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Review

Human and environmental exposure to PCDD/Fs and dioxin-like PCBs in Africa: A review

Patrick Ssebugere [a, *], Mika Sillanpää [b], Henry Matovu [a, b, c], Edward Mubiru [a]

[a] Department of Chemistry, Makerere University, P.O. Box 7062, Kampala, Uganda
[b] Laboratory of Green Chemistry, Lappeenranta University of Technology, Sammonkatu 12, 50130, Mikkeli, Finland
[c] Department of Chemistry, Gulu University, P. O. Box 166, Gulu, Uganda

HIGHLIGHTS

- PCDD/Fs and dl-PCBs were detectable in human and environmental samples from Africa.
- Concentrations of PCDD/Fs and dl-PCBs were high in industrialised and urban centres.
- PCDD/Fs and dl-PCBs in the blood samples from Africa were lower than those from Europe.
- OCDD, OCDF and CB 77 were major contributors to the $\sum$TEQs in the majority of samples.
- Major sources of dioxins were emissions from industries and uncontrolled combustion.

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ABSTRACT

This paper reviews literature for the last two decades with emphasis on levels, toxic equivalencies and sources of polychlorinated dibenzo-p-dioxins (PCDDs), dibenzofurans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs) in Africa. Further, we comprehensively analysed data, interpret differences and identify existing gaps with those from other continents. We observed that high levels of PCDD/Fs and dl-PCBs were reported in environmental and biological samples near densely populated urban and industrialised areas compared to those in rural settings. In general, the concentrations of PCDD/Fs and dl-PCBs in the blood samples from Africa were in the same range as those from Asia but lower than those from Europe. The concentrations of dioxins and dioxin-like compounds in the atmosphere in Africa were comparable to and/or higher than those in developed countries. The reported sources of PCDD/Fs and dl-PCBs in Africa were industrial emissions, obsolete pesticide stockpiles, household heating, recycling of electronic waste, and incineration and combustion of domestic waste. Regional and intercontinental transport of dioxins could not be confirmed because of the lack of sufficient literature on them. Further data about the levels and sources of PCDD/Fs and dl-PCBs in Africa need to be generated to complete the chemical inventories for the continent and to facilitate the implementation of the Stockholm Convention on persistent organic pollutants. The reviewed literature shows that most analyses have been carried out in laboratories outside Africa because of the limited institutional capacity in Africa. More support needs to be given to laboratories in Africa to develop the capacity to accurately quantify dioxins on routine basis.

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* Corresponding author
E-mail addresses: ssebugerep@gmail.com, ssebugere@cns.mak.ac.ug (P. Ssebugere).

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1. Introduction

Polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) constitute compounds that are unintentional by-products of industrial and incineration processes involving chlorine (Kulkarni et al., 2008). The compounds are characterised by being persistent in the environment and can bioaccumulate in adipose tissues of living organisms, biomagnify in food chains and travel long distances to non-point sources (Binelli and Provini, 2003; Hagenmaier et al., 1992; Johnson-Restrepo et al., 2005; Letcher et al., 2009; Shaw et al., 2014).

PCDD/Fs constitute 210 theoretical congeners (75 PCDDs and 135 PCDFs), but 17 of the congeners are substituted with chlorine in the 2, 3, 7 and 8 positions and have aroused concern because of the evidence of widespread environmental contamination and toxic effects on living organisms (Corsolini et al., 2002; Fiedler et al., 1990; Johnson-Restrepo et al., 2005; She et al., 2016; Van den Berg et al., 1998). The congeners of toxic concern include 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7,8-HpCDD, OCDD, 2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8,9-HpCDF and OCDF. The PCDD/Fs have similar chemical properties to those of the dioxin-like polychlorinated biphenyls (dl-PCBs). dl-PCBs are co-planar compounds and constitute four non-*ortho* substituted congeners (CB 77, 81, 126 and 169) and eight mono-*ortho* PCBs (CB 105, 114, 118, 123, 156, 157, 167 and 189).

2. Environmental and human exposure to PCDD/Fs and dl-PCBs

2.1. Sources of PCDD/Fs and dl-PCBs in the environment

Africa has experienced rapid urbanisation and industrialisation over the last two decades (Turok and McGranahan, 2013). These developments together with increasing energy requirements for a rapidly growing population have resulted in increased pollution of environmental systems on the continent. Furthermore, the illegal *trans*-boundary movements of electric and electronic waste (e-waste) banned under the Basel convention from developed countries in the name of recycling is creating new sources of pollutants in Africa (Gioia et al., 2014; Grant and Oteng-Ababio, 2012). In most developing countries, management of e-waste is done using inappropriate and unregulated methods (Sjödin et al., 2003). These methods constitute a major source of dioxins (PCDD/Fs and dl-PCBs) in Africa, for instance, Tue et al. (2016) reported higher levels of dioxin-related compounds in soils from an informal e-waste (open burning) site in Ghana than those from non-e-waste burning sites suggesting that e-waste burning was the principal source of dioxins.

Other sources of dioxins and related compounds include industrial processes such as thermal processes in the metallurgical industry (sinter plants in iron and steel industry and production of aluminium, cement and zinc) (Ba et al., 2009; Schecter et al., 1988); pulp bleaching and synthesis of chlorinated phenols and phenol-derived chemicals, hexachlorobenzene, technical hexachlorocyclohexanes and chlorides of aluminium, copper and iron (Liu et al., 2009; Öberg and Rappe, 1992; Quaβ et al., 2004; Swanson et al., 1988). In addition, incineration of municipal and hazardous waste contributes to the release of PCDD/Fs and dl-PCBs into the environment, particularly if combustion is incomplete (McKay, 2002). Furthermore, pesticide stockpiles, electricity generation facilities, paint and printing ink and dielectric fluids of capacitors and transformers constitute possible sources of dioxin emission (Folarin et al., 2018; IPEN, 2005b; Samara et al., 2006). Non-point sources of dioxins include volcanic eruptions, burning of domestic waste and forest and agricultural fires (Kim et al., 2003).

