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Occurrence and dietary exposure to organophosphorus esters in foodstuffs of Nigerian origin

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ABSTRACT

Despite extensive evidence of the presence of organophosphate esters (OPEs) in various environmental matrices including foodstuffs in some developed countries, there are no data on the presence of OPEs in African foodstuffs. This study provides the first data on concentrations of OPEs in foods of Nigerian origin by measuring the concentrations of eight OPEs in 85 food items either purchased in Nigeria or from major African stores in Birmingham, UK. Foodstuffs analysed were divided into ten categories. Median concentrations of $\sum_8$OPEs in melon (Egusi) (17.4 ng/g wet weight (ww)), cassava flour (Fufu) (11.6 ng/g ww), and milk (9.62 ng/g ww) were the highest of the food categories examined. Tris(2-butoxyethyl) phosphate (TBOEP) and triphenyl phosphate (TPHP) were the dominant OPEs. Cassava flour (Fufu) was the major contributor to the estimated daily intakes (EDIs) of Σ_8OPEs, accounting for 26 and 33% of the EDI for children and adults respectively. For individual OPEs, TBOEP contributed the highest median EDI of 183 ng/kg bw/day (42% of the EDI for Σ_8OPEs) for children and 159 ng/kg bw/day (39%) for adults, followed by TPHP (median – children: 92.3 ng/kg bw/day (21%); adults: 90.6 ng/kg bw/day (22%)). The range of OPE concentrations in the present study were comparable to data reported from other countries and EDIs for all OPEs were well below their corresponding health-based reference doses. This study identifies diet as a substantial human exposure pathway to OPEs in Africa.

1. Introduction

Organophosphate esters (OPEs) are chemical additives integrated into combustible materials and other consumer products to reduce the spread/risk of fire, meet flammability standards, and/or modify the properties of the material to which they are added (Castro et al., 2020; Van der Veen & De Boer, 2012). Their extensive use as flame retardants, stabilisers, anti-foaming agents, or plasticisers in various consumer and industrial products such as: flexible polyurethane foam (PUF), electronics, furniture, plastics, construction materials, floor polish, and paints has led some OPEs to be catalogued as high production volume chemicals (Castro et al., 2020). In 2017, the global consumption of phosphorus-based FRs reached 797,234 tons, accounting for more than 30% of the total global consumption of FRs, compared to 20% for bromine-based FRs (McWilliams, 2018; Wang et al., 2019). Due in part to the regulation of brominated flame retardants such as polybrominated diphenyl ethers and hexabromocyclododecane, production and usage of OPEs has increased and led to their ubiquitous presence and detection in various environmental matrices and human samples (Gbadamosi et al., 2021; Hou et al., 2020; Wang et al., 2019). Pertinently, OPEs are physically added into polymeric materials and not chemically bonded (Zhao et al., 2020); this allows their facile release and pollution of the environment as a result of volatilisation, abrasion, and leaching (Brommer & Harrad, 2015; Hou et al., 2020; Wei et al., 2015). Moreover, several toxicological, and epidemiological studies have reported serious adverse effects of OPEs that include: carcinogenicity, cardiotoxicity, neurotoxicity, genotoxicity, reproductive disorders, and dermatitis (McGee et al., 2013; Wang et al., 2019; Zhao et al., 2021). Based on the potential exposure health risks of OPEs in various products, the European Union (EU) in 2008 set regulations and criteria for the hazard classification and labelling of certain OPEs (Regulation (EC) No 12/72/2008). According to the EU's approved harmonised classification and labelling, many OPEs pose major health and/or environmental hazards (Chen et al., 2021a). Consequently, while over

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100 countries regulate the use of certain OPEs (Blum et al., 2019; Chen et al., 2021b), there is to our knowledge no regulation on the production and use of OPEs in Africa. This is of concern, given that because of the ubiquitous presence of OPEs in various environmental compartments, and their possible accumulation in biota and other fatty tissues; humans can be exposed via: ingestion of dust, water, and food, inhalation of air, and dermal absorption (Abdallah & Covaci, 2014; Al-Omran et al., 2021; Gbadamosi et al., 2021; Lee et al., 2016; Poma et al., 2017, 2018; Wang & Kannan, 2018). Contamination of foodstuffs with OPEs can occur through biomagnification in the food chain via crops, aquatic organisms/seafood, and livestock through their feed and absorption from soil and water (Zhang et al., 2016), as well as directly via migration from food packaging materials (Wang & Kannan, 2018) and contamination from food processing and storage (Ding et al., 2018; Gbadamosi et al., 2021; Poma et al., 2018). Consequently, it has been established that dietary intake is one of the main human exposure pathways to OPEs for the general population (Gbadamosi et al., 2021; Li et al., 2019a; Wang et al., 2022). However, information on the occurrence of OPEs in foodstuffs is limited to countries such as: Australia (He et al., 2018), China (Chen et al., 2022; Ding et al., 2018; He et al., 2019; Liu et al., 2019; Wang et al., 2022; Zhang et al., 2016; Zhao et al., 2019; Zhou et al., 2022), the USA (Han et al., 2019; Wang & Kannan, 2018), Sweden and Belgium (Poma et al., 2017, 2018), the United Kingdom (Gbadamosi et al., 2022), Canada (McGoldrick et al., 2014), and the Philippines (Xu et al., 2015). While the OPEs measured varies between studies, the reported median concentration of ∑OPEs in these studies varies from 0.08 to 50 ng/g wet weight (ww) in the various foodstuffs analysed. Out of all studies of which the authors are aware, the highest median concentrations of ∑OPEs were found in rice (7.59–55.9 ng/g) from four provinces in China (Zhang et al., 2016), and in fat and stock (median: 63.5 and 16.7 ng/g ww) in Belgium (Poma et al., 2018). The highest concentrations of individual OPEs reported in all the foodstuffs from these studies were TDCIPP and TCIPP in fat (16.1 ng/g and 31.0 ng/g ww) (Poma et al., 2018) and EHDPP in stock and desserts (12.3 and 8.72 ng/g ww) from Belgium (Poma et al., 2018). In studies in the USA, China, Sweden, Belgium, and Australia, mean EDIs of ∑OPEs were in the range 1.9–601 ng/kg bw/day for the general population (Wang & Kannan, 2018; (Chen et al., 2021a; He et al., 2018; Poma et al., 2017; 2018; Zhang et al., 2016; Zhao et al., 2019). However, current data and understanding on the occurrence of OPEs in foodstuffs and dietary exposure and consequent health risk is far from sufficient especially given the absence of data for Africa. Therefore the aims of the current study are: (i) to report for the first time the concentrations and distribution of eight OPEs in a wide range of Nigerian foodstuffs collected directly from local markets in Nigeria along with some collected from major African stores in Birmingham, UK that could be identified as originating from Nigeria; (ii) to investigate the likely contamination sources of OPEs in Nigerian foodstuffs; and (iii) to estimate human dietary exposure to OPEs to individuals consuming foods of Nigerian origin under different exposure scenarios.

