



Review article

Exposure to phthalates from personal care products: Urinary levels and predictors of exposure

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ABSTRACT

Phthalates are a large group of chemicals used in many everyday consumer products such as food packaging, household cleaners, cosmetics, fragrances and personal care products (PCPs). A number of diseases such as obesity, hypertension, as well as reproductive system effects and endocrine disorders have been linked to phthalate exposure through the use of PCPs, due to their frequent use and high phthalate content. In this study we review available literature on phthalates and their metabolites in urine and report the various determinants of exposure through the use of PCPs in infants, toddlers, children and adults. The range of creatinine-adjusted concentrations for each phthalate was 1.5–14956.1 µg/g for MEP, 0.4–94.5 µg/g for MEHP, 0.39–425.9 µg/g for MEHHP, 0.5–481.3 µg/g for MEOHP, 0.1–755.1 µg/g for MBzP, and 0.3–401.4 µg/g for MiBP. Time of sampling, frequency of use, race and age are critical factors that influence phthalate concentrations. Using PCPs 48 h before urine collection, using a combination of PCPs (i.e., particularly leave-on versus rinse-off products), being younger (i.e., children compared to their mothers), and being a woman of colour (i.e., Mexican-American and black versus white) leads to higher phthalate levels in urine. The most striking association between any phthalate and PCPs was observed between MEP and perfumes or fragrance-containing products such as shampoos, body lotions and hair products. Future studies should focus on different types and brands of PCPs (i.e., branded versus generic), explore possible ethnic/racial differences and the applicability of non-invasive matrices such as nails and hair for phthalate biomonitoring, as well as intervention studies that explore behavioural changes.

1. Introduction

Phthalates are a group of chemicals that are used as plasticizers, stabilizers and preservatives. They can be found in a wide variety of products such as personal care products (PCPs) (e.g., cologne/perfume, body lotion, shampoo), building materials (e.g., flooring, wall materials), food packaging and children's toys (Berman et al., 2009). They have short half-lives and are easily metabolized in the human body to their monoesters, while some phthalates can endure further metabolism (Guo and Kannan, 2013). Most human biomonitoring studies sample urine to assess phthalate exposure in the human body, because the majority of their metabolites are discharged through urine in less than 48 h (Koch et al., 2017). Due to their presence in many consumer products and their non-covalent bonds, they can be released into the environment resulting in increasing human exposure (Wong and Durani, 2017). Phthalates are divided in two groups: low-molecular weight

(LMW) and high-molecular weight (HMW) phthalates. Each group includes different types of phthalates, depending on their nature or their use; an example of a LMW phthalate is diethyl phthalate (DEP) which is used as a dissolvent in cosmetics (i.e., perfumes), while a HMW phthalate is di (2-ethylhexyl) phthalate (DEHP) used as plasticizer-stabilizer in building materials (Cavallari et al., 2015).

In PCPs, phthalates provide stabilization and an extended duration of the product. They are mostly found in creams and lotions (e.g., after-shave, shaving cream, bar/liquid soap, facial/body creams), cosmetics, deodorants, mouthwash, nail polish, hair products, baby products such as diapers, talc, and many feminine hygiene products (e.g. pads, tampons, and vaginal douche). Due to their frequent use and high phthalate content, PCPs are both an environmental concern and a human health risk (Ferguson et al., 2017). A variety of diseases such as obesity, hypertension, reproductive system effects and endocrine disorders have been linked to phthalate exposure through the use of PCPs (Ferguson et al., 2017; Dodson et al., 2012; Kabir et al., 2015; Katsikantami et al.,

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Abbreviations

BBP	benzyl butyl phthalate
BzBP	benzyl butyl phthalate
DBP	dibutyl phthalate
DCHP	dicyclohexyl phthalate
DEHP	di(2-ethylhexyl) phthalate
DEP	diethyl phthalate
diBP	di-isobutyl phthalate
diPP	di-isopentyl phthalate
DMEP	dimethoxyethyl phthalate
DMP	dimethyl phthalate
DPP	dipentyl phthalate
EPA or USEPA	United States Environmental Protection Agency