Availability of organic pollutants in Africa is largely influenced by atmospheric deposition, precipitation, inflowing rivers, urban centre drainage channels and near-shore run-off (Arinaitwe et al., 2016). Atmospheric transport and deposition of pollutants in Africa is driven by oceanic winds, ambient temperatures and relative humidity. The warm ambient temperatures in Africa are likely to favour volatilisation and transport of organic pollutants, while relative humidity facilitates their condensation and deposition (Arinaitwe et al., 2018; Barhoumi et al., 2018; Guazzoni et al., 2013). Owing to their large Koc values, deposited dioxins tend to adsorb strongly onto particulate matter such as dust, surface sediments, macrophytes and organic carbon in the water column. In aquatic media, this partition pattern makes them bioavailable to benthic organisms and micro-invertebrates, which form part of the fish diet, as well as to birds and humans, who feed on the contaminated fish (Omwoma et al., 2015).

Overall, the sources of dioxins in Africa are anthropogenic in nature and are related to industrial processes, open burning of wastes, leakages from electricity generation plants and transformers and obsolete stockpiles of pesticides. While some of the sources from hotspot areas have been established, little has been done to solve the issues related to and establish public health implications of PCDD/Fs and dl-PCBs from these sources. Environmental protection agencies in different countries in Africa need to revisit their management plans to reduce exposure levels.

2.2. Human exposure to PCDD/Fs and dl-PCBs

Exposure pathways of the chemicals to humans include inhalation and dermal contact through the skin (Johnson-Restrepo et al., 2005; Schuhmacher et al., 2009). However, the most significant exposure route is through the consumption of contaminated food, particularly that of animal origin, such as eggs, dairy products, beef and pork (Schlummer et al., 1998). Depending on regional or national food habits and traditions, the actual intake of these chemicals from different food groups may vary considerably. The body burdens caused by the chemicals are dependent on dietary habits, i.e. whether one is consuming a variable diet or food from the top of the food chain. Children receive greater exposure to pollutants because they inhale or ingest more on a body weight basis than adults (EFSA, 2005). Breast milk is a major source of exposure to dioxins and/or related compounds for nursing infants (Alaluusua et al., 1996).

In Africa, data about the dietary intake of dioxins are limited, possibly because of the scarcity of information regarding the quantities of food consumed by different age groups in each country. Dietary intake rates have been calculated for some countries using toxic equivalents (TEQs) based on toxic equivalent factors (TEFs) evaluated by World Health Organization (WHO), 1997 (Van den Berg et al., 1998). In Egypt, dietary intake of PCDD/Fs and dl-PCBs was estimated to range from 6.04 to 6.68 pg TEQ/kg body weight (bw)/day, which was higher than the WHO tolerable intake of 4 pg TEQ/kg bw/day (Loutfy et al., 2006). The major contributor to the total TEQs was cheese (a dairy product). In Kenya, Shih et al. (2016) reported maximum dioxin intake rates as 2.15 pg TEQ/kg bw/day through diet, whereas in Tanzania, Polder et al. (2016) reported a maximum intake rate of dioxins as 6 pg TEQ/kg bw/week through consumption of eggs of free-range chicken. The studies in Egypt, Kenya and Tanzania highlight the importance of continuous monitoring of persistent organic pollutants (POPs) such as PCDD/Fs and dl-PCBs in dietary components such that the data would be used in policy formulation by the relevant government agencies and in implementation of the Stockholm Convention on POPs.

Another study conducted by Reeuwijk et al. (2013) reported that mothers in some African countries (such as Cameroon, Uganda, Democratic Republic of Congo, Zimbabwe and Cote d'Ivoire) are exposed to dioxins through the consumption of clay during pregnancy because the levels of dioxins in clay products correlated well with those in breast milk. The authors reported a maximum concentration of 103 pg TEQ g^{-1} in clay products, which was equivalent to an intake of 333–887 pg TEQ/kg bw/week on the assumption that a 65 kg mother consumes 30–80 g of clay per week. This intake rate was 24–63 times higher than the tolerable weekly intake of 14 pg TEQ kg bw. Compared to the studies conducted in Egypt and Kenya, this study showed that African mothers experience higher dioxin exposure through geophagy than through diet.

2.3. *Effects of PCDD/Fs and dl-PCBs on living organisms*

2.3.1. *Humans*

Most toxic actions of dl-PCBs and 2,3,7,8-substituted PCDD/Fs are mediated through mechanisms where the congeners bind to the aryl-hydrocarbon (Ah) receptor (Ahlborg et al., 1992). Studies have reported that PCDD/Fs and dl-PCBs can activate the Ah-receptor in the cytoplasm of most vertebrates and cause a range of toxic effects in laboratory animals, wildlife and humans (Denison and Heath-Pagliuso, 1998; Denison et al., 2002). Exposure of humans to high levels of dioxins and related compounds may result in skin lesions, altered liver function and sensory abilities and delayed cognitive development (Chen et al., 2006). A study by Wesselink et al. (2014) reported that exposure of mothers to the chemicals in the early stages of pregnancy causes birth defects. 2,3,7,8-Tetrachlorodibenzodioxin (TCDD) is a cancer-causing agent and can disrupt multiple endocrine pathways (Son and Rozman, 2002). The compounds can also interfere with thyroid hormone homeostasis by altering deiodinase activity, and this increases thyroid hormone catabolism through glucuronidation (Hood and Klaassen, 2000). Occupational exposure of humans to dioxins and related compounds has been reported to cause chloracne (White and Birnbaum, 2009). Whereas a significant population of Africans is likely to be exposed to dioxins through the aforementioned sources, literature on the cause–effect relationships is scarce and needs to be generated in African populations.

2.3.2. *Other animals in the ecosystem*

In animals, dioxins and related compounds have been reported to cause a variety of toxic effects, such as suppression of the immune system, weight loss, hepatotoxicity, dermal toxicity, gastric lesions and haemorrhage, thymus atrophy, immunotoxicity, teratogenicity, enzyme induction and vitamin A depletion (Brouwer et al., 1989; Dickson and Buzik, 1993; Mos et al., 2007; Schwacke et al., 2012). Exposure of pregnant animals to dioxins at extremely low levels (doses that do not adversely affect the mother) leads to alterations in the reproductive system. For instance, Allen and van Miller (1978) reported that adult female rhesus monkeys that fed on a diet containing 0.5 μg/kg/day of TCDD for 3 months suffered irregularities in menstrual cycle, poor conception and abortion. Gray et al. (1997) found that exposure of male rat offspring to TCDD caused a reduction in sperm count and alteration of their mating behaviour. A study conducted by Ikeda et al. (2005) found that *in utero* exposure to TCDD induces demasculinisation in male offspring by inhibiting the aromatase activity in the brain during the development of the central nervous system.