2. Materials and methods

2.1. Sample collection

Between June–August 2021, a total of 85 food samples were collected directly from local markets in Lagos, Nigeria (n = 21) and from three major African stores in Birmingham, UK (n = 64). The origin of the samples collected in the UK was established via information provided on product labelling and/or via confirmation from the store owners that the foods were from Nigeria. Collected food samples were stratified into 12 categories: cereal (rice and noodles) (n = 10), dried and smoked fish (n = 26), roasted beef (Suya and Kilishi), cow hide meat (ponmo) (n = 23), milk (n = 6), fried plantain snacks (n = 3), wheat flour (Semovita) (n = 3), yam powder (Poundo yam) (n = 3), cooked cassava flour dough (Fufu) (n = 3), cassava flakes (Garri) (n = 5), and Egusi (melon) (n = 3) (Table S3). Some food samples such as Kilishi and Fufu were purchased in the processed and ready to eat form. Samples collected in Nigeria were collected directly from the local vendors, carefully packaged in aluminium foil, placed in a sealed zip lock bag, and shipped to the laboratory at the University of Birmingham for chemical analysis. Those collected from African stores in Birmingham were taken directly to the lab after collection. On arriving at the laboratory, edible parts of solid foods were freeze-dried and individually homogenised, then stored at −20 °C prior to extraction and analysis. Further information about the foodstuffs analysed are presented in supplementary information (SI) (Table S3).

2.2. Chemicals and reagents

Standards of the 8 OPEs measured in this study namely: tris(2-chloroethyl) phosphate (TCEP), tris(2-chloroisopropyl) phosphate (TCIPP), tris(1,3-dichloro-2-propyl) phosphate (TDCIPP), tris(2-butoxyethyl) phosphate (TBOEP), triphenyl phosphate (TPHP), 2-ethylhexyl-diphenyl phosphate (EHDPP), tri-*n*-butyl phosphate (TnBP), and tri-m-tolyl phosphate (TMTP), internal (surrogate) standards (ISs) d_{27}-TnBP and d_{15}-TPHP, and 2,3,4,6-tetrachlorobiphenyl (PCB-62) used as a recovery determination (syringe) standard (RDS) were purchased from Wellington laboratories, (Guelph, ON, Canada). Hypersep Florisil® cartridges were purchased from Thermo Scientific (Rockwood, USA. All glassware, as well as the knife and weighing spoon used for sample preparation, along with Na_2SO_4 were baked at 450 °C and 600 °C overnight and rinsed before each use with dichloromethane (three times), toluene, and acetonitrile (ACN) to minimise residual organic contamination.

2.3. Analytical procedures

Concentrations of target OPEs in the foodstuffs were determined via GC-MS using a previously reported method (Gbadamosi et al., 2022, 2023). Briefly, for most samples ~1 g of each freeze-dried sample was accurately weighed in a 15 mL polypropylene (PP) tube - with smaller aliquots (ca 0.5–0.8 g) used for high fat matrices (i.e. >20% lipid) to minimise matrix interferences. After adding 5 mL of ACN and 5 mL of 5% formic acid in ACN (v/v), each sample was spiked with 50 ng of both ISs and extracted via vortexing (1 min), sonication (15 min), and centrifugation (3500 rpm for 5 min) in three cycles. The pooled supernatant was then transferred into a precleaned glass tube and evaporated to ~2 mL under a gentle nitrogen stream. This concentrated extract was subjected to dispersive solid phase extraction via addition of 100 mg of C18 and 50 mg of primary-secondary amine sorbent powder to remove fats, sugars, and other polar pigments that may act as interferences, followed by vortexing for 1 min and centrifugation for 5 min at 3500 rpm. The resultant supernatant was collected into a clean glass tube and evaporated to incipient dryness, resolubilised in 0.5 mL of n-hexane: ethyl acetate (ETAC) (1:1 v/v) and further cleaned by elution through a Florisil® cartridge preconditioned with 6 mL ETAC and 6 mL n-hexane. Fractionation was achieved with 12 mL of n-Hexane (F1, discarded) and 10 mL ETAC (F2, containing the target OPEs). F2 was concentrated to near dryness under a gentle flow of nitrogen, and reconstituted in 200 μL iso-octane:ETAC (8:2 v/v) containing 100 pg μL^{-1} PCB-62 as RDS. The resultant cleaned extract was transferred into a clean glass tube and stored at −20 °C to allow precipitation of any residual lipids, after which the clean upper layer was transferred into an injection vial, and stored ready for analysis via gas chromatography electron ionisation mass spectrometry (GC-EIMS) in selected ion monitoring mode. One μL of purified extract was injected using cold split-less injection. Detailed information on the GC-EIMS method used is available elsewhere (Gbadamosi et al., 2022, 2023) but is also presented here as supplementary information, section 1, Table S2.

2.4. Quality assurance (QA) and quality control (QC)

Due to the lack of certified reference materials for food, aliquots of Na_2SO_4 (prebaked at 450 °C for 6 h) were used to assess blank contamination and method accuracy via analysis of aliquots spiked: (a) with ISs, and (b) with ISs and native OPE standards. In addition, the following matrix spike samples were analysed: five samples each of vegetable oil and egg samples (from the same grocery store) were spiked with ISs (50 ng of each) and each of the target OPEs (also 50 ng each) and analysed. Results from these matrix spike samples, showed good IS recoveries ranging from 62.3 ± 16.9 to 79.4 ± 7.3% for d_{15}-TPHP and d_{12}-TBP and the average recoveries of the native OPEs varies from 73.1 to 99.5% (RSD = 4.6–24.8%) and 71.4–99.6% (RSD = 8.8–24.7%) for vegetable oil and egg respectively (Table S4 and Table S5). Average recoveries of the ISs spiked into Na_2SO_4 ranged between 72 and 96% (Table S5). Two procedural blanks comprising 1 g Na_2SO_4 treated as a sample were analysed per each batch of 10 samples (n = 9). This revealed some low-level contamination (ranging between 0.02 and 0.07 ng/g dw) for TCEP only (Table S5). Other quality control measures followed in this research are reported in our previous study of UK foodstuffs (Gbadamosi et al., 2022) The limit of detection (LOD) and the limit of quantification (LOQ) were calculated as the amounts of an analyte that yielded a signal to noise ratio of 3 and 10 respectively (Table S5 and Table S6), while for TCEP (as the only compound detected in the blank), the LOD and LOQ was calculated as 3 and 10 times the standard deviation of the blank values.

