although mothers from Accra had relatively high concentrations. Similarly, differences in concentrations of urban and rural samples were also not statistically significant. Mean levels of $\sum$HBCDs were: Accra (0.80 ng/g lw); Kumasi (0.44 ng/g lw) and Tamale (0.30 ng/g lw). The highest HBCD concentration of 3.2 ng/g lw was detected in a 19-year old mother living in Accra. Differences in concentrations reflect the frequency and extent of HBCD exposure as well as the individual's variability to metabolize these compounds. Human exposure to HBCDs occurs through multiple routes as also in the case of PBDEs. For non-occupationally exposed persons, the major intake of HBCDs is probably from food and indoor air or dust (Sudaryanto et al., 2008). However, the relevance of human HBCD exposure originating from house dust versus food-based HBCD exposure is still scarce (Sudaryanto et al., 2008). With respect to HBCDs, few studies have reported the body burden in humans. The overall mean level of total HBCDs (0.54 ng/g lw) found in our study was higher than in some other studies conducted elsewhere (Table 3), but far lower than in Japan, USA and Belgium, reflecting an extensive usage of HBCDs in those countries.

3.2. Congener profiles and potential source/exposure pathways

Seven prevalent indicator PCB congeners, mainly CB-28, CB-152, CB-101, CB-118, CB-138, CB-153 and CB-180, which are considered as primary indicators of biological

Table 2
Concentrations (ng/g lipid wt.) of major PCB congeners, PBDE congeners and HBCD isomers in human breast milk collected from Accra, Kumasi and Tamale, Ghana.

