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# A review of brominated flame retardants in the environment with emphasis on atmospheric levels, knowledge and information gaps in the African continent

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ABSTRACT

Brominated flame retardants (BFRs) are chemicals that are now omnipresent in the environment and have been detected in different matrices from different parts of the world.

They are semivolatile organic compounds (SVOCs), hence they partition between air, water, soil and sediment. Atmospheric long range transport (LRT) is commonly considered the swiftest route for SVOCs to reach remote environments. As a results, the Stockholm Convention on Persistent Organic Pollutants (POPs) advocates air monitoring as a key measure to assess the effectiveness of global control initiatives. There is little information of atmospheric BFR levels in the African continent, which makes it impossible to understand the regional as well as global perspective of these pollutants in the atmosphere. The aim of this review is therefore to establish scientific reasons to explain why it is necessary to have atmospheric research in Africa, and provide research gaps that need to be considered in further studies in order to better understand these pollutants in the atmosphere.

To date there are about 57 published studies on BFRs in different matrices in various countries of the African continent, out of which, only three have concentrated on the atmosphere. The amounts are not as high as detected in other regions but are likely to increase due to the fact that the continent is developing at a fast rate thus an anticipated increase in BFRs use. The review therefore provides different recommendations which all clearly develop a foundation of the necessity of air monitoring of BFRs in the African continent.



**Contents**



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## 1. Introduction

Brominated Flame Retardants (BFRs) are organic chemicals that have been added into consumer products such as carpets, upholstered furniture, electronic devices and curtains since 1970s in order to delay ignition and slow the spread of fire in order to minimize fire related damages and deaths (Hale et al., 2006; Qi et al., 2014; Zhou et al.,2014).

These chemicals, however, are now omnipresent in our environment and have been detected in both human and wildlife tissues in sediments, air, soil, house dust and water samples from different parts of the world (Hale et al., 2003; Janssen, 2005; Law et al., 2006; Watanabe and Sakai, 2003).

There are more than 175 different types of flame retardants categorized into different classes including: nitrogen-containing, organophosphorous, inorganic and halogenated organic (brominated or chlorinated) (Birnbaum and Staskal, 2004). The BFRs are currently the largest market group because of their low cost and high performance efficiency. So far, about 75 different BFRs have been produced on commercial scale (Altarawneh and Dlugogorski, 2014).

There are two categories of BFRs that are commonly reported in literature: Legacy BFRs, which include Polybrominated diphenyl ethers PBDEs, tetrabromobisphenol A (TBBP-A), Polybrominated biphenyls (PBBs) and hexabromocyclododecane (HBCDD) (Fig. 1) and Alternative or Novel BFRs (NBFRs), whereas important representatives include; 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (TBB or EHTBB), hexachlorocyclopentadienyldibromo-cyclooctane (HCDBCO), decabromodiphenyl ethane or 1,2-*bis*(pentabromodiphenyl) ethane (DBDPE), 1,2-*bis*(2,4,6-tribromophenoxy)ethane (BTBPE), bis(2-ethylhexyl)-3,4,5,6-tetrabromo-phthalate (TBPH or BEHTBP), and tetrabromobisphenol A-*bis*(2,3-dibromopropylether) (TBBPA-DBPE) (Janssen, 2005; Covaci et al., 2011; Olukunle et al., 2012). Polybrominated diphenyl ethers have three major commercial products which were produced globally in the last decade: penta − BDE, octa-BDE, and deca-BDE. The high manufacture volume, widespread consumption and environmental persistence of

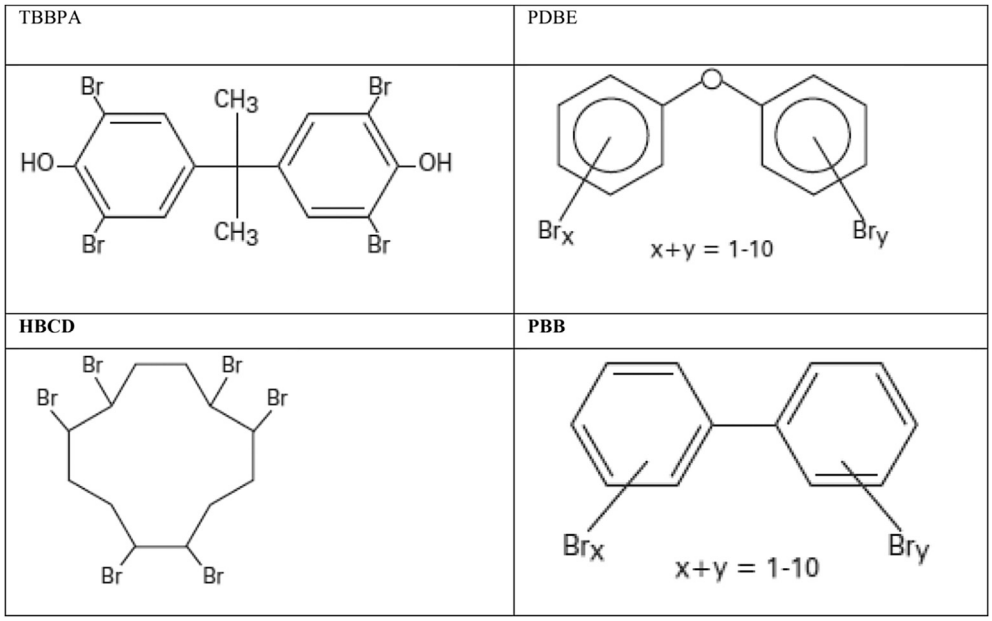


**Fig. 1.** Chemical structures of the common BFRs.

the PBDEs, have led to their pervasive presence in the environment (Yang et al., 2013).

The different classes of BFRs have different properties; PBDE, PBB and HBCD are considered additive, and consequently can leach out of the products into the environment (Eljarrat and Barceló, 2011). TBBP-A is a reactive BFRs as it is covalently bonded to plastic, thus more stable and not easily released to the environment (De Wit, 2002). As a result, the additive BFRs are more likely to be present in the atmosphere than the reactive BFRs. However, few studies have acknowledged the presence of TBBP-A in the indoor and outdoor environment (Sjödin et al., 2001; Tollbäck et al., 2006; Abdallah et al., 2008; Harrad et al., 2010; Abafe et al., 2016).

The urge to monitor BFRs presence in the environment stems from the fact that some of them are toxic. For example, PBDEs have been identified as neuro-toxicants and can cause neurochemical and hormonal deficiencies (Eriksson et al., 2001; Viberg et al., 2002; Besis and Samara 2012; Akortia et al., 2016). As a result, penta- BDE and OctaBDE were banned in Europe and the United States in 2002 and 2003, respectively, and added as new members of persistent organic pollutants (POPs) in the Stockholm Convention on Persistent Organic Pollutants at the Fourth Conference of the Parties (COP-4) held in Geneva Switzerland in May 2009 (UNEP, 2009). Furthermore, deca- BDE is also banned in electrical and electronics in Europe and is being phased out in United States (Dodson et al., 2012; Stapleton et al., 2012; Ma et al., 2013). Hexabromocyclododecane (HBCDD) has also been known as potentially harmful to wildlife and humans and thus was added to the Convention on POPs during COP-6 which was also held in Geneva in April 2013 (Lin et al., 2013).