2.5. Data analysis

Descriptive statistics, specifically: mean, median, maximum, minimum, standard deviation, and the 97.5th percentile were obtained using Microsoft 365 excel, while the multivariate statistical tests were carried out using IBM SPSS statistics version 28 and GraphPad prism 9 (version 9.5.0). Our data were shown to be not normally distributed using Shapiro-Wilk and Kolmogorov normality tests and visual inspection. As a result, nonparametric statistical tests such as Mann-Whitney *U* test and Kruskal-Wallis H test were employed used to test for differences in concentrations of OPEs in foodstuffs (P-values lower than 0.05 ($p < 0.05$) were deemed statistically significant). Moreover, bivariate correlations (Spearman's correlation analysis) were used to investigate associations between OPEs. The data were log10 transformed prior to analysis to permit use of component analysis. Following satisfactory outcomes from a Kaiser-Meyer Olkin (KMO) measurement of sampling adequacy and Bartlett's Test of sphericity (Table S7); principal component analysis (PCA) was employed (Gbadamosi et al., 2018). Combined with the Spearman's correlation analysis, PCA helped identify whether common sources exist of our target OPEs in foodstuff, and if so, for which OPEs. A one-way analysis of variance (ANOVA) was used to test for differences in concentrations of OPEs in foodstuffs (P-values lower than 0.05 ($p < 0.05$) were deemed statistically significant). Where concentrations of a given OPE in a sample were <LOQ, the concentration value was assigned as half the LOQ for that OPE (< LOQ = 0.5 x LOQ).

2.6. Estimated dietary intakes and risk assessment

Estimation of human dietary intake was undertaken using the formula (Gbadamosi et al., 2022; Zhao et al., 2019):

$$EDI = \frac{\sum_{i=1}^{n} C_i x\ CF_1}{BW} \quad (1)$$

Where EDI is the estimated dietary intake of a given OPE(s) (ng/kg bw/day), Ci is the median/97.5th percentile concentrations of OPEs in each food category (ng/g); CFi is the daily food consumption rate of the corresponding food category (g/day/person) obtained from previous studies in Nigeria (Ayanda et al., 2020; Giri et al., 2021; Hayford et al., 2009; Ijeoma et al., 2020; Nejabat et al., 2017; Orisakwe et al., 2015; Patrick-Iwuanyanwu & Udowelle, 2017; Sanusi et al., 2021) (Table S8). EDIs were calculated for children (4–18 yrs) and adults (19–64 yrs). BW is the average body weight (kg) of the population assumed to be 20 kg and 60 kg for children and Nigerian adults respectively (Brommer & Harrad, 2015; Charles et al., 2017; Walpole et al., 2012). In this study, we assumed that the population consumes all the food analysed in this study in quantities that correspond to those that a typical Nigerian consumes, even though some of the foods were bought from African stores in UK and some in Nigeria. The median and the 97.5th percentile concentrations of each foodstuff were multiplied by the median consumption rate data under the average and the high-exposure scenarios for the same foodstuffs for humans and are listed in Table S8. The high-end exposure data was derived by multiplying the median consumption rate data with the 97.5th percentile concentrations for each food category. This is due to the fact that only median consumption rate data are available for Nigeria (Table S8).

3. Results and discussion

3.1. Occurrence of OPEs in foodstuffs of nigerian origin

A statistical summary of the concentrations of OPEs in foodstuffs of Nigerian origin is presented in Table 1. Detection frequencies (DFs) of the chlorinated OPEs (TCEP, TCIPP, and TDCIPP) exceed those of our other target OPEs (DF = 67–100%) except in smoked and dried fish samples, where the DF for TDCIPP is 23%. DFs for the two aryl OPEs: TPHP and EHDPP each vary from 0 to 100%, with TMTP not detected in any food sample in this study (Table 1). The DFs for the alkyl-OPEs: TBOEP and TnBP, vary between 0 - 67% and 0–100% respectively (Table 1). This variation in the DFs of our target OPEs likely arises from a combination of factors, including whether the foodstuff is processed, and the use volumes, physicochemical properties, half-lives, degradation, and metabolism of the OPEs (He et al., 2018). Our frequent detection of the Cl-OPEs in food samples, confirms reports of high DFs of Cl-OPEs in various environmental compartments due to their wide application (Brandsma et al., 2014; Gbadamosi et al., 2022). Notwithstanding their frequent detection, Cl-OPEs were present in this study at relatively low concentrations in animal-based foods which may be because their transfer from feed is inefficient (Gbadamosi et al., 2022; Zhang et al., 2022).

The highest concentrations of $\sum_8$OPEs were obtained in melon (median: 17.4; range: 14.9–23.6 ng/g ww), followed by cassava flour (median: 11.6; range: 8.60–18.5 ng/g ww), milk (median: 9.62; range 5.68–18.0 ng/g ww), roasted meat and cow hide (median: 9.08; 1.30–47.1 ng/g ww); while the least contaminated food was dried and roasted fish (median: 3.90; range: 1.61–28.1 ng/g ww) (Table 1). The elevated OPE concentrations in melon may result from uptake of OPEs from soil, as Egusi melon belongs to the genus *cucurbitae*, and previous studies have shown that plants from this genus display unusually high propensity for root uptake from soil and translocation to fruits, of organic chemicals such as chlorinated dioxins (Huelster et al., 1994).

The values obtained in this study are comparable to those observed in our previous study of OPEs in UK foodstuffs (Gbadamosi et al., 2022). However, our detection of the highest Σ_8OPE concentrations in melon, contrasts with previous studies in the USA, China, Australia, Belgium, Sweden, and the UK, where the highest median concentrations of OPEs were reported in fish, rice/meat/cereals, vegetables, fats/oils, cereals and milk and milk products (Chen et al., 2021, Ding et al., 2018; Gbadamosi et al., 2022; He et al., 2018; Poma et al., 2017; 2018; Wang & Kannan, 2018; Zhang et al., 2016). Across all food samples, TBOEP was present at the highest median concentration (36.2 ng/g ww, 41% of Σ_8OPEs), followed by TPHP (20.0 ng/g ww, 22%), EHDPP (13.8 ng/g ww, 15%) with the lowest median concentration reported for TMTP

Table 1
Overall descriptive statistics of OPEs in foodstuffs of Nigerian origin (ng/g, ww).

