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## Review

# DDTs, PCBs and PBDEs contamination in Africa, Latin America and

# South-southeast Asia—a review

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**Abstract:** Levels of polybrominated biphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs), and dichlorodiphenyltrichloroethane and its degradation products (DDTs) in the environment (ambient air, soil and aquatic mammals) and in humans from the developing regions (Africa, Latin America, and South-southeast Asia) are reviewed. Higher DDTs levels in certain parts of the developing regions due to agricultural applications and disease control measures are evident. The data however do not indicate higher levels of PCBs and PBDEs in the developing regions compared to developed countries. We also compared globally the levels of these chemicals in human milk sampled since year 2000. Human milk data again showed higher DDTs levels in the developing regions do not exceed levels found in the developed countries, data suggest the levels of PBDEs in the developing regions may be on the rise.

Keywords: persistent organic pollutants; developing regions; developed regions; trends; breast milk

# 1. Introduction

Polychlorinated biphenyls (PCBs), DDTs, which include dichlorodiphenyltrichloroethane (DDT) and its degradation products dichlorodiphenyldichloroethylene (DDE) and dichlorodiphenyldichloroethane (DDD), and polybrominated diphenyl ethers (PBDEs) are persistent organic pollutants (POPs) and are bioaccumulative. They can be transported over long distances. These chemicals also show toxicological responses in human and wildlife. PCBs and DDTs are commonly referred to as legacy POPs, while PBDEs as emerging POPs [1].

PCBs act in humans on multiple organs and organ systems including the liver, kidneys, pancreas, immune and nervous system functions and sex steroid hormonal systems, leading to increased risk of cancer, diabetes, liver disease, infertility, ischemic heart disease and hypertension [2]. A weak

epidemiologic association has been identified between low-level perinatal exposure to PCBs and effects on cognitive and motor development of children [3]. Co-planar PCBs exhibit teratogenic properties and are also endocrine disrupters [4]. Gestational and lactational exposures to co-planar PCBs have been associated with impaired cognitive development and behavioural problems [5]. Commercial use of PCBs started around 1929 and steadily increased until 1979 when production and sales were banned in the United States. The substances were subsequently banned in many other developed countries.

DDTs belong to a group of organochlorine pesticides (OCPs) that have been used extensively in agriculture and for controlling disease vectors. Experimental and epidemiologic evidence have associated DDTs with various cancers such as those of the pancreas and liver as well as with disruption of the endocrine system [6]. In a United States study, DDTs have been associated with premature births [7]. Chronic, low level exposures to DDTs have been associated with a broad range of non-specific symptoms such as headache, dizziness, fatigue, weakness, nausea, chest tightness, difficulty breathing, insomnia, confusion, and concentration difficulties [8]. Occupational exposure to DDTs has been associated with reduced verbal attention, visuomotor speed, and with increased neuropsychological and psychiatric symptoms among retired workers aged 55–70 years in Costa Rica [9]. DDTs were banned in developed countries in the 1970s and 1980s due to their negative impact on non-target organisms and bioaccumulative potential in biota and humans [10,11].

PBDEs are brominated flame-retardants that have been widely used in upholstered furniture containing polyurethane foam, in insulation material for wires and cables and in high impact polystyrene in electronics and computers [12]. PBDEs have endocrine disrupting effects [13,14]. Because of their ubiquitous presence, bioaccumulation and potential toxicities to wildlife and humans, PBDEs (excluding the DecaBDE technical mixture) have been added to the list of POPs in the Stockholm Convention [15]. The European Union imposed a ban in 2004 on the production, use and import of PentaBDE and OctaBDE. In the United States, efforts are underway to phase out DecaBDEs by all current producers by the end of 2013.

