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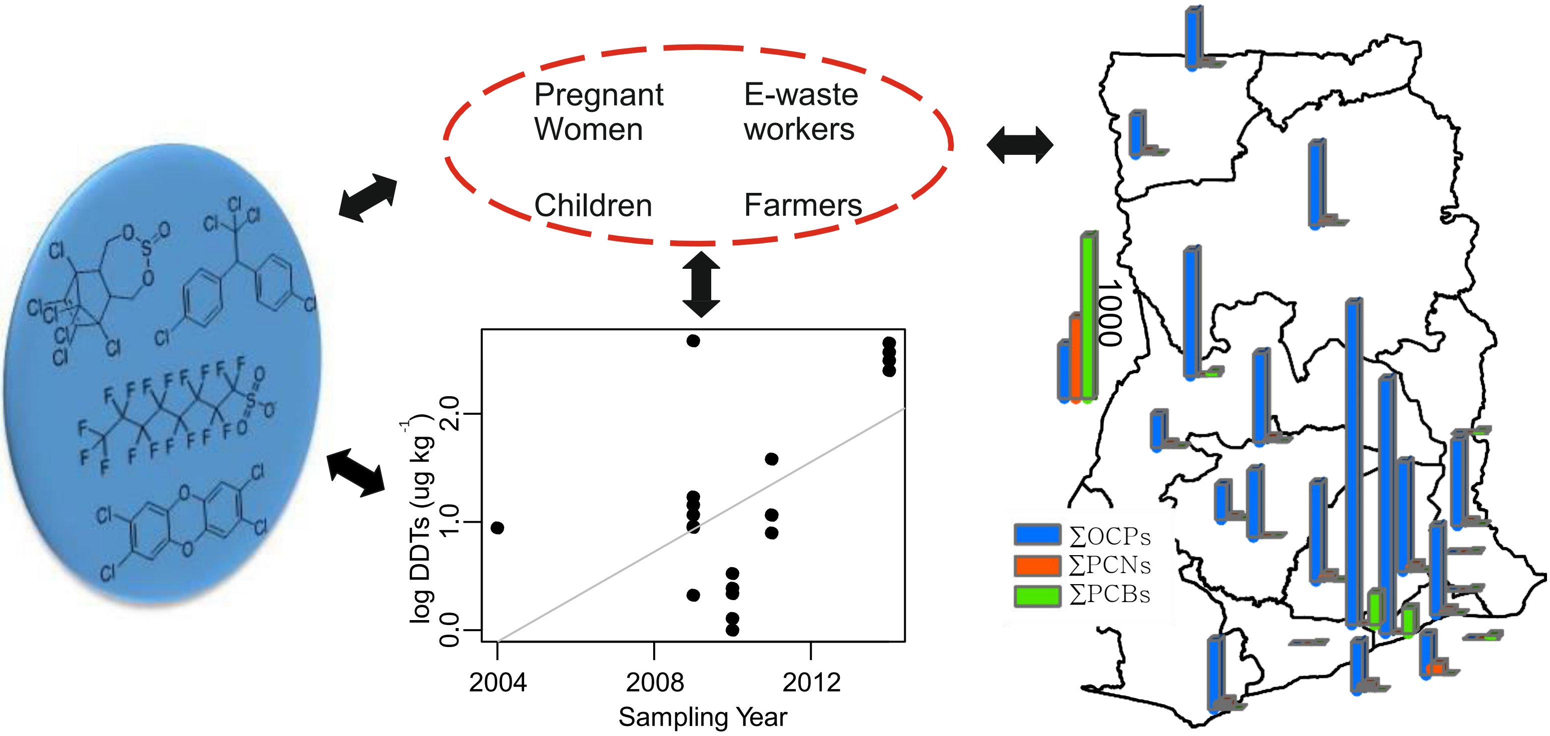
Pregnant Women
E-waste workers
Children
Farmers
log DDTs (ug kg^{-1})
0.0
1.0
2.0
2004
2008
2012
Sampling Year
1000
ΣOCPs
ΣPCNs
ΣPCBs

The State of POPs in Ghana- A Review on Persistent Organic Pollutants: Environmental and Human Exposure.

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Abstract

Ghana is one of the top pesticide users and highest persistent organic pollutant (POP) emitters in sub-saharan Africa. Despite recent increases in published data, there is limited information on how POP concentrations have changed, post ratification of the Stockholm Convention. As a result, this review aims to address these knowledge gaps by collating available data that reported POPs in Ghanaian environmental matrices, identify spatial and temporal trends, and establish potential health risks. It is worth noting that Ghana has not developed its own regulatory standards for POPs, but adapts United States Environmental Protection Agency (USEPA) standards.

Results obtained showed concentrations in excess of USEPA regulatory standards for per- and poly-fluoroalkyl sulphonates (PFASs) and dichlorodiphenyldichloroethane (DDD) in water, polychlorinated and polybrominated dibenzo-p-dioxins and furans (PCDD/Fs and PBDD/Fs) in e-waste soils, and polybrominated diphenyl ethers in aquatic organisms and dairy products. The published studies do not cover major regions nationwide. The inconsistency in methods and analytes measured, along with data scarcity in some regions, makes it challenging to identify temporal trends. However, the data did indicate decreasing concentrations of some legacy POPs in soil/sediment and aquatic organisms, with increasing concentrations of some POPs in water, fish, fruits and vegetables. Studies that performed health risks assessments were limited although the data indicated risks to e-waste workers, some farmers and vulnerable sub-populations. This review identified potential human health risks from POPs in the Ghanaian environment and the need for more consistent and widespread monitoring program.

Capsule: This paper provides a critical review of studies of POPs in Ghana which can be used as a reference for all of Africa, as well as other developing countries, for compliance with the requirements for POPs monitoring in the e-waste, food and environmental sectors to inform the mitigation of health risks.

Persistent Organic Pollutants; Environment; Human Health; Ghana; Africa

1. Introduction

Over several decades, the production of persistent organic pollutants (POPs) has resulted in adverse toxicological effects to human and environmental health. Although POP emissions have been restricted by the Stockholm Convention, exposure continues from a variety of sources: industrial additives in polymers and pesticides, inappropriate waste disposal and long-range transport (Birnbaum, 1994; Gioia et al., 2014; Herrman, 1993; Jones and De Voogt, 1999; Stockholm Convention Secretariat, 2001; Vallack et al., 1998).

Despite adoption and entry into force of the Stockholm Convention in 2001 and 2003, reports still confirm elevated POP concentrations. For instance, in North America, Europe and Asia, POPs in aquatic organisms (Fisk et al., 2001; Hites et al., 2004; Jacobs et al., 2002; Meng et al., 2007), sequestered in soil (Marvin et al., 2002; Zhang et al., 2002), air, dust, particulate matter (Harner et al., 2004; Pozo et al., 2006; Strandberg et al., 2001), wildlife (Mateo et al., 2016), bioaccumulation in serum (Patterson et al., 2009; Sjodin et al., 2008; Thomas et al., 2006) and breastmilk (Kunisue et al., 2004; Schecter et al., 2003; Tanabe and Kunisue, 2007) have been reported. Comparatively, in African countries, although pioneering reports on POPs heavily focused on pesticides (Barakat et al., 2002; Clarke et al., 1997; Darko et al., 2008b; Ntow, 2001; Schulz and Peall, 2001; van Wyk et al., 2001), few studies in Ghana, South Africa and Egypt have documented dietary intake (Adu-Kumi et al., 2010; Asante et al., 2011; Asante et al., 2013), concentrations in serum and breastmilk (Darnerud et al., 2011; Hanssen et al., 2010; Wittsiepe et al., 2015), wildlife, notably birds of prey (Garcia-Heras et al., 2018), atmospheric burdens (Hassan and Shoeib, 2015; Hogarh et al., 2012), water (Essumang et al., 2017), soil, sediment and ash (Caravanos et al., 2011; Fujimori et al., 2016; Nieuwoudt et al., 2009; Tue et al., 2016), and beach pellets (Ryan et al., 2012) for other classes of POPs. Ghana was a signatory to the Stockholm Convention in 2001 and ratified it in 2003 (EPA-Ghana, 2007). Obligations under the Convention for state parties largely resulted in the ban of nine organochlorine pesticides (OCPs) in West Africa (Federal Ministry of Environment Nigeria, 2009; L'Environnement et au Tourisme Guinea, 2012; MINISTERE DE L’ENVIRONNEMENT ET DU CADRE DE VIE, 2007), in addition to PCBs and polychlorinated dibenzo-p-dioxins and furans (PCDD/Fs) (Stockholm Convention Secretariat, 2001). This presents a challenge as Ghana is one of the top pesticide users and POP emitters from major industrial complexes, the agricultural and health sectors (Osibanjo et al., 2002). A map of Ghana is shown in Figure 1, identifying the ten regions. POPs in Ghana are understudied; however, there are potential risks to environmental and human health due to a legacy of widespread pesticide use, along with additional emerging industries such as e-waste processing sites.