Parties to the Convention, have to meet the obligations under the Convention leading to the elimination of the listed BFRs (UNEP, 2009). This can only be possible, if information and data on the sources is available to a particular Party. As such, information on concentrations present in the atmosphere is very important to understand the possible sources and hence possible strategies for control of the same. The increase in population and industries exert great pressures on local environment, therefore, improving air quality is an important factor in promoting sustainable development. Atmospheric monitoring is a basic tool for prioritising policy actions so as to map levels of air pollution in order to identify areas requiring special attention; to help assess the number of people exposed to high levels of air pollution; to monitor levels of compliance with air quality standards; to assess the effects of air quality policies; and to help investigate associations between air pollution and health effects. All these cannot be done if there is no data on the concentrations of the pollutants in the atmosphere. It is therefore, very important to measure the BFR concentrations in the atmosphere in all the continents in order to have an overview of the global distribution via air.

So far, there have been three studies that have been conducted in Africa to determine concentrations of BFRs in the air, particularly PBDEs, HBCDDs and NBFRs (Pozo et al., 2008; Shunthirasingham et al., 2010; Arinaitwe et al., 2014).

Besis and Samara (2012), Shunthirasingham et al. (2010) and Mochungong and Zhu (2015) have pointed out the need to establish concentrations of BFRs in the atmosphere in African countries due to the lack of data which makes it impossible to understand the regional as well as global perspective of these pollutants in the atmosphere. The aim of this review is, therefore, to establish scientific reasons to concur with the aforementioned studies and to provide gaps in the area that need to be further researched on.

## 2. BFRs in the atmosphere

Many BFRs are known to have lipophilic (fat soluble) rather than water soluble characteristics (Qi et al., 2014). BFRs mostly exhibits atmospheric exchange with the terrestrial surface, hence the tendency to illustrate low measurements in water samples and higher measurements in sediment, sewage sludge, and particulate samples such as dust particles, because of their high affinity for binding to particles (Janssen, 2005). Moreover BFRs have high log octanol-air partition ($K_{OA}$) values (8–15) compared to PCBs (7–11), PAHs (6–13), and PCDD/Fs (8–13) (Tian et al., 2012).

BFRs are also considered semivolatile organic compounds (SVOC), hence they partition between air, water, soil and sediment. Atmospheric long range transport (LRT) is commonly considered the swiftest route for SVOCs to reach remote environments. As a results, The Stockholm Convention on POPs advocates air monitoring as a key measure to assess the effectiveness of global control initiatives (UNEP, 2007).

The most fundamental factor affecting atmospheric transport of particles and thus particle-bound flame retardants is their aerodynamic diameter (Okonski et al., 2014). Particles with diameter (0.08 μm - 2 μm) cannot undergo rapid gravitational settling (Bidleman, 1988); hence they tend to linger in the ambient air for a longer time and are not as efficiently removed by wet or dry deposition, leading to an extension in their atmospheric lifetime (Aulinger et al., 2007). Consequently, information on size-specific particle data can assist in understanding and predicting POPs transport in the atmosphere. However, there is limited data on size specific particle distribution of flame retardants (Okonski et al., 2014).

BFR levels in the environment are highest near industrial facilities involved in the production and manufacturing of flame retardants as well as electronic recycling facilities (De Wit, 2002; Butt et al., 2004). However, due to their persistence and potential for LRT through the atmosphere, these pollutants are now present in regions where they were never used, even in the Polar Regions (Birnbaum and Staskal, 2004). Different studies have illustrated the presence of different types of BFRs in areas where they were neither used nor produced. These studies prove that the atmosphere is a communal resource that respects no boundaries. Table 1 shows some published articles which have proven the LRT of BFRs. Some of the studies suggest that atmospheric LRT from lower latitude regions such as Africa is known to be one of the major important sources for the presence of many persistent organic pollutants in the Arctic environment today (Hung et al., 2016; Wang et al., 2016a,b; Salvadó et al., 2016; Kallenborn et al., 2015; Salamova et al., 2014; Arellano et al., 2014; Li et al., 2011; Xie et al., 2011). Furthermore, Klánová, et al., (2009) monitored POPs in the African continent and found lower concentrations in background sites, but significant concentrations in big cities, which indicates the need to monitor other POPs such as BFRs in Africa.

## 3. Sources of emissions of BFRs in the African continent environment

As laid out by the Stockholm Convention on Persistent Organic Pollutants (POPs) and the Convention on Long Range Transboundary Air Pollution (UNECE CLRTAP), a significant approach toward controlling POPs in the environment is identifying emission sources (Csiszar et al., 2012). The emission sources of BFRs in the African continent hence include.

### 3.1. Point sources

#### 3.1.1. Waste electrical and electronic equipment (WEEE)

The most apparent sources of emission of BFRs into the environment are effluents from factories producing BFRs, flame retarded polymers, and plastic products such as electrical

**Table 1**
Different studies showing the concentrations ($pgm^{-3}$) of BFRs in the atmosphere (gaseous and particulate), where they were not produced.

| Location | Year[a] | Statistic | Type | ΣPBDEs | Congener | PBB | PBT | DPTE | HBB | Ref |
|---|---|---|---|---|---|---|---|---|---|---|
| Atlantic and Southern Ocean | 2008 | Range | g | 0.31–2.85 | BDE47, 99 | | nd-0.04 | 0.03–1.89 | 0.04–10.8 | (Xie et al., 2011) |
| | | | p | <0.04–2.16 | | | nd | 0.01–0.4 | nd-0.12 | |
| East and South China Seas | 2008 | Mean | g&p | 10.8 ± 6.13 | BDE47, 99 | | | | | (Li et al.,2011) |
| Bay of Bengal and the Andaman Sea | | | | 3.22 ± 1.57 | | | | | | |
| Indian Ocean | | | | 5.12 ± 3.56 | | | | | | |
| Atlantic Ocean | | | | 2.87 ± 1.81 | | | | | | |
| George Island, Antarctica | 2009–2010 | Range | g&p | 0.67–2.98 | BDE17, 28 | | | | | (Li et al., 2012) |
| North Sea | 2010 | Range | g&p | 0.2–10.7 | BDE47, 99,209 | n.d. - 0.81 | nd-0.24 | nd-2.5 | 0.09–6.3 | (Möller et al., 2012a) |
| Pacific Ocean (East Indian Archipelago and Philippine Sea) | 2010/11 | Range | g&p | 0.14–4.6 | | | 0.71–2.2 | 0.44–2.3 | 3.7–19 | (Möller et al., 2012b) |
| Indian Ocean | | | | nd-6.6 | | | nd-2.8 | nd-1.1 | 0.15–26 | |
| Southern Ocean | | | | 0.13 | | | nd | nd | 0.12 | |
| Tibetian Pleateau | 2006–2008 | Range | g&p | 0.83–5.2 | BDE47, 99 | | | | | (Xiao et al., 2012) |
| Canadian Arctic | 2006–2007 | | | 1.2–55 | | | | | | |
| Longyearbyen (European Arctic) | 2012 | Mean | p | 5.6 ± 1.1 | BDE209 | 0.14 ± 0.05 | | | 0.23 ± 0.06 | (Salamova et al., 2014) |
| Western Sub-Arctic (Canada) | 2011–2014 | Range | g&p | 0.42–18 | BDE47,99 | | 0.007–0.47 | | 0.024–0.013 | (Yu et al., 2015) |
| Chinese Great Wall Station, West Antarctica | 2011–2014 | Range | g&p | 0.60–16.1 | BDE209 | | | | | (Wang et al., 2016a,b) |
| North Greenland | 2008–2012 | Range | g&p | nd-6.26 | BDE99 | | | | | Bossi et al., 2016 |
| Western Antarctic Peninsula | 2010 | Range | g | 1.4–7.6 | BDE47 | | | | | (Khairy et al., 2016) |