Food category and # samples purchased in Nigeria and UK	Statistical parameter	TCEP	TCIPP	TDCIPP	EHDPP	TPHP	TnBP	TBOEP	TMTP	∑OPEs
Dried and smoked fish (n = 26)	Mean	0.48	0.49	0.27	1.40	2.38	<0.53	2.38	<0.16	8.09
Nigeria (n = 9), UK (n = 17)	Median	0.40	0.35	0.06	0.14	2.02	<0.53	0.24	<0.16	3.90
	Range	0.12–1.04	<0.13–1.35	<0.06–1.13	<0.14–7.70	<0.23–7.64	<0.53	<0.24–8.53	<0.16	1.61–28.1
	97.5th Percentile	1.02	1.13	1.09	7.54	6.79	<0.53	8.47	–	
	DF (100%)	100	92	23	27	85	0	31	0	
Roasted meat and cow hide (n = 23)	Mean	0.52	0.40	0.59	2.33	0.97	0.52	7.23	<0.18	12.7
Nigeria (n = 7), UK (n = 16)	Median	0.43	0.29	0.36	2.25	0.23	0.53	4.83	<0.18	9.10
	Range	0.12–1.25	0.11–1.00	<0.07–1.97	<0.14–8.71	<0.22–4.07	<0.53–0.57	<0.26–29.4	<0.18	1.63–47.1
	97.5th Percentile	1.19	0.86	1.94	7.39	3.68	0.53	25.7	–	
	DF (100%)	100	100	70	70	39	4	44	0	
Milk (Liquid and Powder) (n = 6)	Mean	0.18	0.34	0.88	<0.16	4.64	<0.60	3.62	<0.16	10.6
All in the UK	Median	0.18	0.29	0.75	<0.16	4.54	<0.60	3.03	<0.16	9.71
	Range	0.10–0.33	0.18–0.60	0.45–1.63	<0.16	3.87–5.40	<0.60	<0.27–9.20	<0.16	5.79–17.8
	97.5th Percentile	0.31	0.59	1.56	<0.16	5.39	<0.60	8.80	–	
	DF (100%)	100	100	100	0	100	0	50	0	
Cereal (n = 10)	Mean	0.19	0.31	0.66	2.92	3.39	0.31	0.55	<0.17	8.50
Nigeria (n = 5), UK (n = 5)	Median	0.18	0.25	0.42	3.01	3.06	0.33	0.24	<0.17	7.66
	Range	0.11–0.28	0.20–0.56	<0.06–2.12	<0.16–5.93	<0.24–7.03	<0.07–0.53	<0.26–2.13	<0.17	1.27–18.7
	97.5th Percentile	0.28	0.54	1.93	5.85	6.94	0.53	1.89	<0.17	18.1
	DF (100%)	100	100	83	67	83	50	17	0	
Wheat flour (n = 3)	Mean	0.28	0.56	1.21	5.04	<0.25	<0.58	<0.26	<0.17	8.35
All UK	Median	0.29	0.64	1.39	4.77	<0.25	<0.58	<0.26	<0.17	8.35
	Range	0.23–0.33	0.25–0.79	0.50–1.74	3.64–6.71	<0.25	<0.58	<0.26	<0.17	5.88–10.8
	97.5th Percentile	0.33	0.78	1.72	6.62	<0.25	<0.58	<0.26	<0.17	10.7
	DF (100%)	100	100	100	100	0	0	0	0	
Cassava flour (n = 3) (Fufu)	Mean	0.36	0.45	0.97	0.35	2.18	1.13	7.31	<0.21	12.9
All UK	Median	0.26	0.48	1.04	0.32	1.49	1.09	6.75	<0.21	11.6
	Range	0.21–0.61	0.38–0.49	0.76–1.11	<0.19–0.60	0.60–4.46	<0.72–1.76	5.82–9.35	<0.21	8.89–18.6
	97.5th Percentile	0.59	0.49	1.10	0.59	4.31	1.73	9.22	<0.21	18.2
	DF (100%)	100	100	100	67	100	67	100	0	
Yam powder (n = 3) (Poundo yam)	Mean	0.22	0.14	0.39	<0.16	<0.23	0.40	6.04	<0.16	7.74
All UK	Median	0.29	0.13	0.48	<0.16	<0.23	0.53	6.50	<0.16	8.48
	Range	0.04–0.34	<0.07–0.23	<0.06–0.61	<0.16	<0.23	<0.14–0.53	5.08–6.55	<0.16	5.94–8.81
	97.5th Percentile	0.34	0.23	0.61	<0.16	<0.23	0.53	6.55	<0.16	8.81
	DF (100%)	100	67	67	0	0	33	100	0	
Plantain snacks (n = 3)	Mean	0.41	0.29	0.90	<0.19	1.06	0.47	4.07	<0.21	7.60
All UK	Median	0.46	0.18	0.71	<0.19	1.19	0.53	5.46	<0.21	8.93
	Range	0.27–0.51	0.17–0.51	0.59–1.39	<0.19	<0.30–1.76	<0.16–0.53	<0.32–6.49	<0.21	2.21–11.6
	97.5th Percentile	0.50	0.50	1.35	<0.19	1.73	0.53	6.44	<0.21	11.5
	DF (100%)	100	100	100	0	67	33	67	0	
Cassava flakes (Garri) (n = 5)	Mean	0.22	0.24	0.66	2.21	2.98	<0.53	1.01	<0.16	8.01
All UK	Median	0.21	0.13	0.67	0.14	2.50	<0.53	0.24	<0.16	4.58
	Range	0.13–0.32	<0.13–0.47	0.64–0.68	<0.15–6.34	2.36–4.08	<0.53	<0.25–2.54	<0.16	4.35–15.1
	97.5th Percentile	0.32	0.46	0.68	6.03	4.00	<0.53	2.42	<0.16	14.6
	DF (100%)	100	67	100	33	100	0	33	0	
Melon (Egusi) (n = 3)	Mean	0.21	0.22	0.37	2.64	4.71	<0.48	9.84	<0.18	18.7
All UK	Median	0.22	0.19	0.45	2.71	4.47	<0.48	8.71	<0.18	17.4
	Range	0.15–0.26	0.16–0.29	0.20–0.47	2.31–2.90	4.11–5.54	<0.48	7.33–13.5	<0.18	14.9–23.6
	97.5th Percentile	0.26	0.29	0.47	2.89	5.49	<0.48	13.2	<0.18	23.3
	DF (100%)	100	100	100	100	100	0	100	0	

* Sum of two isomers (TMTP1 and TMTP2).