HMW	high-molecular weight
LMW	low-molecular weight
MBzP	monobenzyl phthalate
MECPP	mono(2-ethyl-5-carboxypentyl) phthalate
MEHHP	mono-(2-ethyl-5-hydroxyhexyl) phthalate
MEHP	mono-(2-ethylhexyl) phthalate
MEOHP	mono-(2-ethyl-5-oxohexyl) phthalate
MEP	mono-ethyl phthalate
MiBP	mono-isobutyl phthalate
MiBP	mono-isobutyl phthalate
MMP	mono-methyl phthalate
MnBP	mono-n-butyl phthalate
PCPs	personal care products

2016; Amato et al., 2021; Deng et al., 2021). Subsequently, some studies have highlighted the importance of banning products containing phthalates or progressively reducing the use of consumer products containing phthalates in order to decrease exposure in the most vulnerable groups, such as teenagers, babies and infants (Wong and Durrani, 2017).

Dermal exposure, ingestion and inhalation are the most important routes of phthalate exposure (Koppen et al., 2019). PCPs applied to the skin are easily absorbed, thus increasing the body burden of phthalates. Children and infants are particularly vulnerable, because of their thinner skin compared to adults and inappropriate use of PCPs related to their young age (Wong and Durrani, 2017). Infants and toddlers are also exposed through their hand-to-mouth behavior, creating an oral route from many different sources (e.g. plastic toys, food packages, etc.) (Sathyanarayana et al., 2008).

The aim of the present study was to review available literature on phthalates and their metabolites in urine and report the various determinants of exposure through the use of PCPs in infants, toddlers, children and adults.

2. Methodology

2.1. Inclusion and exclusion criteria

The study's inclusion criteria were the following: 1) the studies must be in English, 2) they must refer to direct human exposure to phthalates through PCPs use only (studies referring to exposure via indoor dust, building materials, packaged food, and children's toys were not included), 3) studies must report an association of phthalates with PCPs (studies with no association were not considered), 4) studies must contain questionnaires that measured the frequency of PCPs use. We included all age groups (adults, children, and infants). PCPs use was assessed through questionnaires either qualitatively (yes/no) or quantitatively (0, 1–3, more than 3) in the last 24–48 h prior to the urine sampling. Products that were closely related were grouped into one category (as seen in Table 1). These categories include: bathing products (e.g., liquid soap, body wash, bath oil, bath gel, bubble bath), facial creams (e.g., facial masks, day and night creams), body lotions (e.g., hand lotion, body lotion, body oil), fragrances (e.g., colognes, perfumes), colored cosmetics (e.g., hair dye, foundation, blush, eye shadow, eye liner, mascara), and hair products other than hair spray or hair gel (e.g., mousse, gel, hair bleach, relaxer, perm, dye, hair nutrient products).

2.2. Search strategy description

We searched for relevant articles published in Web of Science, PubMed, and Scopus using the following combination of keywords:

“phthalates”, “human” “exposure”, “(personal care products)”, “(use OR questionnaire OR survey)”, “MEP”, “MEHP”, “MBzP”, “MEHHP”, “MMP”, “MCP”. The search was performed in two stages (until September 2019 and from October 2019 to April 2021) using the same keywords and was completed at the end of April 2021. The selection process of the papers that are included in this review is illustrated in Fig. 1.

3. Results and discussion

Our literature review search included 30 articles that investigated the association between phthalates and PCPs via questionnaires. Most of these studies were from North America (13 from the USA and Canada and 1 from Mexico), 11 studies were from Europe and 6 from Asia. The sampling was conducted between 1999 and 2017 with an average population size of 463 (ranging from 10 to 6470). In terms of the bio-monitoring matrix, the majority analyzed urine samples to assess phthalate exposure. The most common type of sample was spot urine (24 studies), followed by multiple urine samples (3 studies) and 24 h urine analysis (1 study). Our review included mother-child pairs (7 studies), pregnant women (5 studies), men (3 studies) and infants (1 study). The age range for the adult population was 18–70 years, 3–13 years for children and 2–28 months for infants. Most of the studies reported creatinine-adjusted phthalate concentrations (19 studies), and considerably fewer reported unadjusted (5 studies) and specific gravity-adjusted phthalate concentrations (4 studies). The results are summarized in Supplementary Table 1.