[a] Year of sampling.

appliances (Watanabe and Sakai, 2003). Electronic waste (e-waste) recycling has demonstrated to be an important emission source of BFRs to the atmosphere (Li et al., 2007). Electrical and electronic equipment's has also been a major source of emission of BFRs in Africa. The demand for electrical and electronic equipment's (EEE) is growing in Africa, resulting to a higher production of WEEE (La Guardia et al., 2013). According to Baldé et al. (2014) the total WEEE generation in Africa was 1.9 Mt in 2014, which was lowest compared to Asia (16 Mt), America (11.7 Mt), and Europe (11.6 Mt). However, nearly 80% of all WEEE produced in developed countries is presently exported to developing nations under recycling disguise (La Guardia et al., 2013), It has, therefore, been predicted that by 2018, developing nations will dispose more end-of-service computers than developed countries. WEEE recycling in developing countries has attracted substantial attention as a major source of many toxic chemicals. The recycling procedures in these regions are usually primeval and hazardous resulting to emissions of high concentration of pollutants including BFRs in the environment. The atmosphere plays a significant role in transporting BFRs from their sources to rural/remote locations (Tian et al., 2011).

Environmental releases of toxicants from unfettered WEEE salvaging has been observed, which is of concern for countries with weak environmental and human health practices (La Guardia et al., 2013). Most African countries ranked very low according to the Yale University's 2012 Environmental Performance Index (EPI) of 132 countries on 22 performance indicators following 10 policy categories including air pollution (effect on human health), air pollution (ecosystem effects) among others (Emerson et al., 2012).

### 3.1.2. Wastewater treatment plants (WWTP)

Wastewaters usually contain a combination of organic contaminants resulting from raw materials and everyday products. The major source of these contaminants is the collection of input of industrial and urban effluents (Sánchez-Avila et al., 2009). Numerous contaminants including BFRs and NBFRs have been identified in wastewaters (Song et al., 2006; Richardson, 2008; Daso et al., 2012; Lee et al., 2014; Li et al., 2016; Wang et al., 2016a,b). Due to their low aqueous solubility and high hydrophobicity, PBDEs are inclined to be adsorbed on suspended solids in wastewater and thus can be highly detected in the sludge as well (Wu et al., 2016; Kim et al., 2016; Novak et al., 2016; Stiborova et al., 2017). High concentrations of deca-BDE has been found in male birds scavenging in waste management facilities (Gentes et al., 2015), signifying that WWTP was a potential source of PBDEs. There are few studies that have already identified WWTP as a source of PBDE into the ambient air (Weinberg et al., 2011; Martellini et al., 2012). Therefore, WWTP in Africa can also be a contributing source of emissions of BFRs into the atmosphere.

### 3.1.3. Landfill sites (where products flame retarded with BFRs are dumped)

In most developing countries especially in the African continent, waste is not separated before being disposed of in landfills. Most of the landfill sites in Africa are open dumps with little or no sufficient measures to preclude environmental pollution (Kajiwara et al., 2014). Fugitive emissions of dust during transportation, unloading and disturbance of waste can result to emissions of BFRs into the atmosphere (Sakai et al., 2006). Earnshaw et al. (2013) suggests that the major source of emissions to all environmental compartments is the waste management phase predominantly landfill sites; the dominant pathway of BFRs from the Landfill to the atmosphere can be via particle bound emission from use and recycling. Most African countries do not have well engineered landfills, they have open dumpsites and thus most local authorities manage the waste by burning, in order to control the annoyance produced by flying litter (Remigios, 2010). This can be a major source of atmospheric

emissions.

### 3.2. Non point sources

#### 3.2.1. Importation of used vehicles that may have been flame retarded with BFR

The passenger section of vehicles is understood to be a major indoor microenvironment where people are exposed to a wide range of harmful substances including PBDEs (Mandalakis et al., 2008). There has been extensive studies on the presence of brominated flame retardants in dust from cars (De Wit et al., 2012; Harrad et al., 2016; Kalachova et al., 2012; Olukunle et al., 2015c). Many African countries import second hand cars from Japan, UK and other developed countries, due to the high price of owning a new car, some of the used cars are near their end-of -life and hence major sources of BFRs releases. Higher temperatures in African countries can be a huge problem on vehicles that have been flame retarded. It is understood that regular exposure of vehicles to direct sunlight can result to a reach in excessive temperature leading to volatilization of chemical substances from interior surfaces. Solar radiation conduction through glass windows can lead to photochemical reactions and the production of degradation by-products, which may be more dangerous than their precursors (Chien, 2007). Söderström et al. (2004) highlighted the possibility of photolytic degradation of PBDEs to lower brominated BDEs which could result to compounds which are more stable to photolytic biodegradation and more bio accumulative. Hence these compounds could later be present in the atmosphere due to the characteristic of LRAT of these POP compounds. Table 2 shows a preview of the number of cars imported in certain years in some of the top 10 populated African countries.

#### 3.2.2. Indoor environment

Additive BFRs are simply emitted from the surface of consumer products to the indoor environment (De Wit, 2002). The indoor environment contributes a major portion of the overall human exposure to BFRs, as people spend most of their times indoors (>90%), either at home, school or in the office (Ding et al., 2016). High levels of BFRs in the indoor environment can be contributed by electronic equipment's, and upholstered furniture's. Melymuk et al., (2016) found high levels of PBDEs in homes with higher numbers of electronics. In the African continent, most electronics are imported from developed countries, and the demand keeps increasing at a fast rate (La Guardia et al., 2013), this might result to higher levels of BFRs in future. Recent studies have indicated the contribution of the concentration of BFRs from the indoor air to the outdoor environment (Gou et al., 2016; Newton et al., 2015; Ding et al., 2016; Melymuk et al., 2016), Some studies have suggested that higher temperatures contribute to higher emissions of pollutants from the indoor environment to the outdoor environment. Melymuk et al., (2016) found high concentrations of PBDEs in the outdoor air than indoor, suggesting that during summer the indoor environments act as a source and air exchange decreases indoor concentrations and raises outdoor concentrations. Furthermore, high ventilation also leads to great air exchange between indoor and outdoor environments, hence higher concentrations of BFRs in the outdoor environments (Newton et al., 2015; Melymuk et al., 2016). Therefore the indoor environment can be a contributing source of BFRs to the outdoor environment in the African continent.

## 4. Presumed characteristics of BFRs in the African continent

Baklanov et al. (2016) pointed out the importance of atmospheric monitoring studies in the developing world because of the different characteristics that are experienced by different regions. The mentioned factors include topography, meteorology, demography, industrialization and socioeconomic development among others.

Most studies on atmospheric BFRs have been conducted largely in mid-to high-latitude regions, while data from low latitude regions are very limited.