The toxic and biological effects of pollutants depend on several factors such as age, dose, route of administration, species and sex of the organism (Safe, 1986). Despite the broad range of adverse effects of the compounds, dose–effect relationships have not been studied in the African context, which underscores the need to intensify investigation into the current state of affairs. Free-range chicken in Africa have been shown to have high burdens of PCDD/Fs and dl-PCBs as reflected in the levels in their eggs (IPEN, 2005a-e; Polder et al, 2016). The scavenging nature of the chicken exposes them to particle-bound PCDD/Fs and dl-PCBs. Despite availability of such findings, little is still known about the possible effects of the compounds on the chicken in Africa. In some regions of the continent, concerns have been raised over the declining populations of fish-eating birds such as the African fish eagle (*Haliaeetus vocifer*) (Omwoma et al., 2015). However, causal relationship with levels of dioxins in Africa has not yet been established.

To prevent and/or eliminate any impacts on the environment and health of living organisms caused by exposure to specific POPs, the Stockholm Convention was adopted in May 2001 and entered into force in May 2004 through cooperation with several countries around the world (Hagen and Walls, 2005). Many countries in Africa acceded to the convention and pledged commitment to protecting the environment and living organisms from the toxic chemicals of global concern, among them being dioxins and related compounds. Whereas there are several reviews on PCDD/Fs and dl-PCBs in Asia, Europe, and North and South America (Alcock and Jones, 1996; Chan and Wong, 2013; Domingo and Bocio, 2007; Jaacks and Staimez, 2015; Munschy et al., 2008; Ulaszewska et al., 2011), little is known about their status in Africa. This paper is a critical review of the levels, TEQs and sources of the pollutants in Africa. However, the fate, transport and comparison of the current to previous status of dioxins have not been reviewed because of the scarcity of available data.

3. PCDD/Fs and dl-PCBs in Africa

The majority of the papers reviewed in this paper report results of dioxin analyses performed in laboratories in Europe, North America and Asia, because of limited funding and shortage of qualified personnel to perform the analyses. In Africa, only South Africa and Egypt have reported the capacity to carry out these analyses. However, some efforts are being undertaken to develop the capacity in laboratories in Africa. Recently, 14 laboratories (from Nigeria, Egypt, Senegal, Ghana, South Africa, Uganda, Cameroon, Mali, Ethiopia, Tanzania and Kenya) participated in inter-laboratory assessment of POPs and 2 of these analysed PCDD/Fs, while 3 of these analysed dl-PCBs (UNEP, 2017). It is therefore important that African governments continue supporting capacity-building and acquisition of modern instruments. In addition, regional collaborations are encouraged.

3.1. Aquatic environments

Scarce information is available about the status of PCDD/Fs and dl-PCBs in dissolved and suspended phases of water due to the hydrophobicity of compounds, meaning that they prefer adsorption to particulate matter and/or sediments (Eljarrat et al., 2005). Therefore, to assess the current status of the chemicals in the aquatic environment, this study focuses on sediments and fish. The concentrations, toxic equivalents and sources of dioxins and related compounds in Africa are discussed in this section.

3.1.1. Sediments

The loading of dioxins in aquatic environments is driven by atmospheric deposition, riverine inputs and runoff from inland zones. Sediments have been reported to be major reservoirs and sinks for the chemicals because of their large adsorption capacity (Evenset et al., 2007). Several authors have reported the occurrence of dioxins in sediments from water bodies in Africa. In Egypt, El-Kady et al. (2007) reported concentration ranges of 154.94–249 and 239.67–775.09 pg g^{-1} dry weight (dw) for dl-PCBs and PCDD/Fs, respectively, in 36 sediment samples from the Nile River. The TEQ values were 0.19–0.54 pg g^{-1} dw for dl-PCBs and 4.17–16.32 pg TEQ g^{-1} dw for PCDD/Fs.

A study conducted by Nieuwoudt et al. (2009) quantified the concentrations of PCDD/Fs and co-planar PCBs in sediments from central South Africa. High-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS) and H4IIE-*luc* bioassay were used to identify and quantify individual PCDD/F congeners. The maximum concentrations of PCDD/Fs and co-planar PCBs determined by bioassay measurements were 45 pg TEQ g^{-1} dw. The WHO_{2005}-TEQs based on the congener-specific HRGC/HRMS ranged from 0.12 to 32 pg g^{-1} dw. The concentrations were more predominant in sediments from industrialised areas than in those from residential ones. Of the dioxins, octachloro dibenzo-*p*-dioxin (OCDD) was the dominant congener followed by octachlorodibenzofuran (OCDF) and then 1,2,3,4,6,7,8-heptachlorodibenzo-*p*-dioxin (HpCDD). For PCBs, CB 118 was the dominant dioxin-like congener, followed by CB 105. The authors attributed the dominance of OCDD and CB 118 to municipal solid waste and medical waste incinerators in the study areas. The other sources of PCDD/Fs reported by the authors were emissions from coal combustion, iron and steel plants and heating activities. Another study conducted by Rimayi et al. (2016) reported levels of PCDD/Fs as 16–37 pg TEQ g^{-1} dw and 1.5–22 pg TEQ g^{-1} dw in sediments from the Juskei River and the Klip/Vaal River catchment areas, respectively. 1,2,3,7,8-PeCDD and 2,3,7,8-TCDD were the most dominant congeners. The authors attributed the levels to high levels of industrialisation in the studied areas.

In Ethiopia, Urbaniak and Zalewski (2011) analysed PCDD/Fs in surface sediments from Lake Awassa and Koka Reservoir in the Ethiopian Rift Valley. The total ($\sum$) PCDD/Fs in the sediments from the lake were 270.39 and 63.17 pg g^{-1} dw for the reservoir. The authors observed that OCDD was the major contributor of $\sum$PCDD/Fs to the lake (20.31%) and reservoir sediments (43.01%). The WHO-TEQ value for the lake sediments (23.78 pg TEQ g^{-1} dw) was approximately 6 times higher than that of reservoir sediments (4.03 pg TEQ g^{-1} dw). The values exceeded the set interim sediment quality guidelines of 0.85 pg TEQ g^{-1} dw and probable effect level of 21.5 pg TEQ g^{-1} dw recommended by the Canadian Council of Ministers of the Environment (2002). The authors suggested effluents from textile industries, municipal sewerage and household waste as sources of dioxins.