(<0.32; <2%) (Table 1; Fig. 1). The sum of the median concentrations of the three aryl-OPEs (∑aryl-OPEs, TPHP, EHDPP, and TMTP) and the two alkyl-OPEs (∑alkyl-OPEs, TBOEP and TnBP) were respectively 35.3 ng/g ww (40% Σ_8OPEs) and 41.9 ng/g ww (47%), which exceeded significantly those of the three Cl-OPEs (∑Cl-OPEs, TCEP, TCIPP and TDCIPP; median: 12.2 ng/g ww, 14%) ($p < 0.05$) (Table 1; Fig. 1). The $\sum_8$OPE concentrations in most foods in this study were within the range reported previously in other studies around the world (Chen et al., 2021; Gbadamosi et al., 2022; He et al., 2018; Poma et al., 2017; 2018; Wang & Kannan, 2018; Zhao et al., 2019). However, for milk, our concentrations (median: 9.62; 5.68–18.0 ng/g ww) were approximately 100 times lower than those reported for dairy products in China (median: 952,

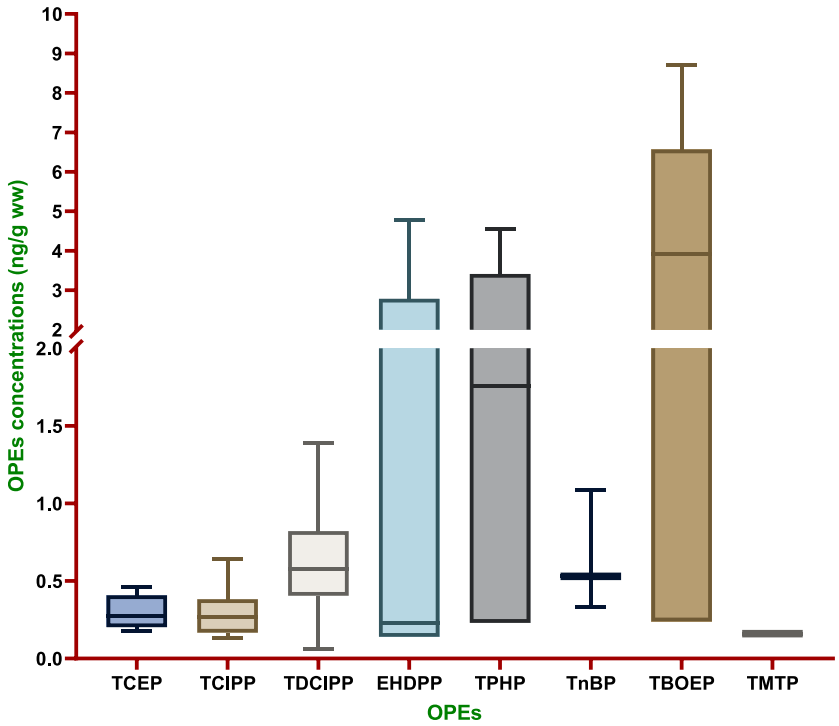

Fig. 1. Box and whisker-plot showing lower and upper concentrations of OPEs in foodstuffs of Nigerian origin (ng/g ww). [The central box represents the 25th to 75th percentile. The middle bold line is the median, while the top and bottom lines of the y-error bars represent the maximum and minimum].

range: 33.8–3065 ng/g ww) (Zhang et al., 2016), but exceeded those reported in studies from: Sweden, Belgium, the USA, China, Australia, and the UK (Gbadamosi et al., 2022; He et al., 2018; Poma et al., 2017, 2018; Wang et al., 2022; Wang & Kannan, 2018; Zhao et al., 2019). This variation in the distribution of OPE concentration data obtained in our study along with those from previous studies from different countries is likely attributed to international variations in the regulation, production, and use of OPEs.

TPHP was the main OPE detected in dried and smoked fish (median: 2.02 ng/g, 52%), milk (median: 4.54 ng/g ww, 47%), cereals (median: 3.06 ng/g ww, 40%), cassava flakes (Garri) (median: 2.50 ng/g ww, 55%), and melon (median: 4.47 ng/g, ww, 26%) respectively (Fig. 2). EHDPP was the main compound detected in wheat flour (Semo) (median: 4.77 ng/g ww, 58%) followed by cereal (median: 3.01 ng/g, ww; 39%). This is consistent with EHDPP being one of the main OPEs used in food packaging materials. Similar to observations in previous studies in the USA (Wang & Kannan, 2018), China (Ding et al., 2018; Zhang et al., 2016), and the UK (Gbadamosi et al., 2022), TBOEP was the major OPE detected in our study, contributing about 41% to the median concentration of $\sum_8$OPEs in the foodstuffs analysed. This may be because of the high bioaccumulation factor (BAF) of TBOEP (25.6) (Wang & Kannan, 2018) or its wide application in various consumer and industrial products leading to its ubiquitous presence in the environment. In contrast, the Cl-OPEs were the lowest contributors to Σ_8OPE concentrations, apart from TMTP that was not detected in any sample. The most abundant Cl-OPE in this study was TDCIPP, which was detected in: cassava flakes (median: 0.67 ng/g, ww: 15% Σ_8OPEs) and wheat flour (median: 1.39 ng/g, ww; 17%); followed by TCEP in dried and smoked fish (median: 0.40 ng/g, ww; 10%). (Table 1). For TnBP, the greatest contribution of 14% Σ_8OPEs occurred in dried and smoked fish (median: 0.53 ng/g, ww).

To evaluate whether transport of foodstuffs (and associated factors such as additional time spent in contact with food packaging) lead to markedly increased concentrations of OPEs; we compared

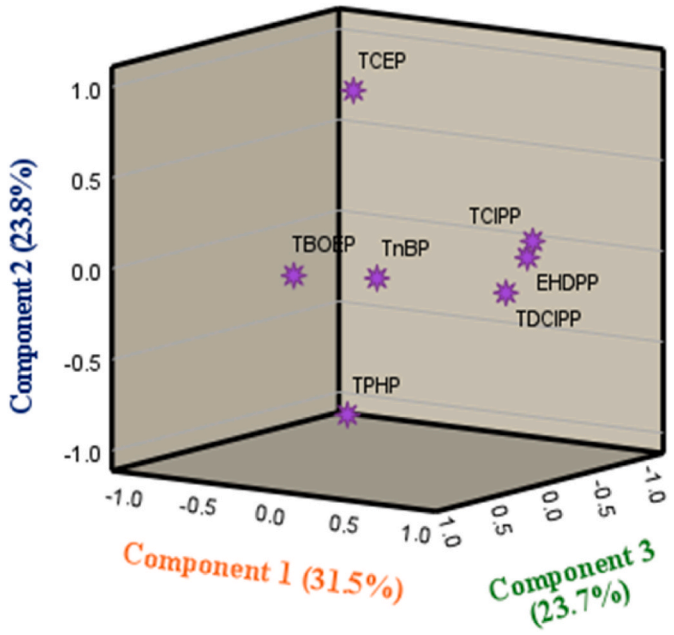

Fig. 2. Graphical representation of components (PC-1 = 31.5%, PC-2 = 23.8%, and PC-3 = 23.7%).