Compound	Accra (2009)			Kumasi (2009)			Tamale (2009)			Accra (2004)		
	Mean	Median	Range	Mean	Median	Range	Mean	Median	Range	Mean	Median	Range
Lipid content (%)	4.8	4.6	1.7–7.8	4.7	4.2	2.3–8.1	4.1	3.7	2.5–6.9	3.1	3.6	0.23–6.7
CB-28	0.61	0.52	0.21–1.6	1.0	0.52	0.01–4.3	2.4	0.55	0.32–12	0.10	<0.01	<0.01–0.66
CB-74	1.2	0.87	0.38–4.9	1.2	0.80	0.27–4.9	1.5	0.76	0.52–5.4	0.86	0.57	<0.01–5.4
CB-99	2.1	1.9	0.94–3.7	1.7	1.2	0.67–5.9	1.2	0.79	0.51–2.9	0.91	0.61	<0.01–8.7
CB-118	3.0	2.9	1.2–5.8	2.6	1.6	0.98–11	1.9	1.6	0.89–3.6	2.0	1.1	<0.01–14
CB-138	16	14	6.4–31	12	11	4.1–30	5.1	5.0	2.5–13	7.5	4.3	0.32–57
CB-153	22	19	8.2–44	17	15	5.9–40	6.4	5.8	3.1–19	8.8	5.1	0.38–73
CB-170	4.6	3.9	1.4–9.5	3.5	3.0	1.0–7.9	1.2	1.0	0.51–3.2	2.0	1.2	0.07–15
CB-180	11	10	4.0–24	9.2	7.4	2.6–24	2.9	2.4	1.5–7.8	4.5	2.9	0.17–33
CB-183	1.7	1.4	0.68–3.6	1.2	0.99	0.43–2.8	0.43	0.37	0.22–1.0	0.81	0.52	<0.01–5.8
CB-187	5.8	5.5	2.3–13	4.6	3.7	1.4–11	1.5	1.2	0.71–4.3	2.6	1.7	0.12–19
ΣPCBs	82	72	37–160	65	63	22–140	30	26	15–84	34	22	1.7–250
BDE-15	0.030	0.016	<0.01–0.17	0.022	0	<0.01–0.090	0.015	0	<0.01–0.080	0.010	0	<0.01–0.060
BDE-28	0.086	0.080	<0.01–0.24	0.079	0.069	<0.01–0.39	0.048	0.043	<0.01–0.13	0.080	0.050	<0.01–0.42
BDE-47	2.0	1.7	0.31–5.5	2.1	1.7	0.47–5.9	0.77	0.49	0.30–2.3	1.2	0.62	0.21–3.9
BDE-99	0.61	0.32	<0.01–4.0	0.72	0.32	<0.01–3.2	0.13	0.046	<0.01–0.84	0.30	0.18	<0.01–1.9
BDE-100	0.39	0.34	0.080–0.91	0.39	0.25	0.080–1.3	0.090	0.067	<0.01–0.32	0.25	0.12	<0.01–1.4
BDE-153	0.31	0.35	<0.01–0.87	0.34	0.22	<0.01–1.1	0.060	0	<0.01–0.33	0.20	0.19	<0.01–1.0
BDE-154	0.049	0	<0.01–0.20	0.022	0	<0.01–0.12	0.005	0	<0.01–0.070	0.010	0	<0.01–0.16
BDE-183	0.086	0	<0.01–0.90	0.063	0	<0.01–0.27	0.061	0	<0.01–0.28	<0.01	0	<0.01–0.18
BDE-196	0.049	0.043	<0.01–0.15	0.049	0.051	<0.01–0.08	0.024	0.016	0.03–0.070	0.010	0	<0.01–0.040
BDE-197	0.20	0.18	0.090–0.50	0.21	0.19	0.070–0.44	0.20	0.18	<0.01–0.070	0.050	0.030	<0.01–0.19
BDE-206	0.038	0.033	<0.01–0.17	0.061	0.053	<0.01–0.29	0.022	0	<0.01–0.12	0.050	0	<0.01–0.78
BDE-207	0.071	0.056	<0.01–0.31	0.11	0.12	0.040–0.24	0.093	0.073	0.050–0.21	0.020	0	<0.01–0.14
BDE-209	0.83	0.55	<0.01–4.0	1.4	0.39	<0.01–11.2	0.84	0.95	<0.01–1.7	NA	NA	NA
ΣPBDEs	4.8	4.1	1.3–12	5.8	4.3	0.89–18	2.5	2.3	0.86–4.9	2.2	1.3	7.2
α-HBCD	0.79	0.62	0.030–3.2	0.44	0.28	0.090–1.1	0.29	0.23	0.010–0.57	2.2	1.0	<0.01–16
β-HBCD	0.01	<0.01	<0.01–0.080	<0.01	<0.01	<0.01	0.010	<0.01	<0.01–0.090	<0.01	<0.01	<0.01
γ-HBCD	<0.01	<0.01	<0.01–0.020	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01–0.070	0.10	<0.01	<0.01–2.2
ΣHBCDs	0.80	0.62	0.030–3.2	0.44	0.28	0.090–1.1	0.30	0.27	0.010–0.66	2.3	1.0	<0.01–18

Note: NA = Not analyzed; ΣPCBs = sum of 62 PCB congeners; ΣPBDEs = sum of 42 PBDE congeners.

PCB burdens (Gynn et al., 2001) are used as marker compounds to monitor occurrence and distribution of total PCBs (Pauwels et al., 2000). The most predominant congeners in the Ghanaian breast milk were PCB 153, 138 and 180 (Fig. 2) which contributed 25%, 19% and 13%, respectively to the total PCB load. A similar result was observed in other studies around the world (Linderholm et al., 2010; Polder et al., 2009; Ennaceur et al., 2008). In Ghana, plasticizers constitute the largest source of PCB releases. Other open applications include certain paints, fire retardants and lubricants (EPA, Ghana, 2007). There are however, no statistics on national imports of these items to allow the