Most of Africa lies in tropical and subtropical latitudes, where temperatures are high throughout the year and vary more from daytime to night-time than during the course of the year (Nicholson, 2001). The 4th IPCC Assessment Report provides evidence that Africa is warming faster than the global average, and this is likely to continue (IPCC, 2007; Parry, 2007).

It has been recognized that the air concentrations of SVOCs have a strong relationship to ambient temperature. The temperature dependence of SVOCs in the air is conveniently described by the Clausius - Clapeyron (CC), which can be expressed as;

$$\ln P = m/T + b \quad (1)$$

Where P is the partial pressure (atm), T is the temperature (K), and m and b are fitting parameters. The CC plot has been used to point out the relative importance of evaporation from local surfaces versus long-range atmospheric transport (LRAT) or advection of background air mass in controlling air concentrations (Hoff et al., 1998). In mid-latitude regions POPs can migrate to higher latitudes in a series of relatively short jumps sometimes termed the “grasshopper effect” (Fig. 2) (Wania and Mackay, 1996). The compounds migrate intermittently in tune with seasonal temperature changes in the regions. For low latitude regions, i.e. Africa, the relatively mobile POPs increase concentrations as they move away from the source because of their tendency to partition more easily from the air at cooler temperatures (Calamari et al., 1991). Warm temperatures favour evaporation from Earth's surface in tropical and subtropical regions while cool temperatures favour greater adsorption of these compounds to the atmospheric particulate matter which then deposit on the surface (Wania and Mackay, 1996). Monitoring the concentrations of additive brominated flame retardants is necessary so as to relate to the amount of concentrations that have been observed in high latitude regions.

Research from these regions are, therefore, needed, given that most worldwide BFR contaminated products are recycled in low-latitude regions where the tropical climate favours the release of

**Table 2**
Preview of imported cars in some of the top 10 populated counties in the African continent.

| Country | Population as on 2014 (UNPD, 2015) | %/Projection of car importation | Reference |
|---|---|---|---|
| Nigeria | 178,517,000 | 335,000 used cars imported in 2015 (269% higher than new cars imported) | (PWC, 2016) |
| Ethiopia | 87,952,991 | 28,654 imported vehicles in 2015 | (ERCA, 2015) |
| Egypt | 87,359,900 | 97,000 imported cars in 2011 (41% of the total cars registered) | (Yehya, 2013) |
| Tanzania | 47,421,786 | 54,452 imported vehicles in 2013 | (CITIZEN, 2014) |
| Kenya | 41,800,000 | 76,518 used cars registered in 2014 (81% of the total cars registered) | (Kigotho, 2015) |
| Uganda | 35,357,000 | 39,000 used cars imported in 2014 (95% of the total cars imported) | (Adengo, 2015) |

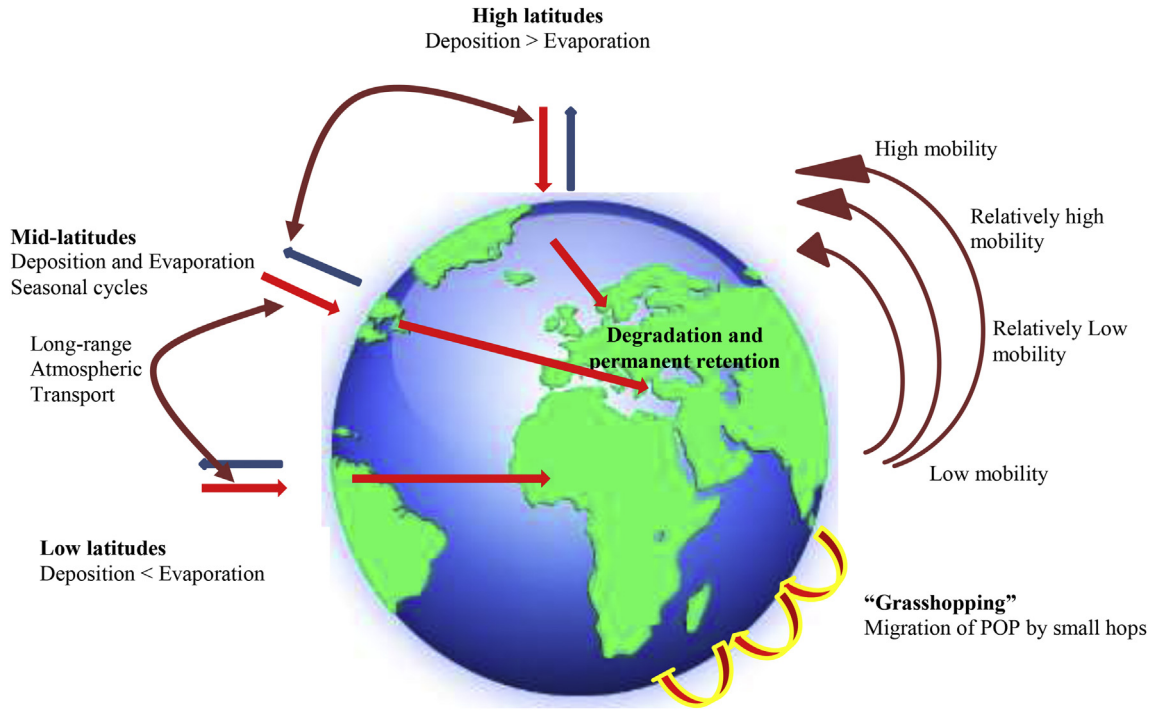


**Fig. 2.** POPs migration process (adapted from Wania and Mackay, 1996).

BFRs into the atmosphere and their subsequent movement to colder regions of the earth (Tian et al., 2012). This indicates a strong evidence of increasing contamination of BFRs to the environment, hence the importance of identifying emerging issues and data gaps as well as generating a future research agenda, particularly in the area of control of releases.

## 5. Intercontinental transport of brominated flame retardants

The atmosphere plays a very significant role in the transportation of POPs (Baek et al., 2013). The nature of the sources and magnitudes of the emissions highly fluctuate across different continents (Piketh and Walton, 2004).

POPs originating from middle latitude regions in the northern hemisphere are thought to be transported by westerly winds (Baek et al., 2013). Conveyance of pollutants from the African continent is also caused by both easterly and westerly synoptic scale circulation patterns (Piketh and Walton, 2004). Higher temperatures experienced by subtropical and tropical regions in Africa and south-eastern Asia are not similar to other regions at the mid latitudes. This characteristic is significant for SVOCs as volatization is a key process by which POPs are emitted in the atmosphere, and it is highly temperature dependent (Breivik et al., 2011).

Table 3 shows the sources and receptor continents of persistent organic pollutants from different sources, which reach different continents depending on the season. Intercontinental transport occurs on a period of about 3–30 days and therefore is significant for compounds with a lifetime of more than three days, such as BFRs (Franklin, 2006). The transport incidents may include either (a) advection relatively horizontally within the planetary boundary layer; or (b) lifting into the free troposphere via deep convection, the influence of mountain slopes on air flow or, in particular, large-scale weather systems, especially the uplifting "warm conveyor belts" that occur ahead of advancing cold fronts. The comparative phenomena of these two incidents differ according to the region as well as the substance (Stohl et al., 2005), hence can neither be ignored nor assumed. Worldwide regulations to protect against significant adverse human health as well as the environmental effects of POPs have been implemented since the early 2000s. However different countries have started to regulate and ban POPs including BFRs at different times, and some countries have not even started regulating them, thus the contamination levels are not presumed to be the same (Glotfelty et al., 2014). It is therefore important to take into account the impact of transport from surrounding countries, as air pollution is not confined to one country. The African continent is neither developed nor industrialized, but it sure makes important contributions to the global atmospheric chemistry, and hence concentrations of BFRs in the atmosphere should be documented.