An initial baseline assessment of POPs in the first National Implementation Plan (NIP) in 2007 by Ghana’s Environmental Protection Agency (EPA), showed limited information on the production, importation, and usage (EPA-Ghana, 2007). A 2018 revised edition of the NIP highlights inventories of 9,972 sources of PCBs. Approximately 1.4 x 10^8 kg of imported electrical equipment and related wastes between 2009-2014 were estimated to contribute to polybrominated diphenylethers (PBDEs). Previous exposure to OCPs were primarily as a result of unsafe agricultural practices and pest eradication (EPA-Ghana, 2018). PCDD/Fs, mixed halogenated compounds (PXDD/Fs), hexachlorobenzenes (HCBs) and PCB contaminants were identified from a variety of sources including medical waste incineration, vehicular transportation, and open-air burning of electronic waste (EPA-Ghana, 2018).

In recent years, importation of electronics to Ghana has promoted technological growth, although less stringent regulations have contributed to legal and illegal electronic wastes (Brigden et al., 2008). Conflicting views on environmental health risks (Asante et al., 2012; Chan et al., 2007; Fu et al., 2008; Leung et al., 2008) and income generation from e-waste scavenging (Oteng-Ababio, 2012; Oteng-Ababio et al., 2014a), necessitates implementation of regulations to ban informal e-waste recycling and make provisions for sound practices.

1.1. Current Legal Framework for POPs management in Ghana

In addition to the Stockholm Convention, the Basel and Rotterdam Conventions (ratified in Ghana in 2003) integrate environmental justice principles, in recognition of hazards pollutants may pose to humans and the environment (Basel Convention Secretariat, 2003; Rotterdam Convention Secretariat, 2003). Contrary to Article 6 (1) d (i) and (ii) of the Stockholm Convention, appropriate measures for handling and disposal of POPs e-wastes are lacking in Ghana. Of relevance are the Environmental Protection Agency Act, 1994 (Act 490) (EPA-Ghana, 1994), and the Hazardous and Electronic Waste Control and Management Act, 2016 (Act 917), for regulation of pesticides and wastes (EPA-Ghana, 2016). Act 917 identifies the need for appropriate recycling facilities for the proper disposal and management of POPs e-waste and hazardous wastes.

1.2. Methodology and Aims

Recent and historic sources, and types of POPs, make the Ghanaian environment an important study area; however, a systematic review of POPs is yet to be completed. This study focuses on previously published data on the Stockholm Convention POPs. As there are several individual Stockholm POPs, similar analytes and congeners were grouped and compared to assess which classes of POPs need further focus. This study reviews POP data for environmental matrices (Section 2), food (Section 3) and humans (Section 4). Data on sample collection, preparation and analytical methods are presented in supplementary information S2, and Table S1. POP concentrations in various environmental matrices were compared with internationally accepted tolerance levels to estimate potential health risks. The data gathered is considered in sections 5 and 6, and Table S6, presenting a critical evaluation and identify considerations for future research prioritization.

Based on the criteria for a systematic review (Liberati et al., 2009), a literature search of peer-review articles published from 2001 to present was conducted using Web of Science and Scifinder databases. The following search terms were used: "persistent organic pollutants"-POPs, "polychlorinated biphenyls"-PCBs, "polybrominated diphenylethers"-PBDEs, "organochlorine pesticides"-OCPs, "polychlorinated dibenzo-p-dioxins and furans"-PCDD/Fs, "polychlorinated napthalenes"-PCNs, "perfluoroalkyl sulphonates"-PFASs and "Ghana". A total of 151 scientific papers were identified (88 from Web of Science, and an additional 63 with Scifinder). Duplicate manuscripts were removed and the remaining papers screened for suitability based on reporting of the following criteria: Stockholm POP congeners, sampling location, type of sample, extraction and detection method, and concentrations. This resulted in a total of 56 papers used to compile this review. Further, 8 papers on social impacts of POPs, the 2007 and 2018 revised NIP reports were reviewed. For temporal trend analysis, the sum of DDTs: [o,p′-dichlorodiphenyltrichloroethane (DDT), p,p′-DDT, o,p′- dichlorodiphenyldichloroethylene (DDE), p,p′-DDE, and o,p′-dichlorodiphenyldichloroethane (DDD)], sum of HCHs (hexachlorocyclohexanes): [α-HCH, β-HCH, γ-HCH, and δ-HCH], and sum of Endosulfans: endosulfan I, II and endosulfan sulfates] were plotted against sampling year. The results of temporal and spatial trends are summarized in supplementary information- S3 Temporal and Spatial Evaluations, and in the conclusion section 5.

Several challenges need to be considered when comparing historical datasets. We have attempted to address these in the supplementary information S1, but acknowledge the resultant inevitable degree of uncertainty.

2. POP Concentrations in the Ghanaian Environment

2.1. Air

POP concentrations in ambient air vary geographically and spatially, and depend on inputs from emission sources. As with other environmental media, POPs partition to particulate matter after pesticide spray application, from combustion processes, and volatilization (Breivik et al., 2002; Jones, 1994). Table S2 and Figure S1 summarize POPs in air in urban and rural areas in Ghana. Analytical methodologies are discussed in Table S1. A review on baseline studies on Ghanaian regions on PCBs, OCPs, PCDD/Fs and PCNs is described below.

2.1.1. PCBs

PCB concentrations in rural and urban areas ranged from below the limit of detection (LOD) to 74 pg m^{-3} (Bogdal et al., 2013; Gioia et al., 2011; Pozo et al., 2009). The highest total concentrations for 7 PCB congeners ranged between 38-74 pg m^{-3} for rural and urban areas (Bogdal et al., 2013). Total concentrations at Wenchi-rural area (Eastern region) for 48 PCB congeners, for different sampling periods, were 35 pg m^{-3} and 68 pg m^{-3} (Pozo et al., 2009). A similar contribution from other rural areas for 29 PCB congeners, was 33 pg m^{-3} (Gioia et al., 2011). For urban areas- Greater Accra region, the lowest concentration range for 48 PCB congeners was in Kwabenya (range: 8.2-12.6 ng sample^{-1}); the highest measurement was at East Legon (range: 6.9-20.3 ng sample^{-1}) (Klanova et al., 2009). Hazard risk assessments for inhalation of ambient PCB air were not reported in studies reviewed, possibly because of their minimal contribution (1-2%) to total exposure from direct inhalation as opposed to dietary intake (WHO, 2000).

The maximum reported exposure concentrations for urban areas (74 pg m^{-3}) was below the accepted World Health Organization (WHO) PCB concentration of 3 ng m^{-3} and 0.003 ng m^{-3} for urban and non-industrialized areas, respectively (WHO, 2000). Although rural exposure concentrations exceeded 0.003 ng m^{-3} (WHO, 2000), associations of ambient PCBs with health risks are low. USEPA suggests possible risk to result from continuous inhalation of concentrations that exceed 1.0 µg m^{-3} (USEPA, 1989). The current data reflects background levels with evidence of minimal primary emissions from agricultural wastes, vehicular transportation, electronic wastes dumping and indiscriminate burning of wastes.