In Uganda, Ssebugere et al. (2013b) reported the maximum concentrations of PCDD/Fs and dl-PCBs in surficial sediments (<60 cm depth) from Napoleon Gulf and Thurston Bay of Lake Victoria as 44.1 and 136 pg g^{-1} dw, respectively. OCDD was the most predominant congener at the majority of the stations (>29.7% for the sum of seven PCDDs). 1,2,3,4,7,8-Hexachlorodibenzo-*p*-dioxin (HxCDD) was detected at low levels in most sediments from the stations. The low concentrations of HxCDD were attributed to biotransformation processes. The authors pointed out that the possible source of PCDD/Fs in the Thurston Bay environment was sugar cane burning on the basis of the ratios of $\sum$PCDFs/$\sum$PCDDs, which were >1. CB 77 was the most predominant congener from the gulf (contributed 83.8–89.3% to the $\sum$non-*ortho* PCBs) followed by CB 126 (4.57–11.5%). CB 169 was detected at low concentrations compared to other congeners, and this was attributed to the fact that the congener shows limited mobility to molecular diffusion processes and is more susceptible to the dissolution loss process. The WHO_{2005}-TEQs for the PCDD/Fs and dl-PCBs were 0.07–5.53 and 0.01–0.21 pg TEQ g^{-1} in the gulf and bay, respectively. The majority of TEQ values were below the interim sediment quality guideline of 0.85 pg TEQ g^{-1} dw. The concentrations of PCDD/Fs and dl-PCBs in the sediments from the gulf, which is near urban and industrial areas, were markedly higher than those from the bay.

In a related study, Ssebugere et al. (2014a) determined PCDD/Fs in sediments from Murchison Bay, Lake Victoria. The Bay is a receiver end for effluents from the capital city of Kampala through the Nakivubo Channel. The $\sum$PCDD/Fs in sediments varied from 68.8 to 479 pg g^{-1} dw. OCDF was the major contributor to the ΣPCDFs at all study stations (it constituted 47–94%), while OCDD was the principal contributor to the ΣPCDDs (accounted for >84% in the majority of the samples). The WHO_{2005}-TEQs ranged from 0.08 to 0.33 pg TEQ g^{-1} dw, and in all cases, they were below the set Canadian quality value. Another study conducted by Ssebugere et al. (2014b) determined dl-PCBs in 24 surface sediments from Murchison Bay, Lake Victoria. The Σ_{12}dl-PCBs varied widely with values ranging from 125 to 809 pg g^{-1} dw. The authors noted that the contaminant levels were significantly higher at stations close to Nakivubo Channel due to effluent discharges. The WHO_{2005}-TEQs for the dl-PCBs varied from 0.04 to 0.64 pg g^{-1} dw. CB 126 was the highest contributor to the Σ_{12}TEQs (70–89%) at all locations. The TEQs in most samples from Murchison Bay were below the interim sediment quality guideline.

In Kenya, Omwoma et al. (2015) studied the status of PCDD/Fs in surface sediments (<30 cm) from three locations of Winam Gulf, Lake Victoria (Kisumu city, Homa Bay and Mbita). The authors reported concentration ranges of 17.4–812 pg g^{-1} dw for dl-PCBs and 38–60 pg g^{-1} dw for PCDD/Fs. The concentrations in sediments from Kisumu city (an industrialised and urban centre) were significantly higher than those from Homa Bay and Mbita, which are far from point sources. The ratios of $\sum$PCDFs/$\sum$PCDDs were 0.06 (Kisumu), 0.07 (Homa Bay) and 0.04 (Mbita), which indicated that the sources of these dioxins were mainly due to combustion processes. The WHO$_{2005}$-TEQs were found to be 0.43, 0.19 and 0.001 pg g^{-1} for $\sum$dl-PCBs and 3.10, 1.20 and 0.09 pg g^{-1} for $\sum$PCDD/Fs at Kisumu, Homa Bay and Mbita, respectively. The TEQ values for the sediments at Kisumu and Homa Bay were higher than those in the Canadian sediment quality guideline. The authors observed that the TEQ values were greatly influenced by high levels of OCDD and OCDF. The congeners are usually produced in high amounts during combustion of fuel oil mixtures and garden waste (Anderson and Fisher, 2002).

Overall, the levels of PCDD/Fs and dl-PCBs in sediments of water bodies in Africa were lower than the data from Lake Maggiore in Italy and Switzerland (Vives et al., 2007); Can Gio, Southern Vietnam and Osaka, Japan (Kishida et al., 2010); Xiangjiang River, China (Chen et al., 2012); Lake Shihwa, Korea (Moon et al., 2012) and Lake Superior and Huron in North America (Shen et al., 2009) but were in the same range as the concentrations reported in sediments from coastal areas of the Istanbul Strait and Marmara Sea, Turkey (Okay et al., 2009); Mondego Estuary in Portugal (Nunes et al., 2011) and Guaymas Basin and Lake Patzcuaro, Mexico (Yunuén et al., 2011) (Table 1). In the majority of cases, the concentrations of dioxins in sediments from water bodies in Africa were below the set guideline value and thus were of no threat to benthic organisms.

3.1.2. *Fish species*

Fish are good bioindicators of contaminants because they accumulate the contaminants from the water column (Bremle et al., 1995). The accumulation of contaminants is mainly influenced by the pH of water and dissolved oxygen content (Eggleton and Thomas, 2004). Most contaminants in fish are derived predominantly from uptake through the diet and through gills. Contaminants are usually ingested by organisms lower in the food chain, which, in turn, are consumed by those occupying high trophic levels (Suedel et al., 1994). Fish consumption is an integral component of a balanced diet, which provides a healthy source of proteins and nutrients. Some of the health benefits of fish consumption include reduced coronary heart disease, stroke and age-related macular degeneration (Seddon et al., 2003).