**Table 3**
Intercontinental emissions of BFRs.

| Source | Continent affected | Reference |
|---|---|---|
| Africa | Australia, South America, North America, Europe and Western Asia | (Piketh and Walton, 2004) |
| Asia | North America, North Pacific, United States, Arctic, Europe | (Glotfelty et al., 2014; Cheng et al., 2007; Stohl, 2006 Wild and Akimoto, 2001; Jaffe et al., 1999; Jaffe et al., 1997) |
| Europe | Asia, United States, Arctic, Africa | (Navarro et al., 2016; Stohl, 2006; Fiore et al., 2002; Holloway et al., 2003) |
| North America | Europe, Arctic | (Stohl, 2006; Pochanart et al., 2003; Holloway et al., 2003) |

## 6. BFR studies in the African continent

Studies on BFR in the African continent are minimal, although the rate of importation of BFRs containing products is high (La Guardia et al., 2013; Schluep et al., 2012). The demand for importation of such products into the African continent is expected to increase in future, considering the fact that the continent is developing at a fast rate (AFDB, 2015). To date a huge majority of studies conducted for PBDEs, have been conducted in Asia, America, and Europe (Harrad et al., 2016). While data is emerging from other regions especially the African continent. To our knowledge below are studies that exist concerning the presence of PBDEs in different matrices.

### 6.1. Aquatic environment

The levels of BFRs in the water samples of the aquatic environment is expected to be small due to the hydrophobic characteristics of these compounds. Higher concentrations will be adsorbed by the particulate matter and sediments (Wurl and Obbard, 2006). There are quite a number of studies that have been done in Africa.

#### 6.1.1. Sediments

A seasonal study on the sediments was conducted from September 2009–February 2011 at the Pangani river basin in Tanzania by Hellar-Kihampa et al., (2013). The $\sum_6$PBDE concentrations ranged from 295 to 2175 pg/g before the wet season 50–940 pg/g during the wet season and 38–920 pg/g during the dry season. The commonly detected PBDEs were BDE47 (50–734 pg/g) and BDE99 (38–1097 pg/g) (Hellar-Kihampa et al., 2013). The obtained values were lower compared to other studies in developed countries.

La Guardia et al. (2013) also determined concentrations of PBDEs, NBFRs and HBCDDs in August 2011 at the coastal area in South Africa. They determined $\sum_{11}$PBDE concentrations and found out that there were higher concentrations of NBFRs than the PBDE and HBCDD levels. The commonly detected compounds were 2-ethylhexyl 2, 3, 4, 5-tetrabromobenzoate (EHTBB) and BDE209 (La Guardia et al., 2013). In comparison to other aquatic systems around the world, Durban Bay's PBDE sediment concentrations were relatively high. Moreover, ∑HBCDD obtained in the study were significantly higher than studies conducted on river sediment in Netherlands and Ireland (La Guardia et al., 2013).

$\sum_8$PBDE, NBFRs and ∑HBCDD levels were also analysed from six rivers between June and August 2013 in Gauteng, South Africa and the concentrations ranged from 0.8 to 44 ng/g, <dl (less than detection) – 310 ng/g and <dl – 186 ng/g respectively, with values lower than those reported in Spain, Korea, and USA but higher than concentrations reported in Hong Kong, Baiyangdian Lake, Da-an River, (Olukunle and Okonkwo, 2015; Olukunle et al., 2015c).

Daso et al. (2015a), analysed $\sum_8$PBDE levels between June 2010 and March 2011 in sediments from two rivers in Capetown, South Africa which ranged from 1.33 to 239 ng/g (dry weight) for both rivers. BDE209 was the major congener in river Diep and BDE47 was the major congener in river Kuils (Daso et al., 2015a). The values were comparable to those previously reported in some European and Asian studies but much higher than those reported In South Africa by Olukunle and Okonkwo (2015).

Concentrations of PBDE levels were also determined in the sediments from the Congo River Basin collected between May and June 2010 and the ∑PBDE ranged from <LOQ to 1.9 ng/g dw with BDE209 as the leading congener (Verhaert et al., 2013). The levels found in the study were lower than values found in developed countries.

Ssebugere et al. (2014) determined concentrations of PBDE in sediments in the Lake Victoria shore in Uganda in March 2011 and the mean $\sum_{11}$PBDE ranged from 60.8 to 179 pg/g dw, BDE47 was the dominant congener followed by BDE99. The concentrations in this study were lower than the data in sediments from other reported studies in Africa, Canada, Spain and United States.

#### 6.1.2. Fish

Verhaert et al. (2013) reported ∑PBDE from fish samples in the Congo River Basin collected between May and June 2010 with a range of <LOQ to 188 ng/g lipid weight (lw), with BDE47 and 99 as the major congeners. $\sum_{14}$PBDE and HBCDD concentrations were determined in the inland and coastal fish from Ghana collected in August 2010. The obtained values for $\sum_{14}$ PBDE and HBCDD ranged from 0.89 to 19 ng/g lw with BDE47 and 99 as major congeners and 0.04–2.2 ng/g lw with α-HBCDD as the main congener respectively (Asante et al., 2013). In Tanzania, a study was done in October and November 2011 to examine PBDE concentrations in fish from four different lakes. The mean $\sum_9$PBDE concentrations ranged from 1.5 to 34.3 ng/g lw with BDE209 as the dominant congener (Polder et al., 2014). Fish samples from the Lake Victoria shore in Uganda were also collected in March 2011 and analysed by Ssebugere et al. (2014). The mean $\sum_{11}$PBDE ranged from 48.2 to 177 pg/g lw with BDE47 as the major congener. Verhaert et al. (2013) and Ssebugere et al. (2014) found the levels obtained in their studies lower compared to those of developed countries while Polder et al. (2014) concluded that the mean levels of ΣBDEs, minus BDE209, were in the same range as corresponding levels from Danube Delta, Romania. Nevertheless, Asante et al. (2013) found PBDE levels higher than studies from Romania, some parts of China, and Germany but lower than Spain, Turkey and another China study. Furthermore the mean HBCDD levels were lower than in China, Europe and several lakes in North America and Canada (Asante et al., 2013).

#### 6.1.3. River

River samples were analysed for PBDEs at the discharge point, downstream and upstream from a wastewater treatment plant in South Africa between June 2010 and March 2011. The mean concentrations of $\sum_8$PBDE were 4.83, 4.29 and 2.60 ng/L for the upstream, point of discharge and downstream respectively with BB153 being dominant with a higher concentration at the point of discharge. The reported levels were lower than those reported in a similar study in China (Daso et al., 2012).

### 6.2. Humans

#### 6.2.1. Breast milk

Different studies have reported BFR levels in breast milk in the African continent. Breast milk provides evidence on transfer of contaminants to infants, it is used as an indicator to measure human exposure to persistent organic pollutants.