2.1.2. OCPs

Total OCP concentrations in rural and urban areas ranged from below LOD to 5,296 pg m^{-3} (Adu-Kumi et al., 2012; Hogarh et al., 2014; Klanova et al., 2009; Pozo et al., 2009). The mid to southern parts of Ghana were dominated by HCHs, DDTs, and endosulfans; chlordanes and heptachlors were detected in the northern parts. The mean OCP concentrations reported in rural-Wenchi (Eastern region), Lake Bosomtwe (Ashanti region), suburban-Accra, and other sites ranged between 19.3-3,700 pg m^{-3} (Adu-Kumi et al., 2012; Hogarh et al., 2014; Klanova et al., 2009; Pozo et al., 2009).

The potential for health risks are low, as concentrations reported were below USEPA estimated carcinogenic assessment inhalation risks, ranging from 9.7 x 10^{-5}-0.0013 µg m^{-3} for selected OCPs (USEPA, 1989). Since their ban in 1985 (except for lindane- banned in 2001 and endosulfan- 2009 in Ghana), concentrations indicate recent pesticide usage in agricultural sectors (EPA-Ghana, 2007). Spatial and temporal trends are displayed in Figures S1 and S2; results are summarized in supplementary S3 and conclusion sections.

2.1.3. PCDD/Fs

One study in literature focused on PCDD/Fs in air. Concentrations of PCDD/Fs in urban-Accra ranged between 370-2,200 pg sample^{-1}, with hazard risk assessment of 10-100 pg International (I)-Toxic Equivalency (TEQ) sample^{-1} (0.2 pg I-TEQ m^{-3}) (Klanova et al., 2009). Industrial and statistical emission estimates from 2002 baseline inventory to 2015 indicate an increase from 665 to 1485 g TEQ PCDD/Fs in Ghana (EPA-Ghana, 2018). Assuming a sampling volume between 300-600 m^{3} (Klanova et al., 2009), concentrations exceed USEPA urban emission estimates of 0.1 pg m^{-3} (USEPA, 1989). At these concentrations, a low to medium health risk of skin and eye irritation from PCDD/Fs inhalation can occur (USEPA, 1989). The current data reflects background concentrations with potential emissions from agricultural wastes, vehicular transportation, electronic wastes dumping and burning of wastes (EPA-Ghana, 2018).

2.1.4. PCNs

One study has been completed on PCN emissions in Ghana. Total concentrations of 63 PCN congeners were low and high in the middle and southern belts: ~30 and 100 pg m^{-3}. TEQ calculations of 17 PCN congeners resulted in concentrations ranging between 0.5-6 fg TEQ m^{-3} for dioxin-like (dl) toxicity (Hogarh et al., 2012). The potential for eye and skin irritations, and liver tissue lesions to result from prolonged inhalation of ambient PCN exposure are low, as emissions were lesser than WHO estimates (tri- to hexa-, and octachloronapthalene range: 0.1-5 mg m^{-3}) for occupational exposure (WHO, 2001). PCNs are yet to be banned in Ghana; the high emissions may be attributed to point sources including industrial production sites: smelting and used car incineration. Additional sources could be from volatilization and wind trajectories from illegal toxic waste dumped by Trafigura in 2006 on the south coast of Cote d'Ivoire (White, 2008).

2.2. Water

POP exchange between the atmosphere, aquatic ecosystems and terrestrial surfaces influence POP loadings in aquatic media and sediments (Jozef M. Pacyna, 2000). In Ghana, lake, river and stream contamination can stem from agricultural run-off during rainy seasons, and household use of pesticides. A review of data on OCPs, PFASs and PCBs in water is described below. These studies highlight important findings which indicate potential pesticide contamination in 3 of 5 drinking water sources in Ghana- River Densu, White Volta (Volta Lake), and Pra River (Lake Bosomtwe).

Table S1 includes an analytical summary of POP residues in water, in Ghana. Temporal trends in water are shown in Figure S3; results are summarized in supplementary S3 and conclusion sections.

2.2.1. PCBs

Mean PCBs in Lake Bosomtwe ranged from 1,090-7,190 ng L^{-1}, with PCB-52 as the dominant congener (Afful et al., 2013b). PCB concentrations exceeded USEPA maximum allowable limit of 500 ng L^{-1} in drinking water (USEPA, 2009). Majority of local communities in Ashanti region depend on water and fish from Lake Bosomtwi; extensive exposure to higher levels for extended time periods could potentially result in skin disorders and immune deficiencies (USEPA, 2009).

2.2.2. OCPs

Residues of OCPs in water were greater in rural than urban areas. Mean OCPs in rural water- streams around agricultural irrigation sites in Tono (Upper East Region) and Akumadan (Ashanti Region), standing pipe water source, drinking groundwater from dugwells, the Volta Lake, and Lake Bosomtwe ranged from below the LOD-6,350 ng L^{-1} (Afful et al., 2013b; Akoto et al., 2016; Darko et al., 2008b; Fosu-Mensah et al., 2016; Kuranchie-Mensah et al., 2012; Ntow, 2001; Ntow, 2005; Ntow et al., 2008a). Mean OCPs in urban water-River Densu in Nsawam and Weija (Greater Accra Region), ranged from below the LOD-180 ng L^{-1} (Kuranchie-Mensah et al., 2012). OCP residues at Tono irrigation site were below the LOD (Akoto et al., 2016). In both rural and urban waters, endosulfans were present. Lake Bosomtwe was the most contaminated, with high concentrations of endosulfan sulfate (5,630 ng L^{-1}) and p,p'-DDD (6,350 ng L^{-1}) (Afful et al., 2013b). In the above studies, OCPs were below WHO MRLs for surface (WHO, 2017), and groundwater (WHO, 2006), except for p,p'- DDD (6,350 ng L^{-1} Lake Bosomtwe) (Afful et al., 2013b), which exceeded the MRL of 1,000 ng L^{-1} (WHO, 2017). The potential for carcinogenic risks to result from oral exposure to OCPs below the MRLs are low; risk levels estimated by USEPA (for DDD, DDT, DDE, aldrin, HCH) that can induce carcinogenic risks (for 1 in 10,000 persons) range between 0.6-10 µg L^{-1} (USEPA, 1989). Recent widespread use of endosulfan within the agricultural sector, and potential illegal use of DDT, lindane, amongst other banned pesticides in rural areas, are the suspected sources responsible for contaminating water resources.

2.2.3. PFASs

Of the 15 perfluoroalkyl acids (PFAAs) congeners, high concentrations of perfluorooctanoic acids (PFOAs) and perfluorooctane sulfonates (PFOSs) were detected in two river basins- River Pra and Kakum, and tap water from rural areas. The mean PFOAs and PFOSs concentrations ranged between 113-205 ng L^{-1} for river basins, and 103-107 ng L^{-1} for tap water (Essumang et al., 2017). The sum of concentrations [PFOSs] + [PFOAs] at each site, exceeded USEPA health advisory levels of 0.07 ng L^{-1} in drinking water (USEPA, 2016). Potential health risks of thyroid disease, kidney and testicular cancer could result from prolonged exposure to PFOSs-contaminated water (Essumang et al., 2017). The data reported suggests that treatment of Pra and Kakum river basins for tap water is not efficient at removing PFAA contaminants (Essumang et al., 2017). PFASs are yet to be banned in Ghana; the limited data gathered as part of this review indicates there may be significant PFAS contamination in Ghanaian drinking water.

2.3. Soil and Sediment

Pesticides introduced into soils are taken up by plants, degrade or transported to groundwater and accumulate in sediments (Ilyina, 2007). POPs are hydrophobic in nature, strongly bind to soil and sediments rich in organic carbon matter, and can be slow to degradation processes (Van Metre and Mahler, 2005). Soil and sediment act as reservoirs or sinks (Moeckel et al., 2008; Van Metre and Mahler, 2005); thus, long-term deposition make it possible to detect accumulated POPs. A discussion on studies of PCBs, OCPs, PBDEs and dioxin-like compounds (DLCs) in soil and sediment is given below. These studies indicate contamination of soil and sediment was as a result of recycling, dismantling and combustion of e-wastes, leakage of oils from transformer storage, and agricultural pesticide usage. Residents and workers in close proximity to e-waste soils, and soils surrounding transformers, can be exposed to pollutants from inhalation, dermal contact and ingestion of deposits on food.