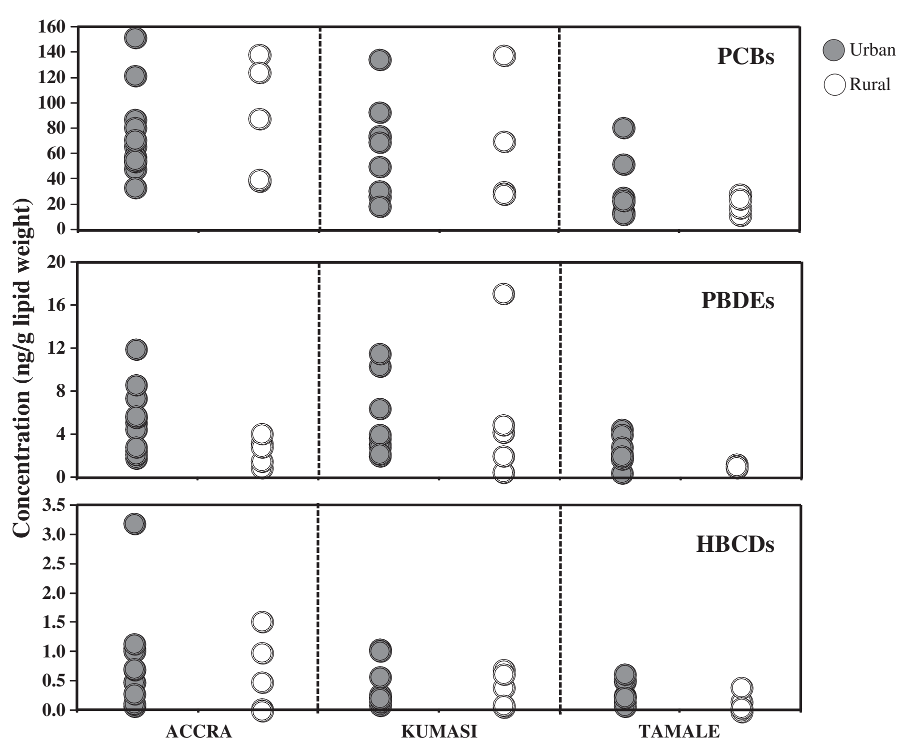

Fig. 1. Concentrations of PCBs, PBDEs and HBCDs from urban and rural locations in Ghana.

Table 3
Mean concentrations (ng/g lipid wt.) of PCBs and BFRs in human breast milk samples from various countries.

Country	Sampling year	*n*	PCBs	PBDEs	HBCDs	References
Africa						
Ghana	**2009**	**42**	**62**	**4.5**	**0.54**	**This study**
South Africa	2006	29	10	NA	NA	Darnerud et al., 2006
Zimbabwe	1997	175	26	NA	NA	Chikuni et al., 1997
Kenya	1991	11	ND	NA	NA	Kanja et al., 1992
Tunisia	2003–2005	237	196	NA	NA	Ennaceur et al., 2008
Asia						
Indonesia	2001–2003	30	27	1.5	NA	Sudaryanto et al., 2008
China	2002	20	42	NA	NA	Kunisue et al., 2004
Vietnam	2007	33	46	0.57	0.33	Tue et al., 2010
Vietnam	2000	42	74	NA	NA	Minh et al., 2004
Philippines	2004	33	70	7.5	0.86	Malarvannan et al., 2009
China	2003–2005	21	206	7.1	0.86	Zhao et al., 2007
Japan	2001–2004	93	120	NA	NA	Kunisue et al., 2006
Japan	2004	30	NA	2.4	NA	Eslami et al., 2006
Japan	2006	Pool	NA	NA	4	Kakimoto et al., 2008
Europe						
U. K.	2001–2003	27	200	7.8	NA	Kalantzi et al., 2004
Sweden	2004	Pool	NA	NA	0.39	Fängström et al., 2008
Sweden	2002–2003	31	111	2.93[a]	0.35	Lignell et al., 2003
Sweden	1997	Pool	NA	4	NA	Noren and Meironyte, 2000
Poland	2004	22	153	2.5	NA	Jaraczewska et al., 2006
Italy	2000–2001	10	240	4.1	NA	Ingelido et al., 2007
Norway	2000–2002	29	172	3.8	0.13	Polder et al., 2008a
Belgium	2006	197	89	2.1	1.5	Colles et al., 2008
Russia	2002	23	191	1.1	0.71	Polder et al., 2008b
Russia	2003–2004	33	240	0.96	NA	Tsydenova et al., 2007
USA and Canada						
USA	2004	38	NA	76	NA	Johnson-Restrepo et al., 2007
USA	2002–2003	8	NA	NA	3.8	Ryan et al., 2006
USA	2002	47	NA	74	0.5	Schecter et al., 2003
USA	2002–2003	40	126	50.4	0.5	She et al., 2007
Canada	2001–2002	20	NA	43	NA	Ryan et al., 2002