Asante et al. (2011) investigated human breast milk collected in Ghana in 2004 and 2009. The mean $\sum_{16}$PBDE (excluding BDE209) and $\sum_{17}$PBDE (including BDE209) for the years 2004 and 2009 were 2.2 ng/g lw and 4.5 ng/g lw with BDE47, 99, 100, 153 and 209 as major congeners. The concentrations were higher in urban areas in comparison to the rural ones and the mean concentration of total PBDEs in this study was higher than some studies in Asian and European countries, but lower than in Canada and USA (Asante et al., 2011).

Müller et al. (2016) presented concentrations of PBDEs in breast milk collected between October and December 2012 in Tanzania which were reported higher than those in Asia and Europe. The $\sum_7$PBDE ranged from <LOD – 785.8 ng/g lw with BDE47 being a major congener especially to mothers living in urban areas (Müller et al., 2016).

Research was also done between May and June 2010 Tunisia by Hassine et al. (2012), whereby the reported $\sum_8$PBDE ranged from 2.49 to 22.62 ng/g lw with BDE183 as a dominant congener. The reported levels were comparable to corresponding levels in milk from Australia, Philippines and China, much lower than those in USA, Canada and Pacific Northwest, and higher than those in developed countries such as Sweden, Norway, Russia, French, Germany, Italy, Taiwan and Japan. Darnerud et al. (2011) investigated PBDE levels in the rural districts of South Africa from April to November 2004 and reported $\sum_8$PBDE ranging from 0.7 to 6.3 ng/g lw with BDE47, 99, 153, and 183 as dominant congeners. The concentrations were comparable to those of Sweden, but lower than other developed countries.

#### 6.2.2. Human serum

So far there is only one study which has investigated concentrations of PBDEs in human serum in Africa. Linderholm et al. (2010) obtained serum samples from an open cohort in Guinea-Bissau between 1990 and 2007. The study detected low levels of PBDEs with BDE 209 as a major congener. The study found an increasing trend of BDE 153 which was comparable to observations found in Sweden and the United States (Linderholm et al., 2010).

### 6.3. Eggs

Eggs from different species have been analysed for the presence of BFRs in the African continent. Polder et al. (2008) investigated BFRs in eggs of eight different bird species in South Africa between November 2004 and March 2005. The mean $\sum_8$PBDE concentrations for the eggs ranged from 2.3 to 369 ng/g lw with BDE 47 as the major congener while $\sum$HBCDD was 71 ng/g lw (Polder et al., 2008). The investigated values were lower than those found in Europe but higher than the eggs found in Romania and Norway (Polder et al., 2008).

Marine bird eggs were collected in 2008 in Mauritius and analysed for BFRs specifically PBDEs and NBFRs by Bouwman et al. (2012). The mean $\sum_9$PBDE concentrations was 0.7 ng/g lw with BDE47 and BDE100 as dominant congeners while the NBFRs were not detected (Bouwman et al., 2012). Terrestrial and aquatic bird eggs from South Africa were investigated and reported to have a mean $\sum_{10}$PBDE concentration ranging from < LOQ − 61 ng/g lw (Bouwman et al., 2013). Both studies showed low levels compared to other studies elsewhere.

Daso et al. (2015b) examined levels of PBDE in eggshells of the Southern Ground Hornbill and the Wattled Crane during the 2012 and 2013 breeding season, the $\sum_8$PBDE concentrations were 46.6 and 80.8 μg/g lw respectively. PBDE 47 was the predominant congener, and the concentrations were comparable to other studies done in Africa.

Polder et al. (2016) analysed BFRs in native free-range chicken eggs from four villages in Tanzania in 2011. The mean concentration of the $\sum_{11}$PBDE ranged from 19 to 81 ng/g lw with BDE 209 as major congener, the concentrations were higher than those reported in Sweden, Spain, Belgium and Canada. $\sum$HBCDD concentrations were 8.4 ng/g lw with comparable values than those reported in Belgium but higher than in Canada and USA. BTBPE concentrations ranging from 0.79 to 4 ng/g lw which were slightly higher than in Irish eggs (Polder et al., 2016).

### 6.4. Indoor dust/air

Brominated flame retardants are omnipresent in the indoor dust and air resulting to human exposure and subsequent concerns about the adverse impact on health (Rauert, 2014).

In South Africa, several studies have been done in the indoor environment; A study was done at a University in South Africa in 2011 to examine concentrations of PBB and PBDE in offices. The mean concentration of $\sum_6$PBDE was 169 ng/g with BDE209 as the main congener and the mean concentration of $\sum_5$PBB was 38 ng/g. The results were lower compared to reported office studies in developed countries (Kefeni and Okonkwo, 2012).

Olukunle et al. (2015a) collected pooled dust samples from 11 offices and 10 houses in Nigeria in 2012. BDE209 was the dominant congener having mean concentrations of 180 ng/g in office dust and 141 ng/g in house dust, the obtained values were higher compared to South Africa. Indoor dust samples were also collected from cars in four states in Nigeria in 2014 and the $\sum_7$PBDE ranged from 159 to 735 ng/g with BDE209 as major congener (Olukunle et al., 2015b). The levels found were comparable to other studies done in the UK and USA.

Abafe and Martincigh (2015) investigated PBDEs in the indoor dust of computer laboratories, homes and offices in 2012, the $\sum_8$PBDE ranged from 818 to 1710 ng/g, BDE153 being the dominant congener in the computer laboratories while BDE209 being a major congener in the homes and offices (Abafe and Martincigh, 2015). Abafe & Martincigh (2016) also collected indoor dust from an electronic repair workshop and two e-waste dismantling and recycling facilities. The $\sum_8$PBDE ranged from 2632 to 44203 ng/g with BDE99 and 209 as the main congeners with levels higher than those reported in Vietnam and Guiyu, China, but lower than concentrations reported in Thailand and Southern China (Abafe and Martincigh, 2016).

Hassan (2015), analysed PBDE and NBFR in car dust, houses and workplace in Egypt. The study reported mean concentrations of $\sum_{14}$PBDE ranging from 159 to 36,927 ng/g in cars manufactured from 1999 to 2012, 2.2–591 ng/g in houses and 26–72,096 ng/g in workplaces (Hassan, 2015). $\sum$HBCDD was the dominant NBFR found in the three matrices analysed, with mean concentrations ranging from 20.7 to 47.7 ng/g (Hassan, 2015). The concentrations of PBDE and HBCDD reported were relatively low compared to those in developed countries.

### 6.5. Landfills

The leaching process of additives in plastic wastes, etc in landfills is difficultto understand. Due to their hydrophobicity, PBDEs are not completely dissolved and their leachability could be influenced by other constituents present in the leachate. (Osako et al., 2004).

Leachate samples were also collected from five landfill sites by Odusanya et al. (2009), reporting the mean concentrations for $\sum_{13}$PBDEs (excluding BDE209) ranging from 8392 to 54,761 pg/L with BDE47 as the major congener in three of the landfill sites. The obtained levels were higher than those reported for landfill sites in Japan and the USA (Odusanya et al., 2009).