Table S1 includes an analytical summary of POPs in soil and sediment. Table S4 and S5 summarizes POP data in sediments. Temporal and spatial trends are shown in Figures S4 and S5; results of trends are summarized in supplementary S3 and conclusion sections.

2.3.1. Soil and ash from e-waste sites

2.3.1.1. DLCs

Concentrations of dioxin-like PCBs (dlPCBs), PCDD/Fs and PBDD/Fs at Agbogbloshie e-waste site in Accra, were among the highest measured in Ghanaian soils. Soils from open-burning of electronic wastes and metal sites were contaminated with PBDFs: 83-3,800 µg kg^{-1} dry weight (dw), followed by PCDFs: 11-390 µg kg^{-1} dw, PCDDs: 6.6-120 µg kg^{-1} dw, PBDDs: 0.12-4 µg kg^{-1} dw and dlPCBs: 3.4-82 µg kg^{-1} dw (Tue et al., 2016). Soils from open-burning sites were more contaminated than non-burning and non e-waste sites. The formation of dlPCBs was mainly attributed to catalytic abilities of Cu, Zn and Pb to release active chlorine and bromine species from e-waste combustion (Fujimori et al., 2016). Median WHO-TEQ for DLCs were 7.1 µg kg^{-1} TEQ dw- open burning, 0.12 µg kg^{-1} TEQ dw- non-burning and 0.00016 µg kg^{-1} TEQ dw for non e-waste sites. Median TEQ values for e-waste soils exceeded the Canadian Soil Quality Guidelines (SQG) for PCDD/Fs (0.004 µg kg^{-1} TEQ dw), indicating a potential risk to human health (Canadian Environmental Protection Act, 2002).

2.3.1.2. PBDEs

Concentrations of PBDEs in Agbogbloshie e-waste soils ranged between 16-100 µg kg^{-1} dw. A variation in distribution of PBDE congeners was attributed to non-specific sources from e-waste activities. The dominant congener was PBDE 28, followed by PBDE 209 and PBDE 47 (Akortia et al., 2017). In contrast to the expected theory of lower brominated congeners partitioning to air particulates and higher brominated depositing on soil, lower brominated congeners was attributed to possibilities of atmospheric transport and deposition, and de-bromination of higher congeners during dismantling and open-air burning processes (Akortia et al., 2017; Oteng-Ababio et al., 2014b).

POPs in non-agricultural soils become a concern when there is a significant pathway for exposure and receptors. The Agbogbloshie e-waste area is centred in a vegetable and food market place surrounded by children of vendors, e-waste workers and the public. Despite the large potential for exposure, no risk assessment of combined multiple exposure from inhalation and food consumption has been completed. Given the elevated concentrations recorded at this and other global e-waste sites, there may be a significant risk to human health.

2.3.2. PCBs in oil, and soil around transformer oil storage sites

Using neutron activation analysis, higher total chlorine-^{38}Cl contents of PCBs were measured by irradiation of 94 transformer oils collected from schools, hospitals, and water treatment plants (71,340-266,920 µg kg^{-1} wet weight (ww)) (Buah-Kwofie et al., 2011), in comparison to soil extracts from 4 transformer oil storage sites (7,690-51,920 µg kg^{-1} dw) (John et al., 2014). The concentrations indicate major contamination of soils around transformer storage sites, which present a local environmental and human health risk.

2.3.3. Agricultural soil

Studies of POPs in agricultural soils, in Ghana, were scarce and mainly focused on OCPs in surface soil (Bentum et al., 2006; Fosu-Mensah et al., 2016; Ntow et al., 2007). Variable depths of cored soils from cocoa farms and a tomato field in rural areas were measured for OCPs. Mean concentrations of lindane in cocoa farm soils ranged between LOD-50 µg kg^{-1} dw in Brong Ahafo region (Fosu-Mensah et al., 2016), and between 2,100-15,500 µg kg^{-1} dw for Central region (Bentum et al., 2006). Lower mean concentrations of p,p'-DDT, β-HCH and dieldrin residues in cocoa soils ranged between 5-50 µg kg^{-1} dw

(Fosu-Mensah et al., 2016). Endosulfan dissipation in tomato soils showed α-endosulfan (mean: 230-2300 µg kg^{-1} dw) and endosulfan sulphate (mean: 40-650 µg kg^{-1} dw) were retained on top soil; β-endosulfan leached to lower depth (mean: 110-650 µg kg^{-1} dw) (Ntow et al., 2007). Concentrations of lindane exceeded the Canadian Environmental Quality Guideline (CEQG) of 10 µg kg^{-1} in agricultural soils (Canadian Council of Ministers of the Environment, 1991); other OCPs monitored in soil were within the CEQG limits.

2.3.4. Sediments

POPs in sediments (from rivers, lakes, streams and coastal areas) are the most studied matrix in Ghana. Although cored sediments reflect historical records of POP pollution, surface sediments have been the focus, limiting the ability to understand sediment temporal trends. A discussion on OCPs and PCBs in surface sediments from coastal marine, lakes, streams, river basins and irrigation dams is given below.

2.3.4.1. PCBs

Mean PCB concentrations in sediment for 11 coastal areas (15.5-47.89 µg kg^{-1} dw) (Dodoo et al., 2012), was higher than for river sediments: 8 sites (0.57-32.2 µg kg^{-1}dw) (Hosoda et al., 2014), and lake sediments: 11 sites (1.09-19.17 µg kg^{-1} dw) (Afful et al., 2013b). The prevalence of higher concentrations of lower PCB congeners (PCB- 28 and 52) (Afful et al., 2013b; Dodoo et al., 2012; Hosoda et al., 2014), supports the theory of sediment historic contamination and subsequent degradation. To evaluate probable toxic effect levels of PCBs on aquatic organisms, a comparison of sediment mean concentrations with the CSQG, showed PCB concentrations fell within the accepted value of 21.5 µg kg^{-1} dw (Afful et al., 2013a, 2013b; Dodoo et al., 2012; Hosoda et al., 2014). A low health risk from human exposure to coastal sediments was identified from hazard index (HI) assessment of < 1 (Hosoda et al., 2014).

2.3.4.2. OCPs

DDTs and HCHs were frequently detected in surface sediments covering coastal Tema harbour areas, Weija dam and Nsawam (Densu river basin) in Greater Accra, Eastern region, Lake Bosomtwe and 4 streams in Ashanti region, Volta lake (6 sites), and Tono irrigation reservoir in the Northern region. The sum of DDTs were highest for irrigation sediment (47-70 µg kg^{-1} dw) (Akoto et al., 2016), followed by Volta Lake (61.30 µg kg^{-1} dw) (Ntow, 2005), coastal sediments (6.0-12.8 µg kg^{-1} dw) (Botwe et al., 2017), lake sediments (LOD-12.75 µg kg^{-1} dw) (Afful et al., 2013b; Darko et al., 2008b), river basin sediments (3.289 µg kg^{-1} dw) (Kuranchie-Mensah et al., 2012), and streambed sediments (0.46 µg kg^{-1} dw) (Ntow, 2001). Mean HCHs (0.75-13.6 µg kg^{-1} dw) were much lower. Contributions of aldrin and dieldrin were very low for sediment types (range: LOD- 0.95 µg kg^{-1} dw), except for irrigation sediment (aldrin: 90 µg kg^{-1} dw) (Akoto et al., 2016). Similarly, low mean concentrations of endosulfan sulphate were detected in all sediments (0.18-1.61 µg kg^{-1} dw), except for Lake Bosomtwe (37.68 µg kg^{-1} dw) (Afful et al., 2013b).

Predictors of past or recent DDT usage are based on aerobic and anaerobic degradation of DDT to DDE and DDD. Provided the ratio of DDT to its metabolites is <1, past usage is predicted. Calculated ratios observed were <1 for river, lake and coastal sediments (Afful et al., 2013b; Botwe et al., 2017; Darko et al., 2008b; Kuranchie-Mensah et al., 2012; Ntow, 2005), indicating a decline in DDT usage with an increase of its metabolites over the years.