Although fish is an integral part of the diet, limited literature is available regarding the levels of dioxins in Africa. In South Africa, Munschy et al. (2016) determined levels of dl-PCBs in albacore tuna (*Thunnus alalunga*), a fish species from Southeast Atlantic Ocean. In their study, the authors reported levels of the pollutants as 5.1–331 pg g^{-1} wet weight (ww) with a corresponding TEQ value of 0.147 ± 0.047 pg g^{-1} ww. CB 126 was the most predominant congener (contributed 98% to the TEQs). In Egypt, El-Kady et al. (2007) determined the levels of PCDD/Fs and dl-PCBs in fish collected from the Nile River. The $\sum$PCDD/Fs and $\sum$dl-PCBs were 27.7–121 and 58.5–133.9 pg g^{-1} ww, respectively. PCDFs were more predominant congeners in muscle tissues (constituted 77.2–87.0% of the $\sum$PCDD/Fs) than PCDDs. The results were in contrast to those in liver tissues collected from the Biobio River in Chile, where PCDDs were the predominant congeners (Orrego et al., 2005). The WHO$_{1998}$-TEQ values ranged from 0.06 to 0.32 for dl-PCBs and 4.56–20.4 pg g^{-1} ww for PCDD/Fs. The values reported by El-Kady et al. were within the Tolerable Weekly Intake (PTWI) of 14 pg TEQ/kg/bw/week set by the European Union Scientific Committee on Food and the Provisional Tolerable Monthly Intake of 70 pg TEQ/kg/bw set by the Joint FAO/WHO Expert Committee on Food Additives (WHO, 2001). El-Kady et al. attributed the occurrence of dioxins to commercial formulations of dichlorodiphenyltrichloroethane (DDT), which were once used along the shorelines of the Nile River.

In Ghana, Adu-Kumi et al. (2010) reported PCDD/F concentration ranges of 0.22–0.88 pg g^{-1} ww in fish species from lakes Volta, Bosumtwi and Weija. The $\sum$dl-PCBs in fish from the three lakes varied from 10 to 165 pg g^{-1} ww. CB 77 was the major contributor compared to other PCBs (CB 81, 126 and 169). The authors observed that the relatively big, fatty predatory fish had high concentrations of the pollutants compared to the small ones. The average TEQ value for the two fish species was 0.3 pg g^{-1} ww. The reported dioxin sources by the authors included emissions from

Table 1
Concentrations of PCDD/Fs and dl-PCBs (pg g^{-1} dw) in sediments from water bodies in Africa compared to those in other continents.

Location	$\sum$PCDD/Fs	$\sum$dl-PCBs	References
Africa			
Napoleon Gulf and Thurston Bay, Lake Victoria, Uganda	3.36–51.4	21.4–247	Ssebugere et al. (2013b)
Murchison Bay, Lake Victoria, Uganda	68.8–479	125–809	Ssebugere et al. (2014a; 2014b)
Winam Gulf, Lake Victoria, Kenya	38–860	17.4–812	Omwoma et al. (2015)
River Nile, Egypt	240–775	154.94–249	El-Kady et al. (2007)
Lake Awassa and Koka Reservoir, Ethiopia	63.17–270.39	-	Urbaniak and Zalewski (2011)
Vaal River, South Africa	1.4–183	120-4700	Nieuwoudt et al. (2009)
Europe			
Lake Maggiore in Italy and Switzerland	3.5-2211	3000	Vives et al. (2007)
Mondego Estuary, Portugal	109.68	199.23	Nunes et al. (2011)
Istanbul Strait and Marmara Sea, Turkey	2.04–60.5	17.9–746	Okay et al. (2009)
Asia			
Tokyo Bay, Japan	3150–20,300	-	Sakurai et al. (2000)
Han River, Korea	23.1–368	41.5-4530	Kim et al. (2009)
East River, China	2100-5400	48–240	Ren et al. (2009)
Can Gio, Southern Vietnam and Osaka, Japan	264-11,000	251.74–116,251	Kishida et al. (2010)
Coastal lagoons, Central Vietnam	192-2912	-	Piazza et al. (2010)
Liaohe River, China	13.74–458.5	57.6-4314.7	Zhang et al. (2010)
Daliao River estuary, Bohai Sea, China	11.3–133.2	-	Zhao et al. (2011)
Xiangjiang River, China	876-497,759	-	Chen et al. (2012)
Lake Shihwa, Korea	40-100,000	50-335,000	Moon et al. (2012)
Central Vietnam	203.9–462.9	1.6–22	Tri et al. (2016)
Xiamen Bay, China	60-4089	3–76	Cai et al. (2016)
North America			
Detroit and Rouge River, Michigan, USA	69-1420	-	Kannan et al. (2008)
Lake Superior, North America	5-18,000	9-11,000	Shen et al. (2009)
Huron, North America	3-6100	9-27,000	Shen et al. (2009)
Guaymas Basin and Lake Patzcuaro, Mexico	24.8–121	-	Yunuén et al. (2011)

uncontrolled combustion, particularly indiscriminate bush fires and medical waste incineration.

Recently, Ssebugere et al. (2013a) reported concentrations for $\sum$PCDD/Fs and $\sum$dl-PCBs as 0.07–0.59 pg g^{-1} ww and 0.3–19.0 pg g^{-1} ww, respectively, in *Lates niloticus* and 0.06–0.59 and 0.2–15.7 pg g^{-1} ww in *Oreochromis niloticus*, from Napoleon Gulf and Thurston Bay, Lake Victoria. The authors observed differences in congener concentrations between the two fish species and study sites and attributed this to differences in feeding habits and trophic levels. OCDF was the major contributor to $\sum$PCDD/Fs in most *L. niloticus* muscle homogenates (18.6–45.6%). In Napoleon Gulf, CB 77 was the only detected non-ortho congener in *L. niloticus* (mean 2.1, range 1.8–2.4 pg g^{-1}), while two congeners (CB 77, 0.4 ± 0.1 pg g^{-1} and CB 126, 0.2 ± 0.1 pg g^{-1}) were detected in *O. niloticus*. The TEQ values were 0.01–0.16 pg TEQ g^{-1} for PCDD/Fs and 0.001–0.74 pg TEQ g^{-1} for the dl-PCBs. The values were within the permissible level of 4 pg g^{-1} ww set by the European Union (EU, 2001). In a related study, Ssebugere et al. (2014a) analysed PCDD/Fs in fish from Murchison Bay on the Uganda side of Lake Victoria. The ΣPCDD/Fs in fish ranged from 5.32 to 49.0 pg g^{-1} ww. The authors observed a significant difference in pollutant concentrations between *L. niloticus* and *O. niloticus* and attributed this to species differences in lifestyles and feeding habits. The TEQ values for *L. niloticus* (range 0.001–0.019 pg TEQ g^{-1} ww) and *O. niloticus* (0.001–0.033 pg TEQ g^{-1} ww) were below the European Union set value. OCDD was more predominant than other congeners (the ΣTEQs contributed to >38% to at all locations for both fish species).