Daso et al. (2013) collected leachate samples between April 2010 and March 2011 from three landfills in South Africa. The mean concentration of $\sum_8$PBDEs ranged from 0.2 to 2240 ng/L where BDE209 was reported as the dominant congener and BB153 concentrations ranged from 7.14 to 70.4 ng/L. The BDE concentrations increased with increasing rainfall frequency, and the levels were comparable to those found in Minnesota, USA but higher than those obtained in Osaka, Japan and Pretoria, South Africa. (Daso et al., 2013).

Olukunle and Okonkwo (2015) and Olukunle et al., (2015c) analysed NBFRs, HBCDD and PBDEs in leachates and sediment from six landfill sites between June and August 2013 with concentrations of NBFRs ranging from 8.7 to 142 pg/L for EHTBB, 4.5–15 pg/L for BTBPE and 4.8–40 pg/L for $\sum$HBCDD. The concentrations of $\sum_7$PBDEs ranged from 127 to 3703 pg/L for the leachates, and 0.8–8.4 ng/g dw for the sediments with BDE209 as a

dominant congener (Olukunle et al., 2015c). The HBCDD levels were significantly higher than studies conducted on river sediments in Netherlands and Ireland. The NBFRs and PBDE levels were lower compared to other studies done in South Africa on sediments as well as other developed countries.

### 6.6. Soil

Sun et al. (2016) reported levels of PBDE in soil samples collected in January 2015 from the rural part of Kenya. The mean $\sum_7$PBDE concentrations ranged from 2.54 to 13.65 ng/g dw from the three rural areas. The reported values were higher than those in China, Pakistan, UK and Norway but lower than that in Turkey (Sun et al., 2016). Surface soil samples were also collected in Arusha district, Tanzania in February 2011 at the final period of the dry season (Parolini et al., 2013). The $\sum_{13}$PBDE ranged from 136.4 to 952.2 pg/g dw which were lower than Italian Central Alps and most of the European background soils but higher than Russian Arctic, the central, southwestern and the east edge of the Tibetan Plateau (Parolini et al., 2013). Both studies found BDE47 as the major congener (Parolini et al., 2013; Sun et al., 2016).

### 6.7. Atmosphere

Pozo et al. (2008) and Lee et al. (2016) reported concentrations of POPs including PBDEs and HBCDD respectively from the first full year of the Global Atmospheric Passive Sampling (GAPS) Network in 2005 for three sites in Africa. The sites investigated included one agricultural site in Accra, Ghana and two background sites in De Aar and Kalahari in South Africa. The PBDE values obtained were below detection limit (Pozo et al., 2008), while the HBCDD concentrations ranged from <0.1 to 2.7 pg/m$^3$. The HBCDD values were comparable with the values from east Africa (Lee et al., 2016).

Shunthirasingham et al. (2010) investigated PBDE levels in Botswana from July 2006–August 2007 and reported the $\sum$7PBDE of the gas phase ranging from 24 to 59 pg/m$^3$ with BDE99, 28 49/71 and 209 as dominant congeners. The particulate PBDEs were reported to be very low ranging from non – detectable levels to 2.8 pg/m$^3$. The reported concentrations of PBDEs were higher in Botswana as compared to Costa Rica which was also investigated at the same time. Furthermore, the levels obtained were similar to measured concentrations in some European countries and background sites in North America but higher than levels measured at some sites around the Great Lakes in the Eastern Mediterranean and above the open Indian Ocean (Shunthirasingham et al., 2010).

Arinaitwe et al. (2014) also reported levels of PBDEs and alternative/novel flame retardants collected from October 2008–July 2010 in air and precipitation samples from the Lake Victoria shore. The study reported that BDE28, 47, 99 and 209 were the major congeners in the air samples with arithmetic mean concentrations

**Table 4**
BFR Studies conducted in the African continent in different matrices.

| Region | Country | Matrix | Reference |
|---|---|---|---|
| Eastern Africa | Uganda | Air and precipitation | (Arinaitwe et al., 2014) |
| | | Fish and sediments | (Ssebugere et al., 2014) (Ssebugere, 2015) |
| | Tanzania | Soil | (Parolini et al., 2013) |
| | | Aquatic ecosystem | (Polder et al., 2014) (Hellar-Kihampa et al., 2013) |
| | | Chicken eggs | (Polder et al., 2016) |
| | | Breast milk | (Müller et al., 2016) |
| | Kenya | Soil | (Sun et al., 2016) |
| | Ethiopia | Aquatic ecosystem | (Yohannes, 2014) (Yohannes et al., 2013) |
| Central Africa | Democratic Republic of Congo | Sediments, Biota | (Verhaert et al., 2013) (Mwanamoki et al., 2014) |
| | Gabon | Leatherback turtles | (Stewart et al., 2011) |
| West Africa | Ghana | Atmosphere | (Pozo et al., 2008) |
| | | Fish | (Asante et al., 2013) (Kuranchie-Mensah et al., 2013) |
| | | Breast milk | (Asante et al., 2011) |
| | | Cow milk | (Asante et al., 2010) |
| | | e-waste open burning soil | (Fujimori et al., 2016) |
| | | Soil | (Tue et al., 2016) |
| | Guinea-Bissau | Human exposure (Serum) | (Linderholm et al., 2010) |
| | Nigeria | sediments | (Adewuyi and Adeleye 2013) |
| | | car dust | (Olukunle et al., 2015b; Harrad et al., 2016) |
| | | indoor dust | (Olukunle et al., 2015a; Harrad et al., 2016) |
| | | plastic | (Sindiku et al., 2014) |
| | Senegal | Sediments and Molluscs | (Bodin et al., 2011) |
| | | Aquatic ecosystem | (Ndiaye et al., 2012; Sun et al., 2016) |
| | Togo | Vegetable samples | (Kolani et al., 2016) |
| North Africa | Egypt | indoor dust | (Hassan, 2015) |
| | | soil and sediments | (Abdallah et al., 2013) |
| | Tunisia | breast milk | (Hassine et al., 2012) |
| | | sediments | (Nouira et al., 2013) |
| | | fish | (Ameur et al., 2013; Ameur et al., 2011) |
| Southern Africa | South Africa | Atmosphere | (Pozo et al., 2008) |
| | | water and sediments | (La Guardia et al., 2013; Olukunle et al., 2012) |
| | | leachetes from selected landfills | (Odusanya et al.,2009; Olukunle and Okonkwo, 2015; Olukunle et al., 2015c; Daso et al., 2013) |
| | | indoor air of some selected sites; | (Abafe and Martincigh, 2014; 2016; Kefeni and Okonkwo, 2012; 2014) |
| | | sewage sludge and wastewater effluent | (Daso, et al., 2012; 2015a); Olukunle and Okonkwo, 2015) |
| | | bird eggs | (Polder et al., 2008; Daso et al., 2015b) (Quinn, 2010) |
| | | human breast milk | (Darnerud et al., 2011) |
| | Botswana | aquatic ecosystem | (Mbongwe et al., 2003) |
| | | atmosphere | (Shunthirasingham et al., 2010) |
| | Mauritius | Bird eggs | (Bouwman et al., 2012) |

of 2.9, 9.84, 4.83, and 8.27 pg/m$^3$ respectively and BDE209 showed the highest range (<1.57–168 pg/m$^3$). The values obtained were higher than those found in the European Arctic and the United Kingdom but lower than the measurements of the Atlantic coast of Africa, the urban centres in Chicago and Cleveland, Ohio as well as China. The study reported the most dominant detected AFR in the air samples was BTBPE with an arithmetic mean of 4.62 pg/m$^3$ (Arinaitwe et al., 2014). The precipitation samples identified BDE47, 99, 153 and 209 as the dominant congeners with average fluxes of 3.40, 6.23, 1.02 and 7.82 ng/m$^3$ respectively.