Ecotoxicological risks of OCPs to aquatic organisms were evaluated by comparing the sum of sediment mean concentrations to SQG. The additive effect of OCPs in river, lake, coastal and irrigation sediments were below the lowest effect concentration (LEL) values of the SQG (Table S3), an indication of low to medium ecotoxicological risk to aquatic organisms. Lindane was identified as the major source of HCH contamination in sediments; based on the predominance of γ-HCH which provides a ratio <1 for $\frac{\alpha\, HCH}{\gamma HCH}$ (Willett et al., 1998)

2.3.5. Pellets

Plastic resin pellets are waste organic micropollutants released from plastic industries; they pose a risk because they adsorb hydrophobic contaminants from aquatic media, and are ingested in large quantities by aquatic organisms and sea birds. Two reports monitored beach pellets as carriers of PCB contaminants in coastal rural and urban areas. The sum of mean PCBs from 17 beaches ranged from 1-98.31 µg kg^{-1} dw (Agbo and Abaye, 2016; Hosoda et al., 2014). PCB concentrations in Accra: 39-69 µg kg^{-1} dw (Ntow et al., 2011), and Tema-Sakumono beaches: 29-46 µg kg^{-1} dw (Ntow et al., 2011), 47.47 µg kg^{-1} dw (Bempah et al., 2012), were higher than rural areas: 1-15 µg kg^{-1} dw (Ntow et al., 2011). Coastal pellets in Accra and Tema were dominated by PCB-110, 138 and 180 (penta, hexa and hepta-PCBs), with rural sites containing a lower proportion of higher chlorinated congeners. PCB contamination of beach pellets was attributed to local inputs from e-waste dismantling and dumping sites.

3. POP concentrations in Food

POPs, once introduced into air, deposit on vegetation, soil and sediments, and bioaccumulate in aquatic fish and farm animals from ingestion of contaminated feed, sediment and plants. Marine and freshwater organisms are used as bioindicators because they accumulate POPs in higher concentrations than their aquatic environment (Gunther et al., 1999). For the majority of population that are not occupationally exposed to POPs, the main route of exposure (> 90% of POP intake) arises from dietary intake of animal products, fish, and seafood (Liem et al., 2000). Fruits and vegetables treated with pesticides, are another source of exposure (Liem et al., 2000).

Table S5 and Figure S6 summarize POP data in food. Methods of extraction, clean-up and analytical detection are summarized in Table S1. A discussion on POP concentrations, in edible fish, seafood, dairy products, beef, game meat, vegetables, fruits and cereals, is given below. Results from these studies indicate that intake of food of animal origin is the major contributor to OCPs and PCBs. On the other hand, relatively small PBDEs and hexabromocyclododecanes (HBCDs) contributions were obtained from fish (Asante et al., 2010; Asante et al., 2013), whilst vegetables, fruits and cereals contributed substantial amounts of OCPs. Data on Ghanaian dietary intake of PCDD/Fs and dlPCBs is scant.

3.1. Aquatic organisms

Freshwater fish is an important part of Ghanaian diet; it is a source of animal protein monitored for bioaccumulated POPs. Biomonitoring activities focused on fish types mostly consumed- tilapia and catfish. Muscle tissue is the commonly consumed fish part frequently analysed for contaminants. Investigations of POPs in molluscs in Ghana are limited, with three papers determining concentrations in oysters, mussels and cockles.

3.1.1. PCBs

Mean concentrations of PCBs in tilapia (*Tilapia zilli* and *Oreachromis niloticus*) and catfish (*Clarias gariepinus* and *Chrysichthys nigrodigitatus*) ranged from LOD-62 µg kg^{-1} lipid weight (lw) (Asante et al., 2013; Kuranchie-Mensah et al., 2011). Mean PCB concentration, reported for tilapia from inland and coastal areas, was 62 µg kg^{-1} lw (Asante et al., 2013); much lower mean concentrations in Lake Volta for tilapia and catfish ranged between 0.9-12.37 µg kg^{-1} ww (Kuranchie-Mensah et al., 2011). Dominant congeners were PCBs- 153, 138 and 180 (Asante et al., 2013), although lower congeners PCBs- 28, 52, 101 and 99, contributed significant amounts (Asante et al., 2013; Kuranchie-Mensah et al., 2011). Potential risks of dietary exposure to tilapia and catfish from Lakes Volta and Weija, and Benya and Keta lagoons were assessed to be low from hazard risk calculations (<1) (Asante et al., 2013; Kuranchie-Mensah et al., 2011), although authors proposed a more detailed assessment using HI and TEQ-WHO (Asante et al., 2013). Mean PCB concentrations were below the United States Food and Drug Administration action level (2000 µg kg^{-1} ww) recommended for fish, suggesting a low health risk (USEPA, 2000).

PCB concentration in bivalves: cockles (*Anadara senilis*), oysters (*Crassostrea tulipa*) and mussels (*Perna perna*) along coastal rural areas (Lake Benya, Ningo, Sakumono) were higher than for fish. Median concentrations for dry and wet seasons ranged between 1,200-3,500 µg kg^{-1} lw, and 1,500-2,100 µg kg^{-1} lw (Otchere, 2005). Lower PCB concentrations in mussels and oysters were detected in Narkwa, Ada and Anyanui; range: 3-11 µg kg^{-1} ww (Dodoo et al., 2013). Seasonal variation of PCBs in mussels was attributed to different source inputs (terrestrial and marine) (Otchere, 2005). Dietary exposure to PCB-contaminated bivalves are potentially high since median concentrations exceeded FDA action levels of 2000 µg kg^{-1} lw for shellfish (USEPA, 2000). Results from calculated risks using PCB 118 (21-112.0 pg WHO-TEQ kg^{-1}) (Dodoo et al., 2013), exceeded the recommended Tolerable Daily Intake of 2 pg WHO-TEQ kg^{-1} with potential risks of low birth weight and neurobehavioural effects in children of exposed pregnant women. The calculated risk contradicts the HI (<1), which indicated low risks of exposure to consumption of oysters and mussels. Typically, TEQ is based on an additive result of 12 dlPCBs; however, the main driver of TEQ is the most toxic: PCB 126. Therefore, an assessment including 12 dlPCBs, rather than the use of PCB-118, would accurately assess risks. For both mussels and oysters, tri and hepta-CBs were dominant congeners (Dodoo et al., 2013). The results indicate there may be a significant risk from consumption of aquatic organisms. However, studies involving determination of WHO-TEQ for PCBs and other DLCs, and a detailed quantitative risk assessment is required to establish risk magnitude.

3.1.2. OCPs

DDTs were detected in fish obtained from lakes and reservoirs. The sum of mean concentrations of DDTs in tilapia, were highest for Tono reservoir-Upper East Region (*Sarotherodon galilaeus*: 250 µg kg^{-1} ww) (Akoto et al., 2016); and Lakes Volta, Bosomtwe and Weija for *Tilapia zilli* and catfish (*Clarias gariepinus*): 253.4 µg kg^{-1} lw (Adu-Kumi et al., 2010). Other studies detected lower mean concentrations of DDTs in tilapia in Lake

Bosomtwe- 8.88 µg kg^{-1} ww (Darko et al., 2008b), Lake Volta- 7.96 µg kg^{-1} ww (Kuranchie-Mensah et al., 2011), 3.81 µg kg^{-1} ww (Gbeddy et al., 2012), and Weija Lake- 0.41 µg kg^{-1} ww.[144] The sum of mean DDTs concentration in catfish in Lake Bosomtwe, Volta and Weija was 2206 µg kg^{-1} lw (Adu-Kumi et al., 2010); DDTs contamination in Tono reservoir was 336 µg kg^{-1} ww in *Schilbe intermedius* (Akoto et al., 2016). HCHs bioaccumulation was high in Kpando Torkor Lake (sum of mean concentration for *Tilapia zilli*: 41.6 µg kg^{-1} ww) (Gbeddy et al., 2012). Mean HCHs and endosulfan concentrations in other fish species ranged from LOD-20.13 µg kg^{-1} ww (Darko et al., 2008b; Gbeddy et al., 2012; Kuranchie-Mensah et al., 2011), and from LOD-4.48 µg kg^{-1} ww respectively (Darko et al., 2008b; Gbeddy et al., 2012; Kuranchie-Mensah et al., 2013). Other OCPs in fish included aldrin, dieldrin, heptachlor and chlordane. Mean of chlordanes (trans-, cis-, oxy- chlordane) ranged from LOD-26.06 µg kg^{-1} ww (Adu-Kumi et al., 2010; Gbeddy et al., 2012; Kuranchie-Mensah et al., 2011).