concentrations of OPEs in dried and smoked fish, roasted meat, and cereals collected in African stores in the UK, with those purchased in Nigeria. Our data showed higher mean concentrations of $\sum_8$OPEs in fish purchased in the UK (n = 17; 10.3 ng/g ww) than in those bought in Nigeria (n = 9; 5.11 ng/g ww) (Kruskal-Wallis H test, $p > 0.05$) (Table S9). For the three Cl-OPEs (TCEP, TCIPP and TDCIPP) there is no significant differences in concentrations between samples of fish bought

in the UK and Nigeria (Kruskal-Wallis H test, $p > 0.05$). In contrast, concentrations of the aryl-OPEs: TPHP and EHDPP, in fish purchased in the UK (3.25 and 2.33 ng/g ww) exceeded significantly those in fish bought in Nigeria (1.20 and 0.14 ng/g ww) ($p < 0.05$) (Mann-Whitney U tests) (Table S9). However, for roasted meat and cereals, there is no significant difference ($p > 0.05$) between concentrations of TPHP and EHDPP in samples bought in the African stores in UK and those purchased in Nigeria. In the same vein, a higher concentration of Cl-OPEs (but not significantly so, Kruskal-Wallis H test, $p > 0.05$) are seen in UK-purchased samples of meats and cereals (Table S9). A slightly different trend was obtained for the alkyl-OPEs. While higher but not significantly higher concentrations of TBOEP (Kruskal-Wallis H test, $p > 0.05$) were found in roasted meat and dried and smoked fish collected in the UK (8.54 and 2.75 ng/g ww) compared to those collected in Nigeria (3.09 and 1.89 ng/g ww); for the cereal group the mean TBOEP concentrations in the UK (0.24 ng/g) was below that from Nigeria (0.87 ng/g ww) (Table S9). Additionally, TnBP was not detected in any of the samples purchased in Nigeria while the median concentrations were 0.18 and 0.55 ng/g ww in cereal and roasted meat purchased in the UK. Our results suggest that some foods of Nigerian origin sold in the UK are more contaminated with aryl-OPEs and TBOEP than in equivalent foodstuffs sold in Nigeria. While our comparison is based on relatively few samples, this raises a potential concern for exposure of individuals consuming such imported foods, and more detailed investigation of this issue is recommended strongly.

Concentrations of OPEs in foodstuffs bought from major African stores in the UK were also compared with those in equivalent "typical" UK foodstuffs bought from major grocery stores and reported in a previous study (Gbadamosi et al., 2022). The median concentrations of Σ_8OPEs obtained in dried and smoked fish (5.91 ng/g ww) and cereal (13.0 ng/g ww) collected from African stores in the UK were comparable to those reported for dried and smoked fish (5.58 ng/g ww) and cereal (13.8 ng/g ww) in our previous study of UK foodstuffs (Gbadamosi et al., 2022). In contrast, significantly higher (Kruskal-Wallis H test, $p > 0.05$) concentrations of Σ_8OPEs were detected in milk of UK origin (median 17.6 ng/g) (Gbadamosi et al., 2022) than in milk from Nigeria (median: 9.62 ng/g ww) bought in the UK; while the median concentration of Σ_8OPEs in roasted meat (11.5 ng/g ww) from Nigeria purchased in African stores in the UK exceeded that detected in meat (median 6.30 ng/g ww) from UK supermarkets (Gbadamosi et al., 2022).

3.2. Source identification and correlation between individual OPEs in foodstuffs of nigerian origin

The results of a Spearman's rank correlation analysis were used to establish the relationship between the concentrations of individual OPEs in the foodstuffs (Table 2) after removing the outliers using the interquartile range method (IQR) and all TMTP data as this OPE was not detected in any sample. TCEP shows a significant strong negative correlation with TPHP ($r = -0.767$; $p < 0.01$), and a weak negative correlation with TDCIPP ($r = -0.164$; $p > 0.05$). A weak non-significant correlation also existed between TCIPP and TDCIPP ($r = 0.207$; $p > 0.05$), moderate between TCIPP and EHDPP ($r = 0.586$; $p > 0.05$), weak between TCIPP and TnBP ($r = 0.312$; $p > 0.05$) and weak negative correlations with TPHP ($r = -0.172$; $p > 0.05$) and TBOEP ($r = -0.191$; $p > 0.05$) (Table 2). In the same vein, weak non-significant correlation existed between TDCIPP with TBOEP and TnBP ($r = 0.119$–0.467; $p > 0.05$). EHDPP also correlated weakly but non-significantly with TnBP and TBOEP ($r = -0.053$ to -0.249; $p > 0.05$) (Table 2). Our results showed that there is no correlation between the OPEs in the foodstuffs except TCEP and TPHP which shows a strong negative correlation which implies that the sources of these OPEs congeners in the Nigeria foodstuffs are not related. This contrasts with what was obtained in some previous studies, where a significant positive correlation between a number of OPEs were found in foodstuffs from the USA (Wang & Kannan, 2018), China (Chen, Fan, et al., 2021), the UK (Gbadamosi et al., 2022) and in other matrices such as house dust (Huang et al., 2020) and human milk (Chen, Zhao, & Shi, 2021).

Principal component analysis (PCA) was also applied to further establish the possible contamination sources of the OPEs in the foodstuffs. Using PCA, the whole dataset was dimensionally reduced to three components which explained 79.0% of the total variation, similar to what was obtained in our previous study in UK foodstuffs where the three extracted components explained 78.9% of the total variation (Gbadamosi et al., 2022). The first component (PC-1) explained 31.5% of the variance and was loaded heavily in a positive direction by TCIPP (0.891) and TDCIPP (0.880) (Table 3; Fig. 2). The second component (PC-2) explained 23.8% of the total variation and was loaded

Table 3
Principal component scores (after varimax rotation) obtained from analysis of OPE concentrations in foods of Nigerian origin.

OPE	Component		
	1	2	3
TCIPP	**0.891**	0.172	−0.150
TDCIPP	**0.880**	−0.078	0.097
TCEP	−0.185	**0.925**	0.067
TPHP	−0.274	**−0.879**	0.000
TnBP	0.495	0.067	**0.808**
TBOEP	−0.110	0.013	**0.759**
EHDPP	0.521	−0.024	**−0.625**
Eigen-value	2.260	1.760	1.512
% of variance explained	31.516	23.824	23.670
Cumulative (%)	31.516	55.339	79.009

Table 2
Spearman's correlation between the concentrations of OPEs in foodstuffs of Nigerian origin.