Regulations now exist for many legacy and emerging POPs, and monitoring spatial and temporal changes is important to follow the results of regulation as well as for monitoring exposure risks and linking these to possible effects [16]. Compared to countries in the developed region, monitoring data of POPs remains inadequate in the developing regions. While informative reviews on countries and regions in the developed region have been conducted [15-18], a systematic review of POPs in developing regions as a whole is lacking. The main objective of this paper is to carry out a comprehensive review on the levels of aforementioned POPs in ambient air, soil and aquatic mammals as well as humans from developing regions, and compare those with countries in the developed region to provide a global perspective on the contamination of these POPs. The developing regions included are Africa, Latin America, and South-Southeast (S-SE) Asia. POPs in China was not included in this review as there are already several published reviews on the subject from China, including a recent review on levels of POPs in general population in China and the development of management policies in China to deal with the pollutants [19]. POPs contamination in China's water has also been reviewed [20].

Figures constructed for this manuscript are presented according to how they are discussed. In some cases, more than one study from a single country is used to demonstrate either spatial or temporal, or both, changes. In order to compare levels reported from different studies, only the central values that are reconstructed from each study are presented in all figures. A central value

(dotted in diamond form) is an arithmetic mean (AM), a geometric mean (GM) or a median value. Some studies only reported range of concentrations, in this case 1/3 of the range was used as the central value, as most of the environmental data is log-normal distributed and 1/3 is roughly in the middle of a log scale. Levels in soil are ng/g dry weight (dw) and in ambient air is ng/m<sup>3</sup>. For aquatic mammals and human blood and milk, values (ng/g lipid weight (lw)) are on the basis of sample content of extractable lipids. Levels of POPs in all figures are shown in log scale.

Levels of DDTs, PCBs and PBDEs are summarized in figures 1, 2 and 3 respectively. Each figure is further divided into a, b and c for different environmental matrices. Levels of POPs in human milk are summarized in Figure 4. Different colors are used in Figure 4: blue for developed regions, red for Africa, green for S-SE Asia, and purple for Latin America. The references for all the data used in the figures are mentioned in the text. We selected soil, ambient air and aquatic mammal as three representative media to evaluate POPs in the environment. Data on ambient air and soil from the developing regions are relatively available compared to other environmental media; besides, they are also important exposure routes to the general population. Aquatic mammals are apex predators, with long life expectancy rates; thus making them sentinel or indicator species for environmental perturbations [21]. No specific time period was applied during the data collection phase. Nonetheless, breast milk data focused on post 2000 samples in an effort to give current situation of human exposure. This review focused mainly on spatial comparisons, though temporal comparisons are also considered when data are available. Among OCPs, only DDTs is covered because it is increasingly relied upon in many regions of Africa and Asia where malaria remains a problem [22].

#### 2. Levels of POPs in the environment in developing regions

## 2.1. DDTs

Levels of DDTs in soil from Senegal and Gambia [23], Congo [24], Tanzania [25], Uganda [26,27], and Ethiopia [28] in Africa; Brazil [29] and Chile [30,31] in Latin America; and Vietnam [32] and India [33] in S-SE Asia are summarized in Figure 1a. In general, levels of DDTs in soil samples from both high and low DDT application areas were in the range of 10–1,000 ng/g, with exceptions of background soil samples from Uganda (< 1 ng/g) and with some soil samples from Chile (> 1,000 ng/g). The higher levels in Chile and India can be attributed to the fresh application of DDT to curb the spread of malaria [30,31]. For comparison, levels from Canada [34] and from the Corn Belt of the United States [35] were at 10 ng/g and less (Figure 1a).

Low application and high application areas reflect the intensity of use of DDT and this intensity can vary significantly between countries, between different areas within a country, as well as between different regions of the world. Examples of high application areas include those where DDT was used against pests in agriculture, for the control of malaria (for example through indoor residual spraying programs), and former pesticide storage areas because of possible leaching of DDTs to the soil. Low application areas are the general urban, rural and background areas. Two studies in Gambia [23] and Tanzania [25] provided clear evidence of associations between the levels of DDTs in the soil and the intensity of DDT use by observing the following concentration gradient: the highest levels were found in DDT storage areas, followed by agriculture areas with large scale of DDT application and then by city farms with some degree of DDT use, the lowest levels were reported in soil samples from background areas where there was no DDT use. While some POPs such as PCBs and PBDEs showed strong urban-rural gradients in soil contamination [36], levels of DDTs did not show such an urban-rural gradient due to the presence of local sources of the chemical in rural areas [34]. However, in general DDTs in soil of urban areas were 1–2 orders of magnitude higher than those in rural areas when rural sources were excluded [37,38].