3.1. Factors affecting phthalate concentrations

Mono-ethyl phthalate (MEP) was the most frequently detected phthalate in humans (26 studies) (Table 1), followed by mono-(2-ethylhexyl) phthalate (MEHP), mono-(2-ethyl-5-hydroxyhexyl) phthalate (MEHHP), mono-(2-ethyl-5-oxohexyl) phthalate (MEOHP) and monobenzyl phthalate (MBzP) (23 studies for each phthalate), and mono-isobutyl phthalate (MiBP) (21 studies). The range of creatinine-adjusted concentrations for each phthalate was 1.5–14956.1 µg/g for MEP, 0.4–94.5 µg/g for MEHP, 0.1–755.1 µg/g for MBzP, 0.3–401.4 µg/g for MiBP, 0.39–425.9 µg/g for MEHHP, 0.5–481.3 µg/g for MEOHP. Time of sampling is a critical factor that influences phthalate concentrations. Cavallari et al. (2015) used multiple urine samples (i.e. first void, before and after shift, and before bedtime) to monitor phthalate concentrations in custodians. The results showed a higher phthalate concentration in the first (i.e. morning) void, which becomes even greater before the custodians' shift, and decreases after their shift and before bedtime. This variability is related to PCPs use before bedtime (such as creams and lotions) and leads to a higher concentration of phthalates in urine in the first void sample. In the study by Giovanoulis

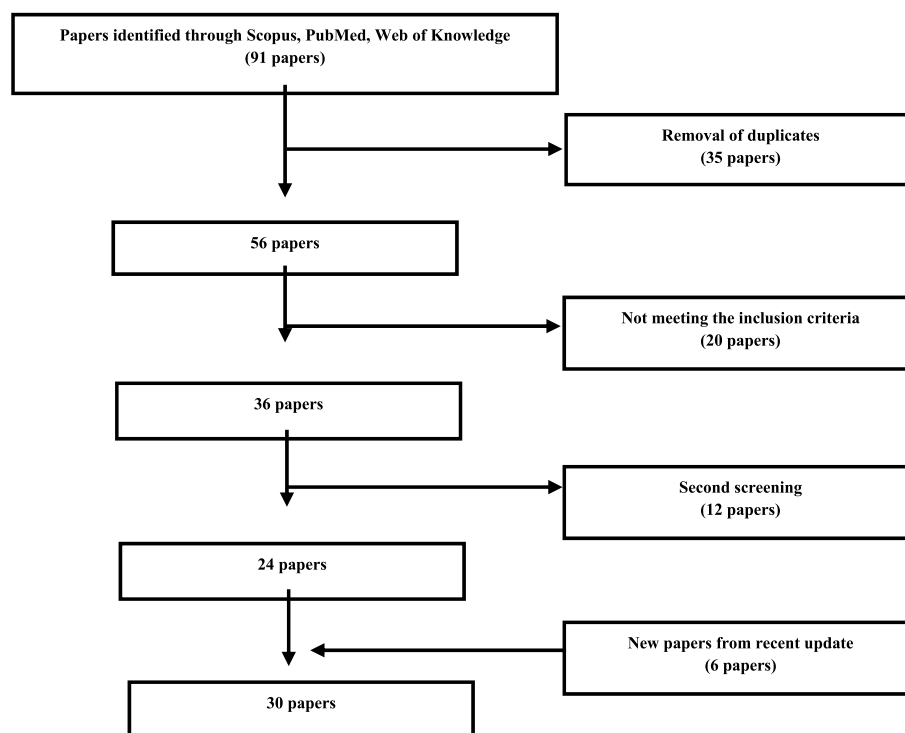


Fig. 1. A flow diagram of the research and selection process.

et al. (2016) three urine samples were taken over two consecutive days (i.e. in the afternoon of day 1, and the morning and afternoon of day 2). The results were similar to the study by Cavallari et al. (2015), with the greater concentration measured in the morning of day 2 due to personal care use before bedtime (i.e. shower gel, deodorant and body lotion).

Age is another important factor in determining phthalate concentrations. Our review included seven studies that report phthalates in mothers and their children. The majority of these studies (Cullen et al., 2017; Cutanda et al., 2015; Koppen et al., 2019; Larsson et al., 2014; Philippat et al., 2015; Sakhi et al., 2017; Runkel et al., 2020) reported higher phthalate concentrations in children than their mothers. Only one of these showed higher phthalate concentrations in mothers compared to their children - except for mono (2-ethyl-5-carboxypentyl) phthalate (MECPP) whose concentration was higher in children than their mothers (Lewis et al., 2013). The higher body burdens in children may be attributed to different PCP usage compared to their mothers, their smaller body size, their significant hand-to-mouth behavior and regular contact with phthalate-containing products (such as plastic toys) (Cullen et al., 2017). Koppen et al. (2019) observed a large variability in urine levels between mothers and their children for MEP (20 and 10-fold difference between P10 and P90).