Another study by Ssebugere et al. (2014b) reported the $\sum$dl-PCBs in fish from Murchison Bay, Lake Victoria as 11–143 pg g^{-1} ww. CB 118 was the most abundant congener, accounting for 46–69% to the Σ_{12}dl-PCBs. The high levels of CB 118 were attributed to the inability of the fish species to metabolise the congener. The presence of CB 118 in the samples suggested that the potential sources were paint additives, municipal waste plants, iron ore sintering plants or medical waste incinerators. The TEQs for dl-PCBs were 0.01–0.39 and 0.01–0.11 pg g^{-1} ww for *L. niloticus* and *O. niloticus*, respectively, and were within the permissible level set by the European Union.

Generally, the concentrations of PCDD/Fs and dl-PCBs in fish from Lake Victoria were in the same range of data as that reported for the Nile River in Egypt (El-Kady et al., 2007) and lakes Volta, Bosumtwi and Weija in Ghana (Adu-Kumi et al., 2010) (Table 2). Similarly, the PCDD/F levels in fish from water bodies in Africa were in the same range of data as that reported for fish from the Turia River in Spain (Bordajandi et al., 2003) and the Qiantangjiang River in China (Han et al., 2007). The levels of the chemicals were lower than those reported in fish from Río de la Plata Estuary in Argentina and Uruguay (Colombo et al., 2011) but were 2–5 times higher than the data reported in fish from Sepetiba Bay, Rio de Janeiro in Brazil (Ferreira, 2013) and Masan Bay in South Korea (Im et al., 2004). The $\sum$dl-PCBs in the fish from water bodies in Africa were in the same range of data as those reported in the Qiantangjiang River in China (Han et al., 2007) but lower than those in the Turia River in Spain (Bordajandi et al., 2003). In the majority of cases, the TEQ values for PCDD/Fs and dl-PCBs for fish in Africa were well below the set European Union value; hence, the dietary consumption of these species was of minimal health risks to humans and fish-eating birds.

3.2. *Ambient air*

Dioxin and dl-PCBs have high melting points of approximately 320 °C; hence, they are solids at ambient temperatures. Because of the high vapour pressure at high temperatures, they easily volatilise and get released into the atmosphere (Rordorf, 1986). The atmospheric compartment acts as an important vector for the transport of dioxins in the environment and may be the largest exposure route to living organisms (Lohman and Seigneur, 2001). However, despite the availability of large data sets about dioxins in other parts of the world, few reports have been documented for Africa. For the available dioxin studies in Africa, passive air samplers were used. High-volume air samplers were unavailable and, in some cases, could not be used in remote regions where electrical power supply was not available. Other possible reasons for the scarce data could be the methodological complexity of sampling and high costs of analysis of the chemicals.

In this review, we present and discuss the available data sets in a wider context, including comparison to existing monitoring studies. In northern Algeria, Moussaoui et al. (2012) conducted ambient air monitoring campaigns (from May to November, 2009) at Baraki (urban and industrialised location) and Bou-Ismail (an industrial site). The reported average concentrations ranged from 249 to 923 fg TEQ m^{-3}. In most cases, the concentrations were a magnitude lower than those reported during the global monitoring plan (GMP) for POPs in Cairo, Egypt (505–616 pg I-TEQ per filter for 3 months of sampling, corresponding to 1 pg I-TEQ m^{-3}) (GMP, 2009). The reported TEQ values exceeded the threshold limit set by the European Union of 100 pg I-TEQ m^{-3}. 1,2,3,4,6,7,8-Heptachlorodibenzofuran (HpCDF) was the most predominant (41–48% to the $\sum$PCDD/Fs at Baraki and 51–61% at Bou-Ismail), while OCDF was the least detected (5–9% at Baraki and 6–9% at Bou-Ismail). Ratios of PCDD/PCDF varied from 0.09 to 0.12 at Baraki and were 0.16–0.17 at Bou-Ismail. According to the ratio values, the sources of dioxins were industrial waste incinerators and an oil refinery in the vicinity of the study site. The dl-PCB levels ranged from 0.04 to 0.07 pg m^{-3} at Bou-Ismail and 0.11–0.15 pg m^{-3} at Baraki. The concentrations were several times lower than those reported in the ambient air of an electronic-waste dismantling area in southeast China (4230–11,350 pg m^{-3}) (Li et al., 2008), Hong Kong (1572–4331 pg m^{-3}) (Choi et al., 2008) and northern Italy (22.7–249.4 pg m^{-3}) (Colombo et al., 2013).

Recently, Lammel et al. (2013) determined the atmospheric concentrations of PCDD/Fs by satellite and a global multi-compartment chemistry-transport model. The reported near-ground atmospheric concentrations of PCDDs of model-predicted vegetation fire ranged from 10^{-5} to 10^{-3} pg m^{-3}. The authors found out that vegetation fires emit 180 kg/year of PCDD/Fs and that fires contributed 1–10% of the $\sum$PCDD/Fs (average 5% of TCDD) in the rural and background atmosphere of sub-Saharan Africa. A study conducted by Bogdal et al. (2013) investigated PCDD/Fs in ambient air collected from northern Africa. The concentrations of the pollutants varied from 18 to 532 fg TEQ m^{-3} (median 51 fg TEQ m^{-3}). The ambient PCDD/F concentrations were in the same range of data as those reported from major urban areas in the United Kingdom (<50 fg TEQ M^{-3}) (Klánová and Harner, 2013). However, the levels were an order of magnitude higher than the concentrations reported in ambient air from the Pacific Ocean (3.1–87 fg TEQ m^{-3}) (Bogdal et al., 2013) but lower than the data reported in Catalonia, Spain (5–1196 fg I-TEQ m^{-3}) (Abad et al., 2007), São Paulo City, Brazil (47–751 fg TEQ m^{-3}) (de Assunção et al., 2005), Athens, Greece (166.6–701.5 fg m^{-3}) (Mandalakis et al., 2002) and Hong Kong (1724–2927 fg TEQ m^{-3}) (Choi et al., 2008).