DDTs in ambient air are presented in Figure 1b. Data are compiled from Congo [39], Malawi [40] and Zimbabwe [41] in Africa; southern Mexico, the Brazilian mountains and Costa Rica [41,42,43] in Latin America; and India, Vietnam, Thailand and the Solomon Islands in S-SE Asia [44]. Central values of DDTs in ambient air samples from urban areas were in the range of 1–10 ng/m<sup>3</sup>, while less than 0.1 ng/m<sup>3</sup> was found in rural areas. This correlates with the difference in DDTs levels in rural and urban soils as we discussed earlier. Soils act as a continuing emission source of atmospheric contamination through volatilization [39]. Air samples from the remote Brazilian mountains had the lowest levels of DDTs (< 0.1 ng/m<sup>3</sup>). Presence of this low background level could be largely due to long range transport of DDTs from other areas, although some unknown local sources might be present too.

Levels of DDTs in ambient air samples from several countries in the developed region such as Japan [45], Canada [46,47], Australia, Sweden and the Baltic Island [41], were also included in the figure and levels in these developed countries were generally lower at 0.01–0.1 ng/m<sup>3</sup>. Most of the developing countries described in this review are located in the tropical regions where climatic conditions favor dissipation of DDT and other chlorinated pesticides. Continuing use of DDT in indoor residual spraying programs in some parts of the region and emissions from old pesticide storage areas may be attributed to the higher DDTs levels in the developing regions [37].

Central values of DDTs in aquatic mammals from Argentina [48], Brazil [49], East Greenland [50], Hawaiian Islands [51], India [52,53], McMurdo Sound-Antarctica [54], Norway [50], Russia [55,56,57], South Africa [58], Tanzania [59], Western Sahara [60] are summarized in Figure 1c. Levels of DDTs in the developing regions (100–100,000 ng/g) varied quite a lot and the upper levels (100,000 ng/g) were an order of magnitude higher than those in the developed countries (10,000 ng/g). For aquatic mammals, many factors including species, age, and gender and tissue type can influence the level of DDTs [61]. While DDE was reported to be the major congener of DDTs in the developed regions, most of the studies from the developing regions reported higher levels of the parent compound (DDT) compared to its metabolites (DDE or DDD) in biota. High levels of DDTs in Brazil and India could be attributed to the fact that the sampled aquatic mammals inhabited riverine and estuarine ecosystems that were in close proximity to pollution sources [49,52]. Levels of DDTs in India however, showed a 3- to 10- fold decrease between 1993-1996 and 2000-2001. This corresponds to the decrease in use of DDT from 19,000 tons/year in the early 1990s to 7,000 tons in 2001 to 2002 [49]. Although levels of DDTs in samples from South Africa and Western Sahara fringe were similar to those from developed countries, the former samples contained higher DDT to DDE ratios suggesting fresh DDT applications in the area [60].



Figure 1. Levels of DDTs from countries in the developed and developing regions: (a) levels in soil, ng/g dry weight; (b) levels in ambient air, ng/m<sup>3</sup>; (c) levels in aquatic mammals, ng/g lipid weight. Year of sample collection is indicated in bracket. Central values (\$ = AM;  $\pounds = 1/3$  range; ¥ = GM; € = Median; \* = single data point) are presented. The bar indicates minimum and maximum range if reported in the literature. H.A = High Application; L.A = Low Application; Bgrd = Background. S-SE Asia = South and Southeast Asia.