Mean OCP concentrations detected were below Food and Drugs Administration (FDA) action levels for DDTs (5000 µg kg^{-1} ww), chlordanes, aldrin, dieldrin, and heptachlor (300 µg kg^{-1} ww) for fish and shellfish from freshwater and marine sources (Food and Drug Administration, 1995); an indication of low risk from OCP-contamination in Ghanaian fish. Hazard risk calculation for consumption of OCP-contaminated fish varied in studies. Hazard indices (HI) for fish consumption from Kpando Torkor lake indicated low risks (<1) for HCH, DDT and γ-chlordane (Gbeddy et al., 2012); an HI of >1 was calculated for aldrin-contaminated fish from Tono reservoir (Akoto et al., 2016). Other studies predicted potential risks via consumption of OCP-contaminated fish, although HI were not calculated (Adu-Kumi et al., 2010; Darko et al., 2008b; Kuranchie-Mensah et al., 2013).

3.1.3. PCDD/Fs and dlPCBs

The mean dlPCB concentration (1200 pg g^{-1} lw) in catfish and tilapia exceeded PCDD/Fs (23 pg g^{-1} lw) in Lakes Bosomtwe, Volta and Weija (Adu-Kumi et al., 2010). Estimated WHO-TEQs for dlPCBs and PCDD/Fs was 0.3 pg WHO-TEQ g^{-1} lw (Adu-Kumi et al., 2010). Fish from the three lakes contained relatively low PCDD/Fs-dlPCBs, as calculated WHO-TEQ value was below the permissible European Union (EU) Regulations limit for fish: 8.0 pg WHO-TEQ g^{-1} ww (European Commission, 2006a), posing low health risks.

3.1.4. PBDEs and HBCDs

Mean concentrations of PBDEs and HBCDs in tilapia from Lakes Weija and Volta, and Benya and Keta lagoons were low. Mean PBDEs ranged from 0.89-19 µg kg^{-1} lw; HBCDs ranged from 0.04-2.2 µg kg^{-1} lw. The least and most contaminated lagoons were Keta and Benya. Dominant congeners- PBDE 47 and 209, were attributed to usage of penta and deca-BDEs. Possibilities of degradation of PBDE-99 into PBDE-47, run-off from contaminated areas into lakes, and de-bromination of hepta to hexa-BDEs contributed to their accumulation (Asante et al., 2013). Possible contamination sources of lakes and lagoons were credited to waste discharge from textile industries as well as improper wastewater treatment (Asante et al., 2013). Mean PBDEs- 0.16 µg kg^{-1} ww for 15 PBDE-congeners, inclusive of 6 PBDEs (Asante et al., 2013), exceeded the maximum allowable concentrations for biota- Directive 2013/39/EU: 0.0085 µg kg^{-1} ww for PBDE- 28, 47, 99, 100, 153 and 154 (European Commission, 2013). Low to medium risks of estrogenic activity from dietary exposure to PBDE-contaminated fish are expected, although calculated HI were below the critical value (<1). Low risks from dietary exposure to HBCDs in fish is predicted as the mean HBCD concentration (0.02 µg kg^{-1} ww) (Asante et al., 2013) was below EU Directive-

2013/39/EU levels for biota (167 µg kg^{-1} ww) (European Commission, 2013). Spatial and temporal trends are displayed in Figures S6 and Figure 2; results of trends are summarized in supplementary S3 and conclusion sections.

3.2. Dairy products

3.2.1. OCPs

Dietary exposure to six OCPs was assessed in dairy products. Mean DDTs concentration in cheese ranged between LOD-298 µg kg^{-1} lw. Lower mean concentrations were detected in milk and yoghurt: 4.7-10 µg kg^{-1} lw. The sum of mean OCP concentrations were below WHO MRLs (Darko and Acquaah, 2008a). Mean OCP concentrations in cheese were below the extraneous WHO MRLs for lindane (100 µg kg^{-1}), aldrin (150 µg kg^{-1}), dieldrin (150 µg kg^{-1}), endosulfan (100 µg kg^{-1}) and DDT (500 µg kg^{-1}), an indication of low risk from dairy dietary exposure (Darko and Acquaah, 2008a).

3.2.2. PCBs

A comparison of PCBs in raw cow milk in urban-Accra and rural-Asutuare (Eastern region) showed the sum of mean PCBs (27 µg kg^{-1} lw) in urban areas to be twice that for rural (14 µg kg^{-1} lw). A variation in PCB accumulation in cow milk were mainly attributed to feeding habits (Asante et al., 2010). The mean concentrations for 15 PCB congeners were below the maximum EU limits of 40 µg kg^{-1} lw (European Commission, 2011). Low health risk from milk consumption is expected; however, no studies were completed to ascertain the TEQ.

3.2.3. PBDEs

In urban cow milk, concentrations ranged between 0.47-11 µg kg^{-1} lw (mean: 2.3 µg kg^{-1} lw). Lower concentrations were in rural milk (0.05-2.8 µg kg^{-1} lw, mean: 1.0 µg kg^{-1} lw). Dominant congeners observed were PBDE-47 and 99. HBCD concentrations were below the LOD (Asante et al., 2010). The mean concentrations of PBDE congeners exceeded allowable concentrations set by EU Directive 2013/39/EU for biota (0.0085 µg kg^{-1} ww for PBDE- 28, 47, 99, 100, 153 and 154) (European Commission, 2013). The results indicate potential risks from dietary exposure to PBDEs in cow milk.

3.3. Meat

3.3.1. OCPs

Red meat (beef), is a significant source of protein for Ghanaian diet. Meat was analysed to identify OCPs in beef fat, lean beef and grasscutter (bushmeat) obtained from Kumasi-Ashanti region (Darko and Acquaah, 2007), and Gomoa-Central region (Blankson-Arthur et al., 2012). Elevated mean concentrations of DDE and DDT ranged between 32-545 µg kg^{-1} lw for beef fat; much lower mean concentrations were in lean meat (range: 6-43 µg kg^{-1} lw). Other OCPs ranged from 0.6-4.3 µg kg^{-1} lw (lindane, dieldrin, endosulfan and aldrin) for lean and beef fat. Mean concentrations of OCP analytes in grasscutter ranged from 0.15-0.78 µg kg^{-1} lw. Mean concentrations of pesticides in lean and beef fat were below EPA tolerance levels for DDT, DDE (5000 µg kg^{-1} lw), endosulfan I, II and endosulfan sulfate (beef muscle:13,000 µg kg^{-1} lw, beef fat: 2000 µg kg^{-1} lw), lindane and dieldrin (beef fat: 7000 and 200 µg kg^{-1} lw respectively) (USDA, 2011). Concentrations detected were below EPA tolerance levels, posing a low risk from dietary exposure to OCPs in food. Possible sources were attributed to cattle feed-contamination with pesticides, and use of pesticides to control ectoparasites (Darko and Acquaah, 2007).

3.4. Cereal products, maize, cowpea and cocoa beans

3.4.1. OCPs

Infant and adult dietary exposures to OCPs were assessed in local and imported cereal-based food and cocoa beans. The highest mean OCP concentrations were recorded in cowpea and maize (LOD-123 µg kg^{-1} dw) (Akoto et al., 2013), followed by cocoa beans (LOD-40 µg kg^{-1} dw) (Okoffo et al., 2016), and cereal (LOD-22 µg kg^{-1} dw) (Akoto et al., 2015b). The highest OCPs in cowpea and maize were β-HCH, β-endosulfan and DDTs (Akoto et al., 2013); that for cocoa beans: γ-HCH and p,p'-DDT (Okoffo et al., 2016). In cereal, the highest contributions were from γ-HCH (local cereal-22 µg kg^{-1} dw) and β-HCH (imported cereal-14 µg kg^{-1} dw) (Akoto et al., 2015b).