Correlations								
OPEs	Spearman's	TCEP	TCIPP	TDCIPP	EHDPP	TPHP	TnBP	TBOEP
TCEP	Correlation Coefficient	1	0.182	−0.164	−0.071	−0.767[a]	0.234	0.119
	Sig. (2-tailed)		0.614	0.651	0.845	0.010	0.516	0.744
TCIPP	Correlation Coefficient		1	0.207	0.586	−0.172	0.312	−0.191
	Sig. (2-tailed)			0.567	0.154	0.634	0.380	0.597
TDCIPP	Correlation Coefficient			1	0.071	−0.092	0.467	0.119
	Sig. (2-tailed)				0.845	0.800	0.173	0.744
EHDPP	Correlation Coefficient				1	−0.098	−0.249	−0.053
	Sig. (2-tailed)					0.787	0.488	0.884
TPHP	Correlation Coefficient					1	−0.236	−0.051
	Sig. (2-tailed)						0.511	0.890
TnBP	Correlation Coefficient						1	0.522
	Sig. (2-tailed)							0.122
TBOEP	Correlation Coefficient							1
	Sig. (2-tailed)							

[a] Correlation is significant at 0.01 level (2-tailed).

significantly by two OPEs: TCEP (0.93) in a positive direction and TPHP in a negative direction (−0.88), while the third component explained 23.7% of the total variation and was significantly positively loaded by two OPEs: TnBP (0.81) and TBOEP (0.76) and significantly negatively loaded by EHDPP (−0.63) (Table 3; Fig. 2). The PCA results are in one respect consistent with the correlational analysis' negative correlation between TCEP and TPHP i.e., PC-2 ($p < 0.05$).

3.3. Evaluation of human dietary exposure to OPEs from consumption of foods of nigerian origin

Median and high-end EDIs for $\sum_8$OPEs based on consumption of the foods analysed in this study were respectively: 431 and 771 ng/kg bw/day for children and 405 and 798 ng/kg bw/day for adults (Table S10 a-b; Fig. 3). The median and high-end EDI for the aryl-OPEs: TPHP (children: 92.3 and 176 ng/kg bw/day; adults: 90.6 and 176 ng/kg bw/day) and EHDPP: (children: 58.2 and 159 ng/kg bw/day; adults: 52.2 and 162 ng/kg bw/day) exceed those obtained for the UK population (Gbadamosi et al., 2022). The same trend was observed for the Cl-OPEs. However, the median children EDI value for the Cl-OPEs: TCEP (12.6 ng/kg bw/day), TCIPP (13.5 ng/kg bw/day) and TDCIPP (33.8 ng/kg bw/day) in our study were comparable to those reported in US (Wang & Kannan, 2018). The exposure data obtained for OPEs in foodstuffs of Nigerian origin for children and adults fall mainly within the international range reported previously for foodstuffs from other countries/regions. Compared to previous observations whereby cereal and milk and milk products were the highest contributor to OPE exposure for adults in Belgium, China, and the UK (Ding et al., 2018; Gbadamosi et al., 2022; Poma et al., 2017); in our study, dough cassava flour (Fufu), a locally processed food was the main contributor to human dietary exposure to OPEs in this study accounting for between 23 and 33% of Σ_8OPE EDIs for adults and children respectively (Tables S10a–b; Fig. 3). However, for children, other major contributors to dietary exposure to Σ_8OPEs (median and high-end EDIs) were: yam powder (poundo yam) (72 and 75 ng/kg bw/day), melon (71.6 and 96 ng/kg bw/day), and cereals (52.9 and 125 ng/kg bw/day). For adults, the main contributors in addition to cassava flour were: cassava flakes (Garri) (52.4 and 167 ng/kg bw/day) > milk (41.2 and 74.9 ng/kg bw/day) > wheat flour

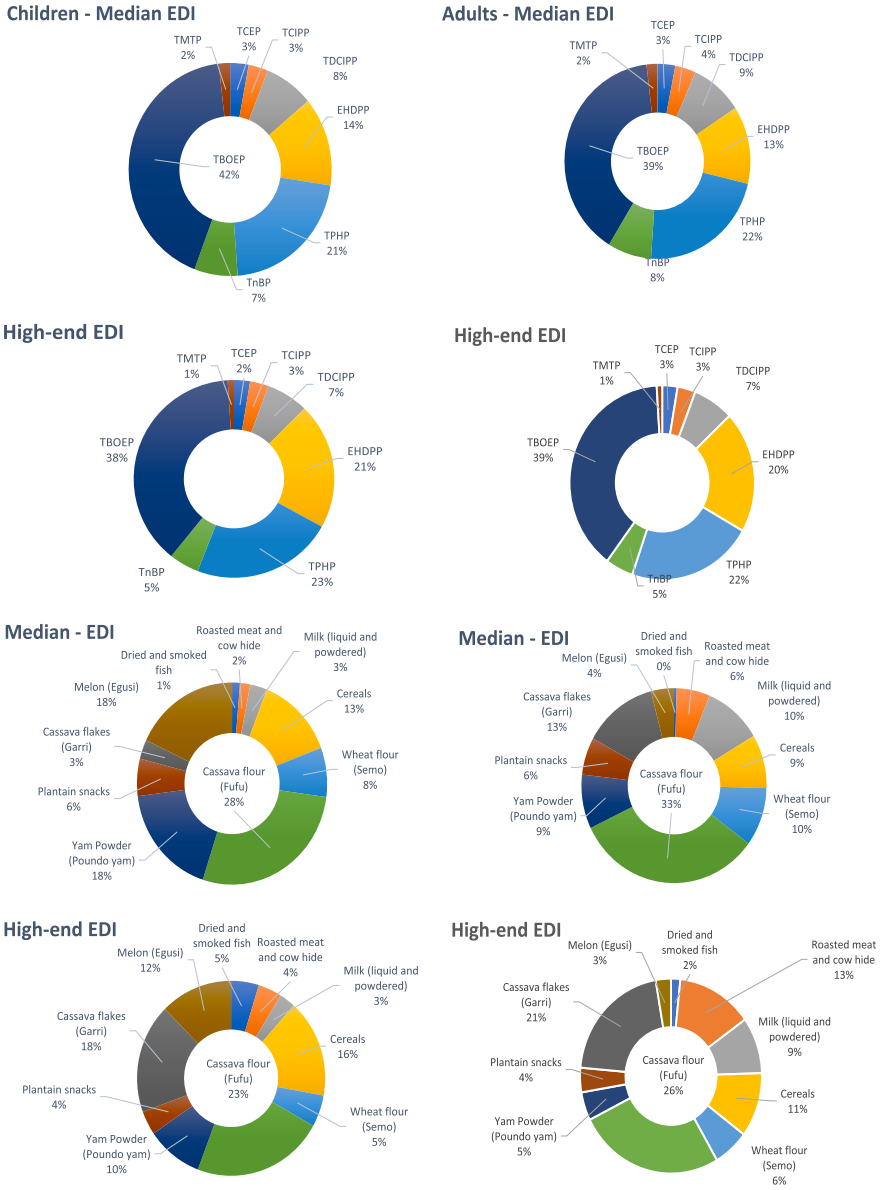

Fig. 3. Median and high-end exposure percent contributions of individual OPEs and different foodstuffs to Σ_8OPE EDIs for children and adults.

