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Environmental occurrence and biota concentration of phthalate esters in Epe and Lagos Lagoons, Nigeria

Aina O. Adeogun [a], Oju R. Ibor [a], Emmanuel D. Omogbemi [a], Azubuike V. Chukwuka [a], Rachel A. Adegbola [c], Gregory A. Adewuyi [b], Augustine Arukwe [d, *]

[a] Department of Zoology, University of Ibadan, Ibadan, Nigeria
[b] Department of Chemistry, University of Ibadan, Ibadan, Nigeria
[c] Department of Chemistry, The Polytechnic, Ibadan, Nigeria
[d] Department of Biology, Norwegian University of Science and Technology (NTNU), Høgskoleringen 5, N-7491 Trondheim, Norway

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ABSTRACT

The high global occurrence of phthalates in different environmental matrixes has resulted in the detection of their metabolites in human urine, blood, and breast milk, indicating a widespread human exposure. In addition, the notorious endocrine disrupting effects of phthalates have shown that they mimic or antagonize the action of endogenous hormones, consequently producing adverse effects on reproduction, growth and development. Herein, we have studied the occurrence of phthalate esters (PEs) in water, sediment and biota of two lagoons (Epe and Lagos) in Nigeria. Two fish species (*Tilapia guineensis*, and *Chrysichthys nigrodigitatus*) and a crustacean (the African river prawn - *Macrobrachium vollenhovenii*) were analyzed for PEs levels using a HPLC method and the derived values were used for calculating bioconcentration factor (BCF), biota-sediment accumulation factor (BSAF) and phthalate pollution index (PPI) in the biota and environment. We observed that the growth and health condition of the fish species were normal with a k-factor of >1. Sediment PE levels were compared with water, at both lagoons showing concentration pattern that is characterized as DEHP = DEP > DBP. We observed that DBP was the predominant compound in *T. guineensis*, *C. nigrodigitatus* and African prawn, at both lagoons, showing organ-specific differences in bioconcentration (BCF and BSAF) patterns in the fish species. While there were no observed consistency in the pattern of PE concentration in fish organs, elevated DBP levels in different fish organs may be related to fish habitat and degradation level of phthalates. Low concentration of DEHP, compared with DBP and DEP, was measured in fish organs and whole prawn body. The BSAF values for DEHP were lowest, and highest for DBP for all species at both lagoons, and DEHP easily accumulated more in the sediment (sediment PPI = 0.28 and 0.16 for Epe and Lagos lagoon, respectively). Overall, our findings suggest a broader environmental and human health implication of the high PE levels in these lagoons since they represent significant sources of aquatic food resources for the neighboring communities.

1. Introduction

Phthalic acid esters (phthalates) are ubiquitous environmental contaminants worldwide, due to their broad usage in flexible plastics and consumer products (Zeng et al., 2008b; Dominguez-Morueco et al., 2014). Phthalates are used to enhance the plasticity of industrial polymers, resulting in their widespread usage in a broad range of products that include – food packaging, toys, paints or internal polyvinyl chloride (PVC) coatings, construction materials, personal care products and cosmetics (such as nail varnish), electronic and medical devices and pediatric articles (such as bags for intravenous fluids, breathing masks or umbilical catheters), among several others products (Berman et al., 2009; Calafat et al., 2004; Kimber and Dearman, 2010; Yan et al., 2009; Dominguez-Morueco et al., 2014). In addition, the widespread application and use of phthalates by the general population, has also resulted in their notorious presence as solid waste products of industrial and domestic activities (Kimber and Dearman, 2010).

* Corresponding author.
E-mail address: arukwe@bio.ntnu.no (A. Arukwe).

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In the European Union (EU), an effective ban was placed on the use of six phthalates – namely: (di-(2-ethylhexyl) phthalate (DEHP), butylbenzyl phthalate (BBP), di-n-butyl phthalate (DBP), di-isononyl phthalate (DINP), di-isodecyl phthalate (DIDP) and din-octyl phthalate (DNOP)) in the production and sale of toys and infant articles that could be introduced into the mouth of children younger than three years old (Directive, 2005/84/EC; European Parliament and the Council, 2013).

The environmental increase and distribution of phthalates has generated strong societal concerns since they are shown to cause cancer, developmental abnormalities and reproductive effects that include reduction in sperm counts in males (Su et al., 2012). Furthermore, due to the high global occurrence of phthalates in different environmental matrices, their metabolites have been detected in human urine, blood, and breast milk, indicating a widespread human exposure (Guo et al., 2012; Hines et al., 2011). Phthalates have been reported to possess overt endocrine disrupting properties, and shown to mimic or antagonize the action of endogenous hormones, which consequently result in adverse effects on reproduction, growth and development (Crisp et al., 1998; Crocker et al., 1983; Fisher, 2004). Although endocrine mode of action is not well understood for phthalates, it may likely be dependent on developmental timing and dosing regimes (Akingbemi et al., 2001). Studies on rodents and avian species have identified Leydig cells as one of the main targets of phthalate-induced reproductive toxicity (Akingbemi et al., 2004; Bello et al., 2014). In humans, exposure to phthalates may be associated with adverse health effects that include, but not limited to neuro-developmental problems (Whyatt et al., 2012), low semen quality (Huang et al., 2011), miscarriages (Toft et al., 2012), asthma and allergies (Hsu et al., 2012), breast cancer progression (Hsieh et al., 2012) and obesity (Hatch et al., 2010), most likely by targeting the peroxisome proliferator (PP) pathways.