Generally, the reported sources of PCDD/Fs and dl-PCBs in Africa were emissions from industries, open burning of domestic waste and e-waste dismantling activities. Other sources of dioxin emission were the burning of coal and wood, particularly in West Africa, and the combustion of biomass, forest fires and open dumping sites, particularly open landfills of solid waste in northern Africa.

Table 2
PCDD/Fand dl-PCB concentrations (pg g^{-1} ww)in fish from water bodies in Africa compared to those in other continents.

Sampling site	Sampling year	Fish species	PCDD/Fs	$WHO_{PCDD/Fs}$-TEQ	dl-PCBs	$WHO_{dl\text{-}PCB}$-TEQ	References
Africa							
River Nile, Egypt	2003–2004	*O. niloticus*	27.7–121	4.56–20.4	58.53–133.9	0.06–0.32	El-Kady et al. (2007)
Lakes Volta, Bosumtwi and Weija, Ghana	2008	*Tilapia zillii, Clarias gariepinus*	0.22–0.88	0.2–0.3	10–165	0.3–0.4	Adu-Kumi et al. (2010)
Murchison Bay, Lake Victoria, Uganda	2013	*L. niloticus,O. niloticus*	5.32–49.0	0.001–0.14	-	-	Ssebugere et al. (2014a)
Murchison Bay, Lake Victoria, Uganda	2013	*L.niloticus, O. niloticus*	-	-	11–143	0.01–0.39	Ssebugere et al. (2014b)
Napoleon Gulf, Lake Victoria, Uganda	2011	*L. niloticus, O. niloticus*	0.06–0.59	0.01–0.16	0.2–18.7	0.001–0.74	Ssebugere et al. (2013)
South East Atlantic Ocean, South Africa	2013	*Thunnus alalunga*	-	-	5.1–331	0.1–0.194	Munschy et al. (2016)
South America							
Río de la Plata Estuary in Argentina and Uruguay	2003–2004	*Prochilodus lineatus*	70–200	11,600–62,200	280,000 –1,100,000	48.8–311.7	Colombo et al. (2011)
SepetibaBay, Rio de Janeiro in Brazil	*2012*	*Lepidopus caudatus, Micropogonias furnieri* and *Mugil cephalus*	0.953 –1.455	0.10,105 –0.2141	4.537–39.168	0.0883 –0.6525	Ferreira (2013)
Europe							
River Turia in Spain	*2000*	*Salmo trutta, Anguilla anguilla* and *Salmo trutta*	1.22–4.39	0.16–0.58	3150–17,000	0.018–0.09	Bordajandi et al. (2003)
River Elbe in Germany	*1984–2005*	*Anguilla anguilla, Abramis brama, Leuciscus cephalus*and *Leuciscus idus*	-	0.48–22	-	8.5–59	Stachel et al. (2007)
Ebro River Basin, Spain	-	Several species	-	3.25–87.1	-	4.0–940	Eljarrat et al. (2008)
Lativian freshwater lakes	-	Several species	0.043–1.84	-	-	-	Zacs et al. (2013)
Mondego Estuary, (Portugal) Vilaine and Slack Estuaries (France), Wadden Sea (The Netherlands) and Sorfjord (Norway)	*2010*	*Platichthys flesus*	0.39–1.30	-	170.3-1002	-	Nanes et al. (2014)
Lake Varese in Italy	*2007–2009*	Several species	-	0.001–1.31	-	0.031–21	Squadrone et al. (2016)
Asia							
Qiantangjiang River in China	2004	*Carassius carassius*and *Parabramis pekinensis*)	1.14–8.18	0.25–2.33	89.83–720.39	0.25–2.33	Han et al. (2007)
Masan Bay in South Korea	1993	*Liza macrolepis, Konosirus punctatus* and *Cleisthenespinetorum herzenstein*	1.4–2.5	0.32–0.53	-	-	Im et al. (2004)
Pearl River Delta in China	-	Several species	-	0.26–3.80	-	0.065–5.25	Wei et al. (2011)
Chenab River, Pakistan	-	Several species	-	-	2600–10,400	0.96–2.9	Eqani et al. (2015)

3.3. *Human blood and breast milk*

Human blood is normally used as a marker to assess human exposure to POPs. In Africa, few studies have determined dioxin concentrations in the human blood. Pieters and Focant (2014) reported mean serum levels of PCDDs, PCDFs and dl-PCBs in a South African Tswana population exposed to burning solid fuels for cooking as 3.29 ± 1.29, 43 ± 0.23 and 2.8 ± 2.82 pg TEQ g^{-1} lipid weight (lw), respectively. The corresponding mean serum levels for participants relying on electricity, gas and paraffin were 3.82 ± 1.41, 1.89 ± 0.62 and 1.83 ± 1.54 pg TEQ g^{-1} lw, respectively. The TEQs were lower (although not significantly) in the latter group for PCDD/Fs but those of dl-PCBs were higher than those in the former group. The study showed that burning of biofuels exposes Africans to higher serum levels of dl-PCBs but not PCDD/Fs. The authors also noted that older participants had higher serum levels of the pollutants than the younger ones.

In Ghana, Wittsiepe et al. (2015) investigated dioxin levels in the blood samples of 21 male e-waste recycling workers (average age 24.7 years) and 21 male controls (24.4 years) from Agbogbloshie in Accra. The reported median concentrations in the exposed and control groups were 6.18 (range 2.1–42.7) and 4.60 (1.6–11.6) pg TEQ g^{-1} lw, respectively. The concentrations were in the same range of data as those reported in the blood samples of children from an e-waste recycling area in Zhejiang, China (mean, 11.66 pg TEQ g^{-1} lw) (Shen et al., 2010), and in the maternal blood and cord blood samples of residents from Tohoku, Japan (7, 1.6–17.5 pg TEQ g^{-1} lw) (Nakamura et al., 2008). However, the levels were lower than those reported in 226 pregnant women aged 19–41 years living in industrialised areas of Germany (4.34–97.3 pg TEQ g^{-1} lw) (Wittsiepe et al., 2008). Recently, Müller et al. (2019) reported dioxin-like activity (dl-activity) in maternal blood samples collected from Northern Tanzania. The dl-activity in maternal blood was determined by Dioxin-Responsive Chemically Activated Luciferase eXpression (DR CALUX®) bioassay, and this ranged from non-detectable levels to 114 pg TEQ g^{-1} lw (median 30.2 pg TEQ g^{-1} lw). The authors, however, observed that brominated rather than chlorinated dioxins and furans contributed to the dl-activity because it positively correlated with polybrominated diphenyl ethers but not PCBs. The findings of the study did not discriminate the contributions of the specific congeners to the TEQs, as confirmation using HRGC-HRMS was not carried out.