Two out of the five studies on pregnant women reported phthalate concentrations for each trimester (Braun et al., 2014; Hsieh et al., 2019). The results showed an increase of phthalate exposure in the 2nd trimester (Berman et al., 2009; Hsieh et al., 2019) and 3rd trimester (Braun et al., 2014). In contrast, Hsieh et al. (2019) observed a decrease in the 3rd trimester. Finally, only two studies explored sex differences in our review. In the study by Müllerová et al. (2016), men had lower phthalate concentrations than women (men: 60.7 µg/g, women: 100.3 µg/g) which may be a result of higher personal care product use by women compared to men.

3.2. Predictors of phthalates through the use of personal care products

Table 1 presents the predictors of phthalates through PCPs use among the eligible studies in our review. It shows the associations

between phthalates measured in urine and PCPs use. We only included associations that were statistically significant (i.e. p -values of 0.05 or lower). For every statistically significant association we marked their positive (+) or negative (−) direction for each PCP. The PCPs list in Table 1 was compiled according to the products included in each study. It should be noted that not all studies have looked into all PCPs categories.

The most frequently reported phthalate across all groups of PCPs was MEP (in 21 products). The use of cologne/perfume and hand/body lotion/oil/cream was positively associated to MEP. Mono-n-butyl phthalate (MnBP) was also positively associated with reported use of many types of personal care products (11 in total), such as colored cosmetics, nail polish and shampoo (with the exception of shampoo, that showed both positive and negative associations). Reported use of cologne/perfume, colored cosmetics and nail polish was positively associated with mono-isobutyl phthalate (MiBP) and mono-methyl phthalate (MMP) (with one study reporting a negative association between colored cosmetics and MiBP). MEHHP and MECPP were least frequently associated with reported use of personal care products (e.g. hand/body lotion/oil/cream, cologne, perfume or fragrance and nail polish). Last but not least, MEOHP was associated with reported use of only 2 types of personal care products (e.g. cologne/perfume/fragrance and colored cosmetics).

Several studies highlight the importance of the time since the PCPs were last used. More specifically, Berman et al. (2009) observed that individuals who used personal care products 48 h before urine collection had higher phthalate concentrations in their urine. Duty et al. (2005) also measured higher MEP levels in participants who used cologne/perfume 3–6 h before urine collection, compared to those whose urine was collected at least 8 h after the use of personal care products. Similarly, Giovanoulis et al. (2016) observed that people who used personal care products on their hands during a 24-h period had higher MEP and MnBP levels, and those who used more than 5 personal care products had higher urinary MEP levels. The associations between the number of PCPs used and phthalate concentrations was also reported by Parlett et al. (2013); women who used at least 8 PCPs had higher MEP levels

Table 1Predictors of phthalates in different types of personal care products (+: positive and -: negative direction of effects, $p < 0.05$).

PCPs	n (%) of studies ^a	MEP	MEHP	MBzP	MiBP	MEHHP	MEOHP	MBP	MnBP	MCP	MECPP	MMP
Creams and lotions												
aftershave	3 (10)	+										
bar soap	8 (27)	++	+									
facial cream/masks (day/night cream)	12 (40)	++++						+		+		
hand or body disinfectant (sanitizer)	3 (10)											
hand/body lotion/oil/cream	22 (73)	+++++--	+-	+-	+	+		+-	+		+	
liquid soap/body wash/bath oil, bath gel, or bubble bath	10 (33)	+		+					-	+		+
shaving cream	6 (20)	+										
sunscreen (suntan, sunblock)	8 (27)	+++						+	+		+	+
wipes/towelettes	1 (3)											
Toiletries and cosmetics												
cologne, perfume or fragrance	20 (67)	+++++++			++++	+	+		+	+		
colored cosmetics (hair dye, foundation, blush, eye shadow, eye liner, or mascara)	16 (53)	+++++	+-	+	+-	+	+	++	++		+	+
conditioner/creme rinse	7 (23)	+										
deodorant (antiperspirant)	14 (47)	+++++			+				+			
fragrance-free products	1 (3)								+			
hairspray/hair gel	13 (43)	+++++		-	-				+-			
lip balm	2 (7)	+										
lipstick	6 (20)	+++										-
mouthwash	6 (20)	+										
nail polish, nail polish remover	13 (43)	+++++-	+	+	++			+	++			
other hair care products (mousse, gel, hair bleach, relaxer, perm, dye), hair nutrient products	14 (47)	+++			++			+	+	+		+
shampoo	16 (53)	++++--	+-		+		-		+-			
toothpaste	1 (3)	+										
Feminine hygiene												
feminine powder	1 (3)											
feminine spray	1 (3)											
feminine wash (vaginal douche)	2 (7)	+										
other feminine hygiene products	1 (3)											
sanitary towel	1 (3)											
tampon	1 (3)											
Infant/baby products												
diaper cream	2 (7)											
infant lotion	1 (3)	+										+
infant powder/talc/cornstarch	1 (3)				+							
infant shampoo	1 (3)											+
infant wipes	1 (3)											