NA: No available data.
ND: Not detected.
[a] : Mono- to hepta-BDEs only.

estimation of potential volumes of such uses. A National Implementation Plan report in Ghana (EPA, Ghana, 2007) indicated that there is enough evidence to suggest that workers in some industries have been exposed to PCBs as a result of bad practices such as using empty transformer oil drums as water reservoirs. Further, PCB oils referred to as 'dirty oil' also finds its way into small-scale industries where they are used to produce pomade (a greasy or waxy substance that is used to style hair) and sold on the local markets for other applications (EPA, Ghana, 2007). The higher relative concentration of the more volatile and less lipophilic lower-chlorinated congeners such as CB-28, -74 and -99 in the samples from Tamale (Fig. 2), resulted in a distinctive congener profile of PCBs compared with those in the other cities and could be due to these PCBs being redistributed through air and air borne particles as the region is prone to the yearly effect of the Harmattan wind which picks up fine dust particles from the Sahara desert. The source of PCBs in the Sahara desert could be atmospheric deposition from Europe and off the West African coast. In a study to elucidate the role Saharan dust may play in the degradation of Caribbean ecosystems, Garrison et al. (2006) observed that air samples from Mali (West Africa) contain a greater number of PCBs, pesticides and polycyclic aromatic hydrocarbons (PAHs) and in higher concentrations than the Caribbean sites. Gioia et al. (2011) also indicated in their study that there are major emissions of PCBs and different source types including ships' graveyard in parts of West Africa than accounted for in current global atmospheric emissions estimates.

Among the PBDEs; BDE-47, -209, -99, -100, and -153 were the dominant congeners (Fig. 3). BDE-47 was found to be the predominant congener in the samples in accordance with other studies dealing with human matrices, such as breast milk (Tue et al., 2010; Daniels et al., 2010; Malarvannan et al., 2009; She et al., 2007) and blood

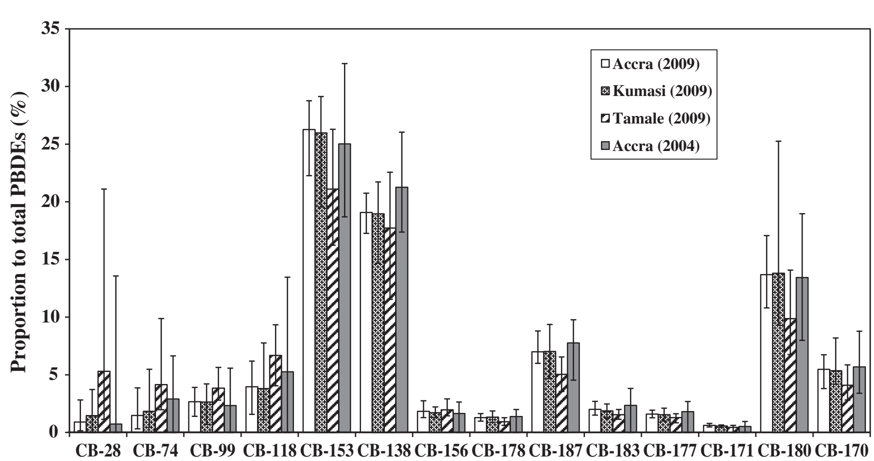

Fig. 2. PCB congener profiles in human breast milk from various locations in Ghana (vertical bars show the range and mean of each congener to total PCB concentration).