#### 2.2. PCBs

Many countries in the developing regions have several PCB sources such as uncontrolled (open) burning of municipal waste, PCB-containing waste imported from developed countries and the use of PCBs in condensers and transformers [62]. So far, environmental monitoring of PCBs in the developing regions is not well developed; only limited data are available. Central values of PCBs in soil collected in urban and industrial areas from South Africa [62,63], Chile [64] and Brazil [65] were in the range of 1–10 ng/g, while the levels in Vietnam [66,67] were between 10 and 100 ng/g (Figure 2a). A general concentration gradient of PCBs in soil (industrial/urban area > agricultural area > background area) was observed in the cases of South Africa [62,63] and Vietnam [66]. This can be expected as samples were collected from urban and industrial areas that are characterized by diverse sources of PCBs than the surface soils (0–0.5 cm in depth), due to possible higher levels of organic carbon in surface soils and atmospheric deposition [62]. On the contrary, lower PCB levels (0.1–1 ng/g) were observed in background areas from Africa, South America and Asia [68].

Background levels of PCBs from Europe and North America however, were about 10 times higher than, while levels in Australia were similar to, the background levels in the developing regions [68]. This is likely due to the fact that countries in the northern hemisphere consumed an estimated 97% of globally produced PCBs between 1930 and1993 [69].

Figure 2b illustrates levels of PCBs in ambient air. Data were collected from Ghana and South Africa [62], the Ivory Coast, Gambia and Cape Verde [70], Southern Mexico [42] and Chile [71], Vietnam, India and the Solomon Islands [41]. Data from countries in the developed region such as Switzerland [72], United Kingdom [73], Japan [74], Spain [75] and the Great Lakes area and Chicago in the United States [76] were also included in the figure. Ambient levels of PCBs in the developing regions were either lower than (in the case of Africa and Latin America) or similar to (in the case of S-SE Asia) those found in countries in the developed region.

The higher PCB levels were found in ambient air from S-SE Asian region (Vietnam, India and The Solomon Islands) than other two developing regions. This echoes the relatively high PCB levels found in soil from countries in S-SE Asian region like Vietnam (Figure 2a). It may also be due to the fact that the air samples from Vietnam, India and Solomon Islands were collected a decade earlier (1989–1991) than the other air samples. The extensive use of PCB transformers and capacitors in the S-SE Asian region might be a contributing factor. Dielectric oils containing PCBs are purportedly still widely used in some countries within the developing region [66,67]. In general, PCB levels in urban areas were higher than those in the rural areas. This can be attributed to the production and application of PCBs in urban areas [77,78]. One exception is however the data from Southern Mexico (2002–2004) and India (1994) (Figure 2b), where PCB levels were higher in rural areas. This

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reversed situation may be due to episodic emissions from non-point sources such as burning of municipal waste, which are common practice in rural areas in the tropical countries [79].

Levels of PCBs in the same or closely related species of aquatic mammals from Argentina [48], Brazil [80,81], Canada [50,82], India [52,53,83], Mauritius[61], Russia [56,84], USA-Florida Coast [85], Western Sahara [60] are shown in Figure 2c. All except one study reported tissue levels in blubber. Blubber is the primary storage location of fat in most aquatic mammals and POPs such as PCBs and DDTs show positive correlation with fat content of the mammals [48]. Generally, PCB levels in aquatic mammals in Africa were in the range of 100–1,000 ng/g, in Latin America were 10–10,000 ng/g, and in S-SE Asia were 100–10,000 ng/g. In comparison, levels were one to two orders of magnitude higher (1,000–100,000 ng/g) in countries in the developed region. The 10 times higher levels of PCBs in aquatic mammals from Florida coast in the United States than those in the neighboring Latin America exemplify higher PCBs levels in aquatic mammals from the developed regions. Very high levels in aquatic mammal samples from Vietnam compared to the other countries could be attributed to the usage of electrical equipment containing PCBs and the use of PCBs in artillery and other chemical weapons during the second Indo-Chinese war (1961–1971) [86].

Despite the fact that PCBs have been banned in developed countries for more than 30 years, levels of PCBs in soil, air and aquatic mammals from developed countries were generally higher than those in the developing region. Especially, aquatic mammals at the top of the food chain are yet to show meaningful declining levels [82]. This might be as a result of heavy use of PCBs in the developed regions in the 1960s and 1970s.