From a developing country perspective, the increasing consumption of consumer products in Nigeria, consequently generates increased amount of solid waste per capita, with associated high proportion of organic material, compared to developed countries (Arukwe et al., 2012; Visvanathan and Glawe, 2006). Recently, we reported high concentrations of DEHP and DBP in leachates and sediment from a municipal waste deposit site at Owerri, Nigeria, whose levels in the sediments were up to 1000 times higher than the run-off water sample from the same sites (Arukwe et al., 2012) and high environmental and biota levels of phthalates from two municipal water supply lakes in Ibadan, Southwestern Nigeria (Adeogun et al., submitted). A similar pattern was previously reported in several other rivers, showing higher levels of DEHP and DBP and lower levels of DMP and DEP (Yuan et al., 2002). The Epe and Lagos lagoons are part of the Southern lagoon system in Nigeria and are important for foraging and breeding of fish species. These lagoons contribute substantial portions of wild type artisanal fish production in Nigeria and serve as fishing habitats as well as nursery, feeding and spawning grounds for a diverse number of fish species. In addition, Lagos lagoon is also a major seaport in Nigeria. The high level of waste materials discharged into the Lagos lagoon has progressively polluted the shores of the lagoon (Eruola et al., 2011). As a result, several locations along the Lagoon are currently facing an increasing number of serious environmental and ecological challenges. Most of these challenges are direct results of urbanization and high commercial activities (including, but not limited to seaport activities) along the axis of the Lagoon, contributing to a massive deterioration of water quality and contaminant load with overt regional consequences on the aquatic ecosystem and on the human health of the user groups (Eruola et al., 2011).

The interplay between urbanization, commercial activities and ecological/human health effects in the Lagos lagoon system is of societal concern, and calls for an urgent management and sustainable use of the lagoon. The level of contaminant load to the lagoon is highest at the harbor area with a decreasing trend towards the metropolitan end of the lagoon (Eruola et al., 2011). In addition, the Epe area of the lagoon is putatively, the least influenced by anthropogenic contamination. However, the degree of contaminant load in the lagoon varies, even within the different areas (Eruola et al., 2011). Therefore, a comprehensive and detailed contaminant monitoring process is urgently needed in order to discern the actual environmental contaminant load and biological consequences. Therefore, the aim of the present study was to investigate the levels of phthalic acid esters in environmental (water and sediment) and biota samples of the two lagoon systems (Epe and Lagos) in Nigeria.

2. Materials and methods

2.1. Sampling sites

The Epe lagoon is located between longitudes 5°30′–5°40′E and latitudes 3°50′–4°10′N. The lagoon receives River Osun that drains a number of cities and agricultural lands (Fig. 1A). The study area is bordered on the west by a number of cultivated lands and receives wood wastes from local wood processing outfits located at the bank of the lagoon. The lagoon is used for transportation of timber logs (possible source of wood particles and leachates) from the villages to the city of Lagos and is the second largest contributor to the viable commercial artisanal fisheries of the southern lagoon complex. The lagoon houses a major jetty at Epe, where different forms of anthropogenic wastes within and around the jetty are indiscriminately deposited (Edokpayi et al., 2010).

The Lagos lagoon is the largest of four lagoon systems in the Gulf of Guinea (Webb, 1958). It stretches for about 250 km from Cotonou in the Republic of Benin to the western edge of the Niger delta. The lagoon includes the forest belt and receives a number of important large rivers such as Yewa, Ogun, Ona and Osun rivers, draining more than 103626 km^2 of the country and empties into the Atlantic Ocean (Fig. 1B). The Lagos opening is by far the largest and forms an extensive harbor, which serve as the major outlet of fresh water from the lagoon system during the rainy season. The central body of the lagoon is located between longitude 3° 23′ and 3° 40′E and latitude 6° 22′ and 6° 38′N. This brackish region is of interest for coastal dynamics and transport of pollutants from the hinterland and the immediate shores of the lagoon (Ajao and Fagade, 1990). The lagoon provides places of abode and recreation, means of livelihood and transport, a dumpsite for residential and industrial discharge and a natural shock absorber to balance forces within the natural ecological system. About 80–85% of the industries in Nigeria are located in Lagos State and they all discharge their effluents into the Lagos lagoon. The effluents discharged are mainly untreated, and very few industries have treatment plants in Nigeria (IPEP, 2006).

2.2. Chemicals and reagents

Acetonitrile, dichloromethane, sodium carbonate, anhydrous sodium sulfate and aluminium oxide, N-hexane and ethyl acetate were of HPLC grade, di-2- ethylhexylphthalate (DEHP), diethylphthalate (DEP) and dibutylphthalate (DBP) standards were purchased from Sigma–Aldrich (Switzerland).

2.3. Sample collection

Biota samples (n = 150/species) of *Chrysicthys nigrodigitatus*, *Tilapia guineensis* and the invertebrate *Macrobrachium*