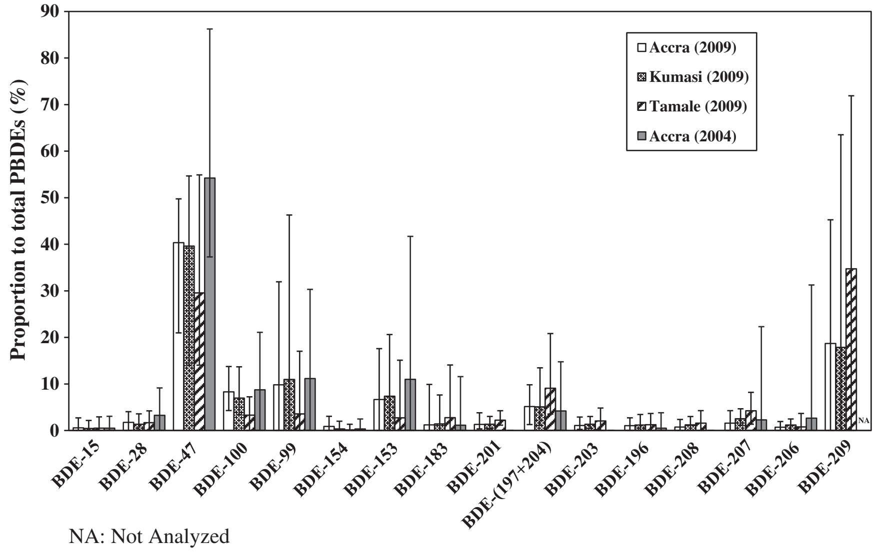

Fig. 3. PBDEs congener profiles in human breast milk from various locations in Ghana (vertical bars show the range and mean of each congener to total PBDE concentration).

plasma (Mazadai et al., 2003). Because BDE-47 is a major constituent of the PentaBDE technical mixtures, its preferential accumulation in the breast milk from Ghana may indicate that PentaBDE technical mixtures have been used in Ghana. Therefore secondary exposure through diet may be the major pathway of the PentaBDEs. Interestingly, BDE-209 was found in some donor mothers at elevated levels compared to BDE-47 in the 2009 samples, but unfortunately we did not analyze BDE-209 in the 2004 samples (Fig. 3). As observed in this study, levels of BDE-209 were also high (up to 23 ng/g fat) in human serum samples from Guinea-Bissau in West Africa, while the other congeners were low (Linderholm et al., 2010). The elevated BDE-209 levels clearly represent ongoing exposure, since the half-life of BDE-209 in humans is very short, estimated in the range of 15 days (Thuresson et al., 2006). In humans, the exposure routes are different for BDE-47 relative to higher brominated congeners like BDE-183 and BDE-209. Food is generally a more important source for BDE-47 because it is the major contaminant in wildlife tissues, whereas dust ingestion seems to be more prominent for BDE-183 and BDE-209 (Sudaryanto et al., 2008).

This study has revealed the exposure of some mothers to higher brominated BDEs which is often attributed to a sustained exposure to Octa and DecaBDE technical mixtures at elevated levels, especially in an occupational context as has been observed in occupationally-exposed workers such as electronic dismantling and rubber factory workers (Sjoedin et al., 2003) and e-waste recycling workers in China (Bi et al., 2007). Therefore, the presence of higher brominated BDEs in breast milk of the mothers in Ghana could be an indication of continuous exposure to Octa and DecaBDE technical mixtures arising from uncontrolled e-waste activities. Large variations in relative concentrations of the congeners observed within each city suggest that the toxicokinetics of these compounds may be different in individual donors (Tue et al., 2010) or human exposure to these contaminants may vary with different microenvironments possibly depending on the composition/duration of usage of the flame retardant goods used by the individual.