In addition to efforts at national levels, global studies are being carried out by the WHO to assess the level of exposure to dioxins and compare differences between countries. The third WHO-coordinated exposure study (2001/2002) included only one country from Africa. A total of 7 pools of breast milk from Egypt were collected and analysed for dioxins. The median level of PCDD/Fs was 22.79 pg TEQ g^{-1} lw (range 17.16–51.5 pg TEQ g^{-1} lw), while that of dl-PCBs was 6.01 (4.43–8.26) pg TEQ g^{-1} lw. The levels of PCDD/Fs were surprisingly higher, while levels of dl-PCBs were comparable to those from other participating countries (Van Leeuwen and Malisch, 2002). The second and third WHO/UNEP Global surveys of POPs including PCDD/Fs and dl-PCBs in pooled breast milk samples showed that Africa had the widest variation in contamination by dioxins (Van den Berg et al., 2017). The studies reported that Kenya and Uganda had the lowest levels of PCDD/Fs, while Cote d'Ivoire and DRC had the highest levels. The surveys also revealed that contamination with PCDD/Fs in the latter countries was not associated with PCB occurrence, as the levels of PCBs in the milk pools were not exceptionally high.

3.4. Chicken eggs

Eggs serve as bioindicators of contamination by organic pollutants because they have high lipid content and are an important component of human diet. Monitoring studies reporting the contamination status of eggs in Africa are limited. The first studies to report levels of dioxin-like compounds in eggs from Africa were carried out by the International POPs Elimination Network, IPEN (2005a-e). Eggs from free-range chicken were collected from hotspot areas in selected countries and their levels exceeded the limits set by the European Commission (EC) (Fig. 1), suggesting potential long-term effects to consumers. The reported sources of dioxins were obsolete pesticides in Tanzania, industries in Egypt, and open burning of wastes (IPEN, 2005a-e). Another independent study (Quinn, 2010) reported mean levels of PCDD/Fs and dl-PCBs in eggs from South Africa as 1.47 and 1.223 pg TEQ g^{-1} lw, respectively. The TEQ values were lower than the EC set limits. Although deliberate temporal trends of dioxins have not been studied in these countries, a recent study by Polder et al. (2016) reported that the mean total dl-activity in eggs from Arusha, Tanzania, was 2.8 pg TEQ g^{-1} lw. Compared to the data reported by IPEN, the levels in the latter study suggest that dioxin levels in eggs from Africa might be declining but at a very slow rate. Therefore, continuous monitoring of spatial and temporal trends is warranted.

Generally, the levels of dioxins in aquatic, atmospheric and biological samples show that dioxins are ubiquitous in Africa. However, baseline levels have not been established in most countries on the continent, which underscores the need for further investigations to understand their sources and fate, as well as establish public health implications of PCDD/Fs and dl-PCBs in Africa. Further, follow-up studies in areas where baseline levels have been established need to be carried out.

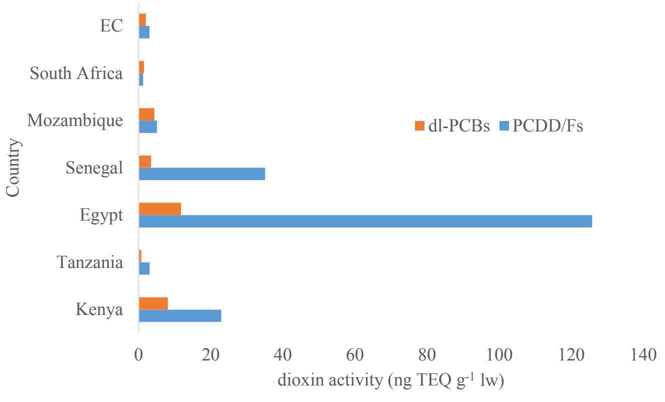

Fig. 1. Mean levels of dioxin-like chemicals in chicken eggs in Africa (IPEN, 2005a-e; Quinn, 2010).

4. Conclusions

The levels, toxic equivalents and sources of dioxins in Africa have been reviewed for the last two decades. Major findings are discussed herein and summarised as follows: studies showed that concentrations of PCDD/Fs and dl-PCBs were generally higher at urban and industrialised sites than at remote and less urbanised areas. Overall, the concentrations of PCDD/Fs and dl-PCBs in the blood samples collected from Africa were in the same range of data as that from Asia but lower than those from Europe. The difference in levels of dioxins in the human blood was attributed to differences in dietary consumption and/or different emission sources. The concentrations of dioxins in the atmosphere in Africa were in the same range of data and/or higher than those in developed countries. The possible reason may be that industrial emissions are more strictly regulated and controlled in developed countries than in Africa, where numerous industries directly dump their waste into the environment to avoid treatment costs. However, more monitoring data are needed to confirm this comparison and explain the differences. Available literature indicated that the largest PCDD/F and dl-PCB emissions were from industrial chemical processes, effluent discharges and open waste burning as well as obsolete stockpiles of pesticides. The contribution of long-range atmospheric transport from developed countries to the levels of PCDD/Fs and dl-PCBs in Africa needs to be investigated further. In terms of institutional capacity, most countries in Africa except Egypt and South Africa have limited facilities specialising in dioxin analysis and, for most studies, analysis was done either in Asia or in Europe. Finally, little data are available about the levels and sources of dioxins in environmental and biological samples from Africa; hence, monitoring of the chemicals in the samples is important to provide a basis for evaluating the effectiveness of the Stockholm Convention on POPs.

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