OCP concentrations in cereal, cowpea and maize exceeded MRLs, whereas concentrations in cocoa beans were below. Approximately 90% of baby food exceeded EU Directive-2006/125/EC of 10 µg kg^{-1} assigned for pesticides in cereal (European Commission, 2006b). Similarly, OCPs in maize and cowpea exceeded EU MRL of 10 µg kg^{-1} for β-HCH, and 50 µg kg^{-1} for β-endosulfan, p,p'-DDE and DDD, an indication of medium risks from dietary exposure (European Commission, 2016). Calculated HIs were >1 (1.62-151), indicating carcinogenic and non-carcinogenic risk for infants and young children from pesticides in cereal (Akoto et al., 2015b). Health risks from consumption of cocoa beans were estimated as low, since pesticide concentrations were below EU MRLs (γ-HCH: 1000 µg kg^{-1}, β-HCH: 20 µg kg^{-1}, DDTs and dieldrin: 500 µg kg^{-1}, and aldrin: 50 µg kg^{-1}) (European Commission, 2016). Temporal trend plots could not be constructed due to the limited number of studies.

3.5. Fruits and vegetable crops

3.5.1. OCPs

Some fruits and vegetables obtained from market places in Accra, Kumasi, Tamale and farm areas contained OCPs, which exceeded MRLs (Amoah et al., 2006; Bempah et al., 2011a; Bempah et al., 2012; Bempah et al., 2011b; Bempah and Donkor, 2011; Ntow et al., 2011; Owusu-Boateng and Amuzu, 2013). For a total of 1137 fruits and vegetables collected from market, grocery, and farm sites, mean OCPs ranged between 2-200 µg kg^{-1} ww (Bempah et al., 2012; Bempah et al., 2011b; Bempah and Donkor, 2011). Mean concentrations for vegetables- Accra, Kumasi and Tamale were 300-500 µg kg^{-1} ww (Amoah et al., 2006), whilst maximum concentration detected in Kumasi for fruits and vegetables was 190 µg kg^{-1} ww (Bempah et al., 2011a). HI >1 calculated for OCPs showed endrin exceeded the critical value for vegetables from Kumasi (Bempah et al., 2011a), posing a concern for vegetable consumption. An assessment of low health risks of decreased thyroid function, and weight loss from dietary exposure to OCPs in fruits and vegetables, can be expected. Although most vegetables are edible in their raw states, washing and cooking before consumption were advised to reduce ingestion of pesticide residues. Spatial and temporal trends are displayed in Figures S6 and S7; results of trends are summarized in supplementary S3 and conclusion sections.

3.6. Honey

3.6.1. OCPs

The concentrations of OCPs measured in honey from various areas in Western, Brong-Ahafo and Ashanti Regions (LOD-0.01) were below recommended EU MRL (Darko et al., 2017). Low health risks can be expected; however, risks from other POPs remain unknown as studies are yet to be completed.

4. POP concentrations in Humans

Biological monitoring of POPs, involving invasive and non-invasive techniques in human, is performed using breastmilk, blood/serum, hair, saliva, semen, fingernails, and urine. These give an indication of how POPs accumulate in the body via exposure, POPs potentially transferred via placenta, and breastmilk from mother to child, and POPs (and their metabolites) excreted through body fluids (Esteban and Castaño, 2009).

Figure S8 shows POPs data in breastmilk. Methods of extraction, clean-up and analytical detection are summarized in Table S1. A discussion of POPs in human breastmilk and serum is given below. Results from these studies indicate that the primary exposure route to POPs bioaccumulation in human fluids is via food intake; a secondary exposure route include inhalation from contaminated e-waste sites and farms. The presence of HCHs and DDTs indicate their long-term usage and exposure to both breastfeeding mothers and infants within the farming, fishing and e-waste communities in Ghana.

4.1. Breastmilk

4.1.1. PCBs

The risks of exposure to PCBs associated with intake of breastmilk by infants were assessed in 304 breastmilk samples, by determining concentrations in exposed and unexposed primparae and multiparae mothers. Surprisingly, the sum of mean PCBs in non-occupationally exposed mothers (for Accra, Kumasi and Tamale, 30-82 µg kg^{-1} lw) (Asante et al., 2011), were higher than for occupationally exposed mothers (4.4 µg kg^{-1} lw) who lived or worked at contaminated Agbogbloshie e-waste site in Accra (Asamoah et al., 2018). The dominant congeners observed for non-occupationally exposed mothers were PCBs- 153, 138 and 180; occupationally exposed mothers was PCB 28. The unexpected concentrations could indicate other exposure sources, in addition to cumulative years of occupational exposure.

Health risk assessments completed on occupationally exposed mothers indicated low risks to infants: hazard quotient (HQ <1) (Asamoah et al., 2018). Low potential health risks to breastfed infants is expected (Asante et al., 2011). However, concentrations were consistently higher than the Agency for Toxic Substances and Disease Registry (ATSDR) safety standard minimum risk level of 7 µg kg^{-1} lw (0.03 µg kg^{-1} bw d^{-1}) for total PCBs in human milk (Agency for Toxic Substances and Disease Registry, 2000).

4.1.2. OCPs

The mean concentrations of OCPs monitored in breastmilk ranged from LOD-490 µg kg^{-1} lw (Ntow, 2001; Ntow et al., 2008b; Tutu et al., 2013). The mean concentrations indicated the greatest exposure of mothers to DDTs and HCHs: 78 and 46 µg kg^{-1} lw, and below the LOD to 490 µg kg^{-1} lw in 2 farming communities (Ntow, 2001; Ntow et al., 2008b); whilst Ada fishing community had the least exposure: 30 and 12 µg kg^{-1} lw (Tutu et al., 2013). In an absence of OCP safety standards in humans, based on recommended safety standards in rats, the equivalent milk OCP concentrations that would induce developmental toxicity: 2300 µg kg^{-1} lw (van den Berg et al., 2017), were not exceeded.

4.1.3. PBDEs and HBCDs

The sum of mean concentrations of PBDEs and HBCDs ranged from 2.2-5.8 and 0.3-2.3 µg kg^{-1} lw respectively, in breastmilk from Accra, Kumasi and Tamale (Asante et al., 2011). In comparison to Tamale (2.5 µg kg^{-1} lw), high mean concentrations in Accra (4.8 µg kg^{-1} lw) and Kumasi (5.8 µg kg^{-1} lw) were attributed to greater exposure to PBDE-consumer products and dietary preferences (Asante et al., 2011). PBDEs and HBCDs in breastmilk provided a low exposure risk to breastfed infants, as the estimated daily intake were below USEPA reference dose for PBDE-47 and 99 (0.1 µg kg^{-1} bw d^{-1}), and PBDE-153 (0.2 µg kg^{-1} bw d^{-1}) in human milk (USEPA, 2008a, 2008b, 2008c).

4.2. Blood/serum

Blood and serum from urban-Accra and rural areas-Offinso and Tono Irrigation sites in occupationally exposed workers, were analysed for PCDD/Fs, PCBs, and OCPs (Ntow et al., 2008b; Wittsiepe et al., 2015).

4.2.1. PCDD/Fs and PCBs

In a cross-sectional study of e-waste workers from Agbogbloshie with control group, median PCDD/F concentrations in exposed populations (6.2 pg WHO-TEQ g^{-1} lw, range: 2.1-42.7 pg WHO-TEQ g^{-1} lw) were higher than in controls (4.6 pg WHO-TEQ g^{-1} lw, range:

1.6-12 pg WHO-TEQ g^{-1} lw) (Wittsiepe et al., 2015). Human exposure assessments to PCDD/Fs and dlPCBs, from body burdens, are relevant when factors such as body weight, fraction of PCDD/Fs and dlPCBs absorbed, and half-life are utilized in estimating daily intakes. In an absence of health risk assessments of body burdens for both e-waste workers and control groups, a feasible estimate of potential risks would have to be based on a comparison of daily intake in order to compare with the recommended guideline range of 1-4 pg WHO-TEQ kg^{-1} lw bw d^{-1}.