Each + or - sign represents either a positive or negative association found in each study.

^a n: number of studies with data on the particular PCPs category.

than those who used 3 products. Another study that corroborates these observations is that by Romero-Franco et al. (2011), according to which women who used a combination of 4 different PCPs (e.g. perfume, deodorant, lipstick, nail polish and/or hand or face cream) had 4 times higher urinary MEP concentrations than women who used fewer than four of these PCPs. The positive associations and exposure risk from fragranced products, makeup and nail polish has also been observed in younger age groups such as preschoolers (Lim, 2020), which poses a health concern.

Another association between MEP and perfumes or fragrance-containing products such as shampoos, body lotions and hair products is evident (Table 1). DEP (the parent compound of MEP) is used as a solvent in fragranced products to make scent last longer (Philippat et al., 2015), so high MEP levels tend to be associated with fragranced personal care product use. According to Cavallari et al. (2015) and Branch et al. (2015) products with fragrances tend to contain higher concentrations of MEP than other products. Another overlooked dimension of this issue is the fact that scented product use differs between individuals of different ethnicities. Cavallari et al. (2015) observed significantly higher phthalate concentrations among Hispanic, non-white people. Branch et al. (2015) also found higher phthalate concentrations in Mexican-American and black women compared to white women. Zota and Shamasunder (2017) found that black women tend to use feminine hygiene products (e.g., vaginal douche, fragranced wipes) more frequently than white women; however they did not find a statistically significant difference in DEP concentrations between black and white women with respect to vaginal douching.

Black women purchase nine times more ethnic hair and beauty products such as hair relaxers and straighteners than any other group of personal care products. Whiteness tends to influence sales such as hair straighteners, skin lighteners, and odor masking products as racial discrimination can influence many peoples' preferences (Zota and Shamasunder, 2017). Phthalate concentrations vary among women all over the world, because of different use patterns, differences between phthalate concentrations in different personal care products worldwide (i.e., different brands, different quality) and each individual's metabolic system and genetic susceptibility (Romero-Franco et al., 2011).

The use of PCPs such as body lotion, skin toner, essential oil, lipstick and hairspray showed a positive association with MMP (Hsieh et al., 2019). A possible explanation is that these leave-on products may result in a higher phthalate exposure than rinse-off products such as body wash, shampoo, face wash and hand wash, which showed a positive association with MBzP levels. The authors also point out that washing behavior seemed to contribute to this negative association because of soap use. The use of soap and water may remove more phthalates than water alone during hand washing, which minimizes the amount of phthalates on the skin, and eventually reduces the amount of phthalates that is being absorbed. Differences in the variability of phthalate concentrations and personal care products may also be attributed to differences in the questionnaires (24 h and 48 h recalls), differences in urine collection (i.e., morning samples and end-of-the-day samples) and the variability in brands and products (Philippat et al., 2015).