### 2.3. PBDEs

Data on PBDEs in environmental media from Africa, S-SE Asia, and Latin America are very limited, particularly for their levels in soil. As a result, we were not able to construct a summary figure for levels of PBDEs in soil. Only the levels in ambient air and in aquatic vertebrates are presented and discussed in this section.

PBDEs in ambient air from Argentina [87], Botswana [87,88], Costa Rica [88], India [89], South East Asian Sea [90] and Vietnam [90] are presented in Figure 3a. For comparison, levels of PBDEs in England and Ireland [91], the United States [78], East Greenland Sea [90], South West Greenland [93], Norway–Svalbard [87] and Canadian and Russian Arctic [17] are included in the figure as well. With the exception of the higher levels in the Canadian Arctic, which was collected in the early 1990s, PBDEs levels in ambient air were in the range of 0.001–0.1 ng/m<sup>3</sup> and there was no clear spatial difference between developing regions and countries in the developed region. Offshore atmospheric levels (Southeast Asian Sea, East Greenland Sea and South West Greenland) were in the range of 0.0001–0.001 ng/m<sup>3</sup>, which were one to two orders of magnitude lower than the levels on land [90].

It is important to note that data from Botswana study showed a possible increase in atmospheric PBDE levels from year 2004 (0.006 ng/m<sup>3</sup>) to 2007 (0.04 ng/m<sup>3</sup>) in the region [87,88]. Some Latin American and S-SE Asian countries are primary destinations for electronic wastes (e-wastes) from Europe and North America for recycling, which may attribute to the levels of atmospheric PBDEs in these regions [89]. Local burning of municipal waste can also be a major contributor to atmospheric PBDEs, particularly in the non- source regions like the Arctic and Subarctic communities in Canada [17].



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Figure 2. Levels of PCBs from countries in the developed and developing regions: (a) levels in soil, ng/g dry weight; (b) levels in ambient air, ng/m<sup>3</sup>; (c) levels in aquatic mammals, ng/g lipid weight. Year of sample collection is indicated in bracket. Central values (\$ = AM;  $\pounds = 1/3$  range; ¥ = GM; € = Median; \* = single data point) are presented. The bar indicates minimum and maximum range if reported in the literature. S-SE Asia = South and Southeast Asia.

Possible evaporation of PBDEs under warm environment could be responsible for levels of PBDEs in the tropical areas where many developing countries are located [94].

Although there is a general lack of soil data of PBDEs for the general environment, PBDEs were monitored in soils around the vicinity of known point sources. For example, Eguchi et al. [95] identified crude recycling of e-waste as the main source of soil pollution by PBDEs in Asian developing countries after analyzing samples from 32 stations around e-waste disposal sites in India, Vietnam, Malaysia, Indonesia and Cambodia during 1999–2004. The contribution of leachates from landfills to the overall environmental burden is also highly significant due to the fact that most consumer products containing brominated flame retardants eventually end up in landfills [96]. Leachate samples collected from the same period from different landfills in South Africa showed wide variations in PBDE levels [97,98]. Levels of PBDEs in sediment samples collected from the estuary of Saigon-Dongnai River, suggesting a movement of PBDEs from urban sources to the surrounding aquatic environment [99]. PBDE levels in eggs of different bird species showed great variation in congener patterns; reflecting differences in trophic levels, migratory behavior, and distance to the source and exposure to different PBDE mixtures, with higher PBDE levels presented in species in close contact with human activities [100].