In this study, the stable isomer, α-HBCD was predominant as also observed in other studies (Shi et al., 2009; Malarvannan et al., 2009; Kakimoto et al., 2008; Covaci et al., 2006), supporting the fact that α-HBCD is more persistent and bioaccumulative. Among HBCDs, α-HBCD is the predominant diastereoisomer in human milk samples, probably due to selective metabolism or biotransformation process. We also detected small amounts of β- and γ-HBCDs in a few of the urban milk samples, possibly indicating recent exposure, but none in the rural milk samples. Among HBCDs, α-HBCD was reported to have a relatively longer environmental and biological half-life than β- and γ-HBCDs. Technical grade HBCD consists of 70–95% γ-HBCD and 3–30% of α- and β-HBCDs (Tomy et al., 2004). While the level of γ-HBCD in sediments appeared to be higher than or the same as that of α-HBCD the dominant isomer in the biota was α-HBCD (Covaci et al., 2006). This alteration in composition is partially due to variations in water solubility (α-HBCD, 48.8; β-HBCD, 14.7 and γ-HBCD, 2.1 μg/l) (Morris et al., 2004; Hunziker et al., 2004) and uptake and metabolism of individual stereoisomers (Covaci et al., 2006). In contrast to the dominance of α-HBCD in this study as well as others, some studies have reported a higher percentage of γ-HBCD in human tissues, such as adipose tissue (Johnson-Restrepo et al., 2008) and serum samples (Thomsen et al., 2007). Eljarrat et al. (2009) recently reported the dominance of γ-HBCD in 24 of 30 Spanish breast milk samples, whereas α-HBCD was predominant in the remainder. Higher percentage of γ-HBCD has also been seen in occupationally exposed workers, with γ-HBCD making up to 40% of ΣHBCDs (Thomsen et al., 2007). Although the reasons for the different isomer profiles in human tissues from different studies are not yet clear, it is reasonable to hypothesize that they arise from a combination of differences in external exposures and inter-individual variations in metabolism. The largest release of HBCDs is estimated to be contaminated water from production of insulation boards, water and air from textile coating and diffuse release during the life cycle of insulation boards and textiles (UNEP, 2010). Judging from this, the small-scale textile industries dotted across Ghana could be a potential source of exposure.

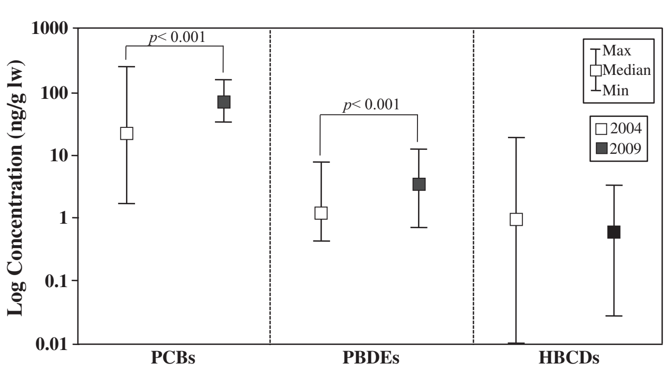

Fig. 4. Temporal variation of PCBs, PBDEs and HBCDs between 2004 and 2009.

3.3. Temporal variation

The mean total PCB concentrations in Accra in 2004 (34 ng/g lw) was lower than the mean in 2009 (82 ng/g lw) and the difference was statistically significant ($p<0.001$) (Fig. 4), possibly indicating that PCBs have increased in the Ghanaian environment within a short period of five years. A report by Ghana's EPA (2007) emphasized that in Ghana, the main potential sources of PCB-containing applications at electric distribution networks, industrial facilities, residential and commercial buildings were found to be transformers and capacitors. In an attempt to know if there was any possible temporal variation, total PBDEs varied between 0.47 and 7.2 ng/g lw (median of 1.3 ng/g lw) in 2004 and from 1.3 to 12 ng/g lw (median of 4.1 ng/g lw) in 2009. In

Table 4
Calculated mean and range values of infants' daily intakes of BDE-47, BDE-99, BDE-153, BDE-209, HBCDs and PCBs through milk in Ghana.

Contaminant	Mean	Range
BDE-47	0.010	0.001–0.039
BDE-99	0.003	<0.001–0.034
BDE-153	0.002	<0.001–0.007
BDE-209	0.005	<0.001–0.041
ΣHBCDs	0.003	<0.001–0.031
ΣPCBs	0.40	0.065–1.0

Note: Values are in μg/kg/day.

comparing the same congeners (up to nona-BDE) between 2004 and 2009, concentrations of PBDEs in 2009 were significantly higher ($p<0.001$) than those in 2004 (Fig. 4), indicating that PBDE levels might have increased in Ghana during this time. Accra is a coastal city and hence the consumption of fish is high compared to Kumasi and Tamale. Aside from this, the unregulated disposal of e-waste and continuous e-waste dismantling activities in Accra could be a major reason for the increase from 2004 to 2009. However, further time-series studies are needed to ascertain these temporal trends in Ghana.