(Semo) (39.3 and 50.6 ng/kg bw/day) > yam powder (poundo yam) (36.3 and 37.6 ng/kg bw/day) > cereals (36.1 and 85.5 ng/kg bw/day) > plantain snacks (25.3 and 32.4 ng/kg bw/day) > roasted meat and cow hide (23.6 and 108 ng/kg bw/day) > melon (16.2 and 21.7 ng/kg bw/day) > dried and smoked fish (1.92 and 13.2 ng/kg bw/day) (Tables S10a–b, Fig. 3).

With respect to individual OPEs, the main contributor to Σ_8OPE concentrations was TBOEP, which accounted for 39% Σ_8OPEs for adults and 42% for children, followed by TPHP (22% adults – 21% children), EHDPP (13–21%), TDCIPP (7–9%), TnBP (5–8%), TCIPP (3–4%), TCEP (3%), and TMTP (1–2%) (Tables S10a–b; Fig. 3). This is similar to the situation for UK foodstuffs where TBOEP was the major contributor to Σ_8OPE exposure for all four of the age groups considered (Gbadamosi et al., 2022) and also consistent with assessment of the dietary exposure of the US population where TBOEP was the principal contributor to ΣOPEs exposure at ~30% for each age group studied (Wang & Kannan, 2018). In line with our previous study of UK dietary exposure (Gbadamosi et al., 2022), much of the contribution of TBOEP to overall dietary exposure originated from consumption of cassava flour, yam powder, melon, and plantain snacks rather than uniformly across all foodstuffs. Among the Cl-OPEs, TDCIPP makes the highest contribution to Σ_8OPE exposure (33.8–36.9 ng/kg bw/day, 8–9%) for adults and children respectively (Tables S10a–b, Fig. 3).

Our mean EDI obtained for five OPEs reported both in our study and previous studies of dietary exposure were compared. The mean EDIs for the sum of: TCEP, TCIPP, TDCIPP, TPHP, and TBOEP for children (335 ng/kg bw/day) were between 3 and 13 times higher than the value reported for children in UK foodstuffs (111 ng/kg bw/day) (Gbadamosi et al., 2022), in Eastern China (39.1 ng/kg bw/day) (Ding et al., 2018) and for children in New York (26.6 ng/kg bw/day) (Wang & Kannan, 2018) (Table S12). The high-end EDI for children (567 ng/kg bw/day) was comparable to the value reported for children in the UK (664 ng/kg/bw/day) under a high-exposure scenario (Gbadamosi et al., 2022) (Table S12). Moreover, the mean and high-exposure EDIs for the five OPEs for adults (314 and 590 ng/kg bw/day) in this study were below those reported for male (378 and 618 ng/kg bw/day) and female (427 and 664 ng/kg bw/day) adults in China (Zhang et al., 2016), in USA (mean EDI = 16.6 ng/kg bw/day) (Wang & Kannan, 2018), in UK (42.5 and 213 ng/kg bw/day) (Gbadamosi et al., 2022), in Belgium (mean: 77.5 ng/kg bw/day) (Poma et al., 2018), and in China (9.10–34.4 and 21.3–309 ng/kg bw/day) (Sala et al., 2022; Wang et al., 2022; Zhao et al., 2019) (Table S12).

3.4. Dietary risk assessment to OPEs in the selected foodstuffs from Nigeria

EDIs for adults and children in this study exceed those obtained based on consumption of non-Nigerian foodstuffs sold in the UK (Gbadamosi et al., 2022) and are more comparable to those obtained in China (Zhang et al., 2016). Table S11 compares the EDIs obtained in this study with reference dose (RfD) values from five different sources for our target OPEs. In all instances, EDI values were several orders of magnitude below RfD values (Table S11).

4. Conclusion

The present study reports the levels and profiles of eight OPEs in 85 foodstuffs of Nigerian origin, of which some were purchased in Nigeria and others bought from major African stores in Birmingham, UK. TBOEP, TPHP, and EHDPP were the predominant OPEs found in the foodstuffs analysed, with highest Σ_8OPE concentrations found in melon (Egusi), cassava flour (Fufu), and milk (liquid and powder). The sum of the total concentrations of the three aryl-OPEs: TPHP, EHDPP, and TMTP significantly exceeds those of the three Cl-OPEs: (TCEP, TCIPP, and TDCIPP). Locally processed foods such as cassava flour, plantain snacks, melon, yam powder, roasted meat, and cow hide were more contaminated with TBOEP and TPHP than other foods studied. Our results revealed significantly higher concentrations of aryl-OPEs in dried and smoked fish bought from African stores in the UK than in similar samples purchased in Nigeria ($p < 0.05$). Cassava flour dough (Fufu) was the major foodstuff contributing to dietary intake of OPEs, comprising 26% Σ_8OPEs for children and 33% for adults. EDI values for OPEs were several orders of magnitude below the current reference dose values, suggesting low risks from these chemicals via consumption of foods of Nigerian origin. Nonetheless, EDIs obtained in this study exceed those for the UK population generally, and more detailed measurement of dietary exposure of Africans is recommended.

CRediT authorship contribution statement

Muideen Remilekun Gbadamosi: Investigation, Software, Validation, Funding acquisition, Visualization, Writing – original draft, Writing – review & editing. **Kehinde Hakeem Moberuagba:** Writing – review & editing, Software. **Mohamed Abou-Elwafa Abdallah:** Methodology, Conceptualization, Supervision, Resources, Validation, Writing – review & editing. **Stuart Harrad:** Conceptualization, Supervision, Resources, Validation, Writing – review & editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Muideen Remilekun Gbadamosi reports financial support was provided by Petroleum Technology Development Fund.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodcont.2023.109880.

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