Levels of PBDEs in aquatic mammals from Cambodia [101], India [52,102], and Brazil [103,104] varied over two orders of magnitude (10–1,000 ng/g) (Figure 3b). Similar variation in PBDE levels was also observed for countries in the developed region such as East Greenland [105], United States [51,106], Russia [55], and Scotland - Isle of May [107]. Levels of PBDEs in aquatic mammals from McMurdo Sound–Antarctica were about one order of magnitude lower at about 1–2 ng/g [51] (Figure 3b). The highest PBDE levels in aquatic mammals were found in the Sao Paulo Coast of Brazil and San Francisco Bay in the United States. The Brazilian coast line, with an estimated population of about 11 million, is one of the most developed areas in the southern Atlantic. It has been impacted by several industries, tourism and agriculture which can be significant sources for PBDEs to the aquatic environment. In contrast, higher PBDE levels in the United States aquatic environment are mainly due to utilization of over 90% of PentaBDE produced globally [15]. PBDE congener patterns in aquatic mammals however, were influenced by the source of the contaminant, transport pathways, diet and species [108].

Although PBDE data for aquatic mammals in and around the African continent are lacking, high levels of the biogenic methoxylated PBDEs (MeO-BDEs) in Indo Pacific bottlenose (mean = 62,000 ng/g) and spinner dolphins (mean = 74,000 ng/g) from the Zanzibar channel in Tanzania were reported [59]. PBDEs were not measured in this study due to lack of available standards.

#### 3. Levels of POPs in human blood in developing regions

Only a handful reports on POPs in human blood from the developing regions are available. Therefore, no summary figures are constructed in this review. More studies were carried out on human milk, which will subsequently be discussed. Since the analytical methods for measuring PCBs and DDTs in humans are very similar, DDTs and PCBs in human blood were often measured and reported together. Therefore, these two groups of POPs in human blood are discussed together in this section.

#### 3.1. PCBs and DDTs

One study on maternal blood from women giving birth in South Africa showed that DDTs were detected in most samples and at higher levels (0.2–14,000 ng/g) than PCBs (0.27–20 ng/g) [109]. The results corroborate an earlier study in Nairobi, Kenya where DDTs (mean = 2,700 ng/g) were detected, but no PCBs, in maternal blood of mothers giving birth by caesarean section [110]. High DDE levels (mean = 380  $\mu$ g/L) were also detected in blood among active pesticide handlers and harvesters in Ghana, Africa [111].

Human exposure to PCBs and DDTs can be influenced by many factors. Levels of DDTs in blood samples were lower in rural population (median DDE = 5 ng/mL) than in urban population (median DDE = 23 ng/mL) in Vietnam [112]. Mean Levels of DDTs in blood from Nagaon district in Northeast India had a positive correlation with age, showing 250  $\mu$ g/L, 720  $\mu$ g/L and 2,000  $\mu$ g/L for age group < 25 year old, 25–50, and > 50, respectively [113]. The same study also found that DDTs were higher in males than in females. DDTs levels in blood were also linked to inhaled vapours of the pesticide [114,115] and diet [116,117]. Two time-trend studies suggested that levels of PCBs and DDTs are decreasing in human from West Africa [118]. Body mass index (BMI) showed no statistical association with levels of DDTs or PCBs in human serum in a Bolivian study [119], but demonstrated a positive association with DDTs in adipose tissue of women in Argentina [114].

### 3.2. PBDEs

Compared to PCBs and DDTs data, there is less data on PBDEs in human blood from the developing regions. However, the limited data indicated a positive association between PBDE levels in general population and urbanization [120,121]. For example, total PBDE blood levels in children living in industrial and urban areas of Mexico were approximately two times higher than those living in rural and municipal areas [122]. Proximity to waste disposal sites and consumption of fish were identified as two important exposure factors for PBDEs in children in Managua, Nicaragua [123]. In Asian developing countries, e-waste recycling plants and municipal solid waste dumping sites were also identified as major PBDE exposure sources for the general population [124]. In countries in the developed region, PBDE congeners in air and dust from e-waste recycling plants have been shown to have similar profiles as those in blood serum from workers in the plants [125-128]. In Africa, an increase in BDE-153 levels in the serum samples of police officers (n = 33) from Guinea Bissau during the study period (1990–2007) were observed [118]. This increase however, was baffling as there were no apparent exposure sources.