The sites investigated included one agricultural site in Accra, Ghana and two background sites in De Aar and Kalahari in South Africa.

Table 4 summarizes the BFRs studies that have been done in the African continent. These studies indicate that BFRs are indeed present in the environment and humans in the African continent and that there is a genuine lack of information about BFRs sources and releases. Nevertheless, current data from these countries indicate concentration of POPs equal or higher than those in temperate or cold regions. This may be due to the fact that products containing these materials are being imported by these countries (Olukunle et al., 2012). Owing to the persistence of BFRs and their potential for long-distance transport through the atmosphere, it is likely that they can potentially affect the developing as well as developed countries.

Table 4 also shows that so far, 3 studies have determined the atmospheric BFRs specifically PBDEs concentration levels, yet air has been suggested as a very useful matrix for regional and global monitoring studies, because it responds rapidly to changes in primary emissions of POPs (Harrad, 2009). Therefore, there is a huge information gap in the African continent which needs to be addressed.

## 7. Research gaps on BFRs in the African continent

Africa is one of the least studied continents in the World with respect to air quality (Laakso et al., 2007), nevertheless it is the second largest continent in the world covering over 30 million km$^2$, including over 54 countries and a population of about 1.17 Billion people (UNPD, 2015). Although only two studies have been conducted on atmospheric BFRs in the African continent. There is no information on atmospheric BFRs in expected high emission areas. Pozo et al. (2008) and Lee et al. (2016) placed passive air samplers in two background sites in South Africa, and one agricultural site in Ghana, Shunthirasingham et al. (2010) placed High – Volume (HiVol) Air samplers in a wetland surrounded by desert in Botswana while Arinaitwe et al. (2014) placed HiVol Air samplers close to the Lake Victoria Shore. Therefore all these studies did not monitor trends of BFRs related to higher risk factors such as landfill sites or residential areas where it is expected to have significant levels of BFRs in the atmosphere. Moreover these studies did not take into account the industrial activities taking place in the area making it difficult to predict whether the obtained concentrations of BFRs found were either from other locations or the specific areas where the studies where done. Fig. 3 shows different sources and conditions that need to be researched with respect to BFRs in the African continent. A good understanding of these sources and factors in the continent can help address the global perspective of these compounds in the atmosphere.

## 8. Conclusion and way forward

From the few studies that have been done in Africa, presenting the levels of BFRs in the environment, it can be concluded that there is a huge gap in the atmospheric concentration of these pollutants in the African continent.

Further research can assist in addressing the concentrations of BFRs present in the remote locations i.e. the Polar Regions. Many studies have just assumed that Africa was among the sources, but none of the studies have come up with the actual levels present in the region to justify it as a major source especially in the case of BFRs.

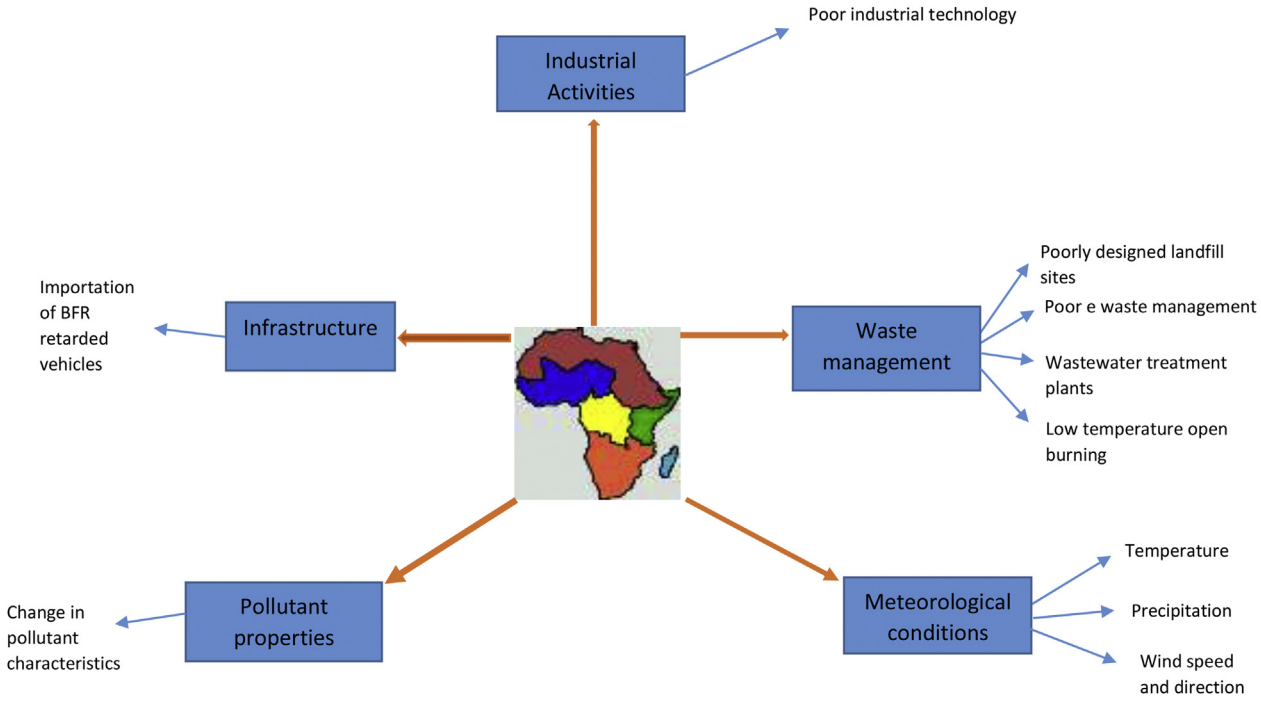


**Fig. 3.** Different sources of BFRs and factors that need to be researched in the African continent.

Most countries in the African continent experience higher temperatures. The effect of temperature and emission of persistent organic pollutants has been observed to be very important. Increasing temperature will directly lead to enhanced emissions of POPs that volatilize from their sources (Lamon et al., 2009).

Higher temperatures also affect partitioning both between bulk phase (air vs surface media i.e. soil, water and vegetation) and between the gaseous particle-bound phases in air. Hence, stronger emissions from primary and secondary sources and a shift towards higher gaseous fractions in the air may be observed. As a result, the additive BFRs will be more available for long-range transport (UNEP/AMAP, 2011). It would therefore be necessary for new research to monitor the air as well as analyse soil samples from the same location to observe the partitioning effect.

Some studies have also indicated that during thermal stress, PBDEs and TBBP-A are converted to the dioxin-like compounds, polybrominated dibenzodioxins (PBDDs) and polybrominated dibenzofurans (PBDFs) (Janssen, 2005). The continent's difference in meteorological conditions might also result in upcoming alternative/novel BFRs (NBFRs). Thus there is also a need to research on these NBFRs, because there is limited information on the atmospheric NBFRs in Africa.

Therefore, monitoring air emission of BFRs in the continent as well as evaluating their elution over longer time periods could be an important foundation in addressing the mentioned gaps.

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