The median concentrations of HBCDs from Accra were 1.0 ng/g lw (2004) and 0.62 ng/g lw (2009) but the difference was not statistically significant ($p=0.141$) (Fig. 4).

3.4. Correlation between contaminants and age/body mass index

In lactating women, factors such as parity, age of mother, food intake preferences and period of breast-feeding have been reported to contribute to the concentrations of lipophilic contaminants in human breast milk (Harris et al., 2001). In this study, we examined the correlation between concentrations of PCBs and age of the mothers. No association was found between levels of PCBs and age or body mass index (BMI) (figure not shown). The lack of correlation could possibly be due to the sample size in the present study. Nevertheless, our results were similar with other studies (Yang et al., 2002; Minh et al., 2004). In lactating mothers, several factors have been associated with the concentrations of organochlorines (OCs) in human breast milk such as food intake preference, age, number of children and other external factors (Harris et al., 2001). For instance, reduction in body burden/tissue concentration of OCs may be due to a combination of metabolism and various excretion routes including milk production/giving birth, etc. while changes in body fat storage may also alter tissue concentration (Juan et al., 2002).

Similar to PCBs, PBDEs and HBCDs did not also correlate with the age and BMI of mothers, as also observed by Schecter et al. (2003) and Sudaryanto et al. (2008). This suggests that there are other factors controlling the variability of these contaminants in human breast milk, or could relate to contemporary exposure to these chemicals in the general population. Thus, the levels in the general population may be due to current exposure/condition rather than their age-dependent accumulation.

3.5. Estimation of daily intakes for infants

One of the aims of this study was to assess the body burdens of contaminants in breast-fed infants. Table 4 shows the calculated infants' daily intakes of BDE-47, BDE-99, BDE-153, BDE-209, HBCDs and PCBs through breast milk in Ghana. The DIs of BDE-47, BDE-99, BDE-153, BDE-209 and HBCDs, were below the reference doses for chronic oral exposure values, implying minimal risk caused by PBDEs and HBCDs at present. However, some nursing mothers had DI values exceeding or close to the RfD (1 μg/kg/day) for PCBs as derived by Health Canada (Oostdam et al., 1999). Moreover, when the minimal risk level of 0.03 μg/kg/day for PCBs (ATSDR, 2000) was considered, the DI values of all the nursing mothers grossly exceeded the RfD, indicating potential health risk to their children. The study highlights the need for further monitoring of these contaminants in Ghana using matrices like fish, food and dust to elucidate the source(s) of exposure and evaluate possible long-term impacts to human health, especially infants.

4. Conclusions

PCBs, PBDEs and HBCDs measured in human breast milk samples in the present study indicate the presence of these persistent contaminants in Ghana. Lack of statistical difference between urban and rural locations was observed for PCBs, PBDEs and HBCDs. However, higher levels of these contaminants were observed in Accra and Kumasi compared to Tamale indicating that inhabitants of Accra and Kumasi are more exposed. This indicates that there are significant sources of these compounds in the more industrialized areas. Significant differences were found in the concentrations of PCBs and PBDEs between 2004 and 2009, while no significant differences were observed for HBCDs. Further studies using fish, food and dust samples may provide further information to explicate the main route(s) of human exposure to PCBs and BFRs in Ghana. Although discussions on the safety of breast-feeding have come to the firm conclusion that exclusive breast-feeding should continue to be promoted, continued breast milk monitoring programs are important to detect higher exposures of the population to PCBs, BFRs and other POPs. The export of e-waste from developed countries to Ghana and processing the same with little or no experience in safe handling could be a threat to the country noted for unregulated disposal of e-waste. PCBs in dirty oils and obsolete equipment should be of concern as potential sources in Ghana. In the near future, studies should be conducted on workers at the e-waste dumping site and other uncontrolled industrial processes in Ghana, and nursing mothers living within the periphery of such potential sources. To our knowledge, this is the first study to report BFRs in human breast milk from Africa.

Conflict of interest

The authors declare that they have no conflict of interest.

Acknowledgments

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