#### 4. Comparison of levels of POPs in human milk (2000-present)

Unlike other matrices, human milk has been used as a viable sample type in a number of studies around the world. For the purpose of this review, we focused on studies in both developing and developed regions reported since year 2000 to provide a global perspective for the levels of POPs in human milk. Comparing levels of POPs amongst different studies to evaluate trends of specific environmental contaminants in human milk samples is a challenging task [129]. Data compatibility issue due to use of different analytical methods amongst various studies and inconsistency in the number of congeners being measured within a single group of contaminants such as PCBs or PBDEs are the two major problems. We have chosen only those congeners that were reported most consistently in human milk for comparing the levels among different studies. For PBDEs, these are BDE-47, BDE-99, BDE-100 and BDE-153; for PCBs, the congeners are PCB-138, PCB-153 and PCB-180. For DDTs, p,p'-DDT and p,p'-DDE were chosen. BDE-209 was excluded because it was not always reported in the PBDE studies despite the fact that it is has been reported in both the general population and in occupationally exposed individuals [130].

#### 4.1. DDTs

DDTs in human milk from countries in the developing region were higher than the levels in the developed region as shown in Figure 4a [131-144]. This observation is consistent with findings from an earlier global trend study on DDTs more than a decade ago [145]. Just as it was observed more than a decade ago, the period that restrictions on the use and application of DDT were enforced in different regions correlated with the trends in the levels of DDTs observed in and between these regions [146]. Early restrictions and/or banning of DDT for agriculture in most parts of the developed regions has accounted for the gradual reductions of these compounds in human milk. DDT was banned in most of Western Europe in the late 60s and early 70s [147]. Although Mexico partially restricted the use of DDT in 1972 and banned its use in 1990, levels of DDTs in human milk sampled in 2000 and 2006 in Mexico were among the highest in the world (Figure 4a). Continuous use of DDT has been reported in both the northern and southern Vietnam, which may contribute to the high level of DDTs in human milk in Vietnam [134].

Ghana had the lowest level of DDTs among countries in the developing region. This should be interpreted as an isolated case since DDT is still used in Ghana to fight cotton pests, to control malaria and for artisanal fishing in the Ofin River [111].

It is important to mention that worldwide ban on DDT came into force in 2004. The United Nations convention provides an exemption for continued use of DDT for the purpose of disease-vector control in malaria-dense areas [148]. Highly infested mosquito areas in most parts of the developing regions have witnessed heavy applications of DDT, which almost immediately led to reductions in malaria cases, and consistently high levels of DDTs in human milk [149].

### 4.2. PCBs

Global distribution of PCBs in human milk is illustrated in Figure 4b [134,137,139,141,142,150-163]. The levels in countries in the developed region (especially in Europe) seem to be quite uniform at around the 100 ng/g. On the other hand, data from the developing regions were scarce and showed huge

variations. Available data from Mexico, Vietnam and Tunisia showed that levels of PCBs in these countries were comparable to those in Europe, while levels from Ghana and South Africa were much lower.

Similar PCB levels among countries in the developed region can be attributed to the regulations on the production and application of PCBs in the developed countries. The variation of PCB levels in countries in the developing region however shows a lack or ineffectiveness of such regulations. Lack of proper hazardous waste treatment facilities in the developing region coupled with mass import of obsolete electrical appliances such as transformers from developed countries lead to on-going human exposure to PCB in some of the developing countries. In Tunisia, for example, a 2004 report identified 1079 PCB contaminated transformers, representing 720 tons of liquid PCBs and 2900 tons of contaminated equipment in the country [163]. There are speculations that similar sites are dotted across countries not only in Africa but also in those located in S-SE Asian region. It is therefore necessary to investigate PCB pollution and all its potential sources, and to curb environmental exposures in these regions where no industrial production was reported [158].