The studies analysing phthalates in cosmetics and PCPs corroborate these observations. The most commonly detected phthalates are DEP (e.g., used as solvent and fixative in fragrances), dibutyl phthalate (DBP) (e.g., used as a plasticizer in nail polish), and dimethyl phthalate (DMP) (e.g., added to hair sprays to form a more flexible film). Phthalates are detected in 60–100% of tested products in most studies (Houlihan et al., 2002; Greenpeace, 2005; Shen et al., 2007; FDA or United States Environmental Protection Agency; Koniecki et al., 2011; Guo and Kannan, 2013; Llompарт et al., 2013). The United States Food and Drug Administration (FDA or United States Environmental Protection Agency,) reported DEP and DBP levels as high as 40,000 ppm in fragrances and 4800 ppm in nail polish, respectively. High concentrations of DEP were also reported by Koniecki et al. (2011) in Canadian products (fragrances: 25,542 mg/g) and Greenpeace (2005) with the highest concentration

being 22,299 mg/kg. DEP was also detected in a diaper cream product at 130 ppm (FDA or United States Environmental Protection Agency,). The occurrence of banned substances (Regulation (EC) No. 1223/2009), DBP and DEHP, at concentrations up to 141 µg/g, as well as those of di-isobutyl phthalate (DiBP), dimethoxyethyl phthalate (DMEP), benzyl butyl phthalate (BBP), di-isopentyl phthalate (DiPP), and dipentyl phthalate (DPP), were found in 16 of the 26 PCPs in a study by Llompарт et al. (2013), despite none of these phthalates being listed as ingredients in the products.

3.3. Limitations and recommendations

The studies included in this review have several limitations. For most (Berman et al., 2009; Lewis et al., 2013; Giovanoulis et al., 2016) the target group was small and as such the results cannot be representative of the whole population. Most studies focus on women and few include male participants, who are underrepresented even though they also use PCPs. A variety of personal care products should be examined in order to identify as many predictors as possible (Braun et al., 2014). In addition, high molecular weight phthalates such as DEHP, are further metabolized into oxidized products and are generally underestimated, and consequently, no statistically significant associations may be observed (Cavallari et al., 2015). Another major limitation is that most of the studies used spot urine sampled during the day to measure phthalate concentrations, and assessed exposure through PCPs use in the past 12 h. As such, products that were not used on a daily basis may not be detected in high concentrations (Cullen et al., 2017). Some questionnaires used categories such as “always”, “sometimes”, “never” for personal care products use, which does not give an accurate estimation of the number or frequency of PCPs use (Ferguson et al., 2017). Finally, we only reported the association of phthalate metabolite concentrations with personal care product use; other sources of phthalate exposure (e.g. diet, building products) were not examined, so exposure routes cannot be fully evaluated from the reviewed literature. In addition, we did not examine the effects of multiple exposures to other chemicals (e.g., parabens in the same personal care products that contain phthalates). Previous studies have reported that biomarkers of chemicals with similar structures and/or uses were more strongly correlated than unrelated chemicals (Kalloo et al., 2018; Lee et al., 2017).

Future studies should focus on different types and different brands of PCPs (e.g., branded versus unbranded/generic), questionnaires with specific questions on PCPs use, frequency, and variety, as well as other routes of exposure (e.g., canned food consumption, environmental parameters such as house construction materials), and should follow a more consistent sampling plan of urine collection, with more than one sample during the day. Another suggestion for future research is to explore possible ethnic/racial differences and the variety of social and economic factors that results in different patterns of PCPs use (Zota and Shamasunder, 2017; Preston et al., 2021; Chan et al., 2021). Moreover, the applicability of non-invasive matrices such as nails and hair for phthalate biomonitoring should be further explored (Giovanoulis et al., 2016; Li et al., 2021). Another factor that should be taken into account is the different dietary patterns among people, which should be considered in order to identify possible phthalate exposure apart from personal care product use (Hsieh et al., 2019; Park et al., 2019). Intervention studies like HERMOSA by Harley et al. (2016) are another valuable tool in estimating the reduction of human body burden to these chemicals by behavioural changes and should be further explored. Such behavioural changes in addition to education and outreach to the general public are two key elements for the reduction in phthalate exposure and public health protection. Finally, since humans are exposed to multiple chemicals at the same time via different exposure routes, future research would benefit from examining the effect of chemical mixtures on human health for more informed risk assessment.

Author contribution

Conceptualization, O.-I. K.; Writing - original draft, O.-I. K. and A.P.; Writing - review & editing, O.-I. K. and O.S.A.; Investigation, O.S.A. and A.P.; Methodology, O.S.A and A.P.; Supervision, O.-I. K. All authors have read and agreed to the published version of the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envres.2022.113194>.

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