In contrast to PCDD/Fs, associations between PCBs in exposed and control populations did not follow the expected trend. High concentrations were observed for PCBs-138, 153 and 180 in control groups; geometric mean concentrations were significantly higher, ~3 times that observed for exposed groups (PCB-138: 0.04 µg L^{-1}, PCB-153: 0.05 µg L^{-1} and PCB-180: 0.03 µg L^{-1} whole blood). A strong correlation was observed between work exposure time for e-waste workers who live on site; no correlation was found between PCBs concentrations and age (Wittsiepe et al., 2015).

4.2.2. OCPs

Serum of male and female vegetable farmers analysed for OCPs, indicated high mean concentrations of dieldrin (127 µg kg^{-1} lw) (Ntow et al., 2008b). No gender dependence of total OCPs was observed on comparison of residues between male and female farmers. Mean concentrations in male versus female serum were 10.6 vs 7.1 µg kg^{-1} lw DDTs, 6.9 vs 8 µg kg^{-1} lw HCHs, and 134 vs 115 µg kg^{-1} lw dieldrin, respectively (Ntow et al., 2008b). Although HCHs are excreted during lactation, higher HCHs residue (8 µg kg^{-1} lw) were observed in female serum. Concentrations of OCPs detected in female serum could indicate possible health risks to foetus when bioaccumulated contaminants are transferred transplacentally (Ntow et al., 2008b). Although there are no tolerance levels for OCPs in blood, an assigned reference dose of 0.5 µg kg^{-1} bw d^{-1} for DDT and 0.3 µg kg^{-1} bw d^{-1} for HCH by USEPA, will not be exceeded if an average body weight of 60 kg is considered.

4.3. Urine samples

Urine is considered an ideal matrix for non-persistent chemicals; it has however been used to monitor pesticides and their metabolites in several studies (Aprea et al., 2002). Within farming communities in Ghana, improper and illegal use of pesticides can expose farmers to absorption from the gut, by lungs and across skin. Long-term farming exposure activities (above 30 d yr^{-1}) such as mixing and application of complex combinations of insecticides/pesticides increased risks of chronic coughs, wheezing, and phlegm production. Out of 8 OCPs determined in 100 urine samples, mean concentrations of β-HCH, heptachlor and endosulfan sulphate (2800 ng L^{-1}, 3600 ng L^{-1} and 3300 ng L^{-1}) were noted to strongly correlate with respiratory symptoms (Quansah et al., 2016).

5. Conclusions

In this comprehensive and systematic review, our purpose was to collate and review data from previous studies undertaken on Stockholm POPs in Ghana, since 2001. We conducted a review on POPs in different matrices, compared concentrations against relevant health criteria, and where data was available provided a discussion on spatial and temporal trends (Figures S1-S8, supplementary information S3). Following this information, we estimated the extent of POP contamination by identifying concern levels in matrices with ranking from low, moderate, and high, to no data. POPs of high concern where assigned due to data scarcity, increasing trends, and exceedances of relevant health criteria (Table S6). For 11 matrices and 10 POP-groups assessed in this review, 52% (58 instances) were classified as no data, 8% (9 instances) were identified as high risk, 13.6% (15 instances) were identified as moderate risk, and 25.4% (28 instances) were identified as low risk (Table S6).

In lakes and drinking water, high risks were observed for PCBs, DDTs and PFASs; moderate risks were identified for several OCPs. Moderate risk for air was identified for

DDTs which showed an increasing trend (Figure S2). In water, moderate risks were identified for endosulfans and HCHs, with increasing concentration trends (Figure S3). A high risk was identified for PCDD/Fs in soil and sediment. Low and moderate risks were identified for most OCPs in sediments, coupled with decreasing concentration trends for HCHs and endosulfans (Figure S5).

Of the different food groups studied, a high risk was identified for PBDEs (in aquatic organisms and dairy products), DDTs in fish (increasing trend) and Drins- sum of endrins and dieldrins (in fruits and vegetables)- Figure S7. In maize, cowpea, fruits and vegetables, moderate risks were identified for DDTs and HCHs. Low risks of DDTs and HCHs were identified for meat, cocoa and dairy products. Large data gaps were identified for PBDEs, HBCDs, PCDD/Fs and some emerging contaminants (PCNs and PFASs). Data on PCBs was scarce.

High risks for humans were noted for both occupationally exposed individuals working at e-waste sites and farming communities, and vulnerable subgroups through exposure to POPs in food. The data reflects a high risk from PCBs due to concentrations in breastmilk exceeding guideline values. A moderate risk was identified for DDTs. The data shows few studies have been undertaken on a limited subset of POPs in humans.

The lack of a widespread consistent monitoring programme, and limited sampling periods, make a robust assessment of spatial and temporal trends challenging. However, there were statistically significant and non-significant temporal trends displaying a decrease in concentrations of some legacy POPs (supplementary S3). The observed decline, although non-significant for some legacy POPs, may be attributed to enforcement of the Stockholm Convention, regulations and legal framework targeting POP elimination and reduction. Conversely, significant and non-significant increases in DDTs, HCHs and endosulfans were observed, and could potentially be attributed to illegal usage, and accumulation of banned pesticides.

From the time-trend analyses, specific POP-pollutants (DDTs, HCHs and endosulfans) in various media are discussed in supplementary information S3.7. These highlight multi-media POP-pollutant occurrences, routes of fate and transport, and differing exposures within the Ghanaian environment.

6. Knowledge gaps and recommendations

Studies undertaken in Ghana over the past 17 years have reported POP concentrations in a wide variety of matrices; however, these have been on local POP distributions. Another issue is the lack of annual measurements and systematic monitoring over time for POPs in all regions.

Temporal data have been assessed, but the majority of datasets do not show trends due to limited sampling periods, and limited sample size. However, the data serves as a baseline for future studies. We hope more consistent monitoring produces nationwide data, leading to informed risk management strategies.

Continuous monitoring should involve screening of matrices via targeted and non-targeted analyses for new and understudied POPs. This would reflect POP contaminants that humans and wildlife are exposed to. This gap could be addressed with a complementary non/semi-targeted analytical approach that would aid in identification of unknown contaminants, and result in more robust risk assessments. Collection of data from a wider range of analytes would be beneficial to help identify the main sources of POPs and establish their importance in different regions. Non-targeted analyses of archived sample extracts could be investigated to assess spatial and temporal trends in data deficient areas.

Table S6 shows a general lack of human, animal and wildlife exposure data. There is no data for various matrices including indoor and outdoor air exposure assessment in workplaces/homes, cored sediments, ground and bore-hole water, wildlife-avian population data, amongst others. To address these gaps in knowledge, further studies would be required. Of high importance would be human exposure studies which could include collection of serum and breastmilk samples from vulnerable groups, occupationally exposed workers, and the general population. Analyses of these samples should ideally be coupled with dietary patterns, and workplace/home exposure hazards in questionnaires to clearly correlate POP concentrations with socio-demographic characteristics.

A potential decline in legacy POPs in Ghana can be foreseen with low-toxicity pesticide alternatives and regulations implemented by EPA-Ghana. However, more consideration could be placed on emerging contaminants (such as PFASs and HFRs), and unintentionally produced POPs (PCDD/Fs, PBDD/Fs, PCNs and dlPCBs), as trends of these contaminants in the environment are less well understood. Similar trends and data gaps identified in this review may be expected in other developing African countries, which highlight these trends as an important area for future study.

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Highlights on Review Article:

1. Current status of POPs in Ghana is reviewed.
2. Health risks from PCDD/Fs at e-waste sites.
3. High health risk from exposure to PFASs and DDT related compounds in drinking water.
4. Large data gaps identified.
5. Future perspectives to include understudied POPs.