## 4.3. PBDEs

PBDEs in human milk are shown in Figure 4c [136,148,152,157-172]. Levels of PBDEs in human milk from India, Vietnam and South Africa were about one to two orders of magnitude lower than those from other countries in the developing regions. Levels from other countries in the developing region were similar to those from European countries. Among the developed countries, the levels in Canada and the United States, depending on the studies, are higher than the levels in other developed countries especially those in Europe. The 2004 ban by the European Union on the production, use and import of penta-BDE and octa-BDE products has been credited for the drop in levels. The comparable levels in European countries, Japan and Australia and the developing regions should be interpreted with caution as the levels in the former are in a decreasing trend due to strict regulations previously alluded to, while those in the latter are on an upward trend. Samples collected in Ghana supports this upward trend in PBDE levels in human milk from 2004 to 2009 [158]. A similar upward trend of PBDE levels was observed in ambient air in another African country of Botswana (Figure 3c).



Figure 3. Levels of PBDEs from countries in the developed and developing regions: (a) levels in ambient air, ng/m<sup>3</sup>; (b) levels in aquatic mammals, ng/g lipid weight. Year of sample collection is indicated in bracket. Central values (\$ = AM;  $\pounds = 1/3$  range; ¥ = GM;  $\pounds = Median$ ; \* = single data point) are presented. The bar indicates minimum and maximum range if reported in the literature. S-SE Asia = South and Southeast Asia.

#### 5. Conclusion and future directions

This review provided a global summary on three groups of POPs, namely DDTs, PCBs and PBDEs in the developing regions as well as some references to the levels in the developed regions. Among these three groups of POPs, PBDE levels in human milk are about an order of magnitude lower than PCBs and DDTs (Figure 4). One noticeable exception is Canada and United States where PBDE levels are comparable to DDTs and PCBs.

Despite of limited monitoring data in the developing regions, lower levels of PCBs and PBDEs, especially in aquatic mammals and in humans, are evident. Higher levels of DDTs in the developing regions, particularly in ambient air and in soil, might be as a result of historical and current use of DDT in agriculture and in disease control in some countries in the regions. Levels reported in the literature are inadequate to be representative of developing regions to establish spatial distribution of POPs in the regions. Data on temporal trends of POPs in the developing regions are particularly lacking. Some data included in this review, especially the levels of DDTs in soil from some countries in the developing region [30,33] are more than a decade old and so might not represent current levels.

PBDEs are relatively new POPs compared to DDTs and PCBs. Higher levels of PBDEs in countries in the developed region, particularly Canada and United States, could be the result of heavy use of PBDEs in the past two decades. Two separate studies measuring PBDEs in ambient air from Botswana [87,88] and in human milk from Ghana [158] showed a possible increase in PBDE levels in these countries. Such upward trend may serve as a warning sign that PBDE levels may on the rise in the developing regions. Therefore, continuing monitoring of PBDEs in the developing regions is crucial to detect possible rising PBDE pollution in these regions.

Most countries in the developed region have taken strong actions against the production and use of DDTs, PCBs and PBDEs as well as control of their emissions. Such effort is still weak in the developing regions due to several reasons of which financial could be considered paramount. For example, due to increased demand of food for domestic consumption and for export, fertilizers and pesticides are widely used in the developing regions [173]. Another concerning area is the export of obsolete electronics and electronic parts as secondhand electronic equipment, or for recycling, from countries in the developed region to some in the developing regions [174]. The latter practice will particularly impact the levels of PBDEs and other flame-retardants in developing regions.

Although this review have provided a global summary of POPs in the developing regions based on data currently available to us, we felt that more monitoring data are required. Several international monitoring activities such as Global Atmosphere Passive Sampling Network for measuring POPs in ambient air [175] and biomonitoring of POPs in human milk by the World Health Organization [176] will continue provide further information on the levels of POPs around the world including the developing regions. International monitoring activities also call for closer collaboration between countries in the developed region and those in the developing regions. Such collaboration is essential for the sharing of technologies and resources as well as generation of compatible data among countries for better interpretation on a global perspective [177].



**Figure 4.** Arithmetic means of POPs in human milk (ng/g lipid weight) sampled post year 2000 from countries in the developed and developing regions: (a) DDTs; (b) PCBs; (c) PBDEs.

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## **Conflict of interest**

All authors declare no conflicts of interest